`qlanth`

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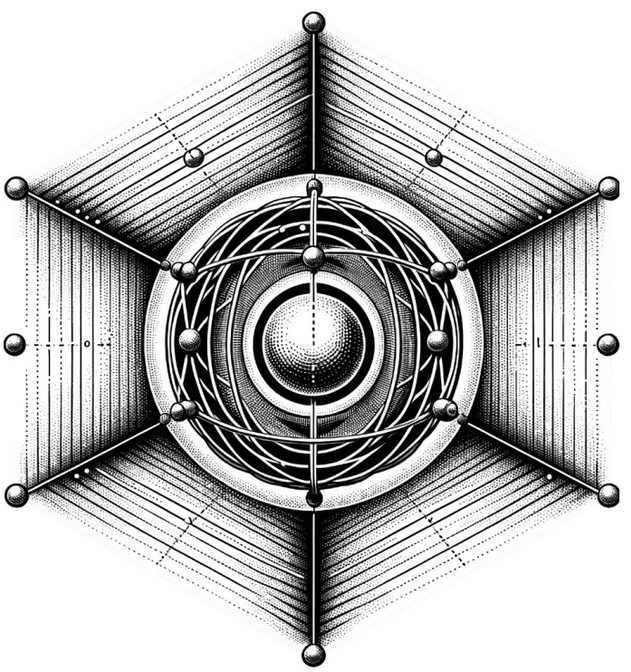

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2025 AD

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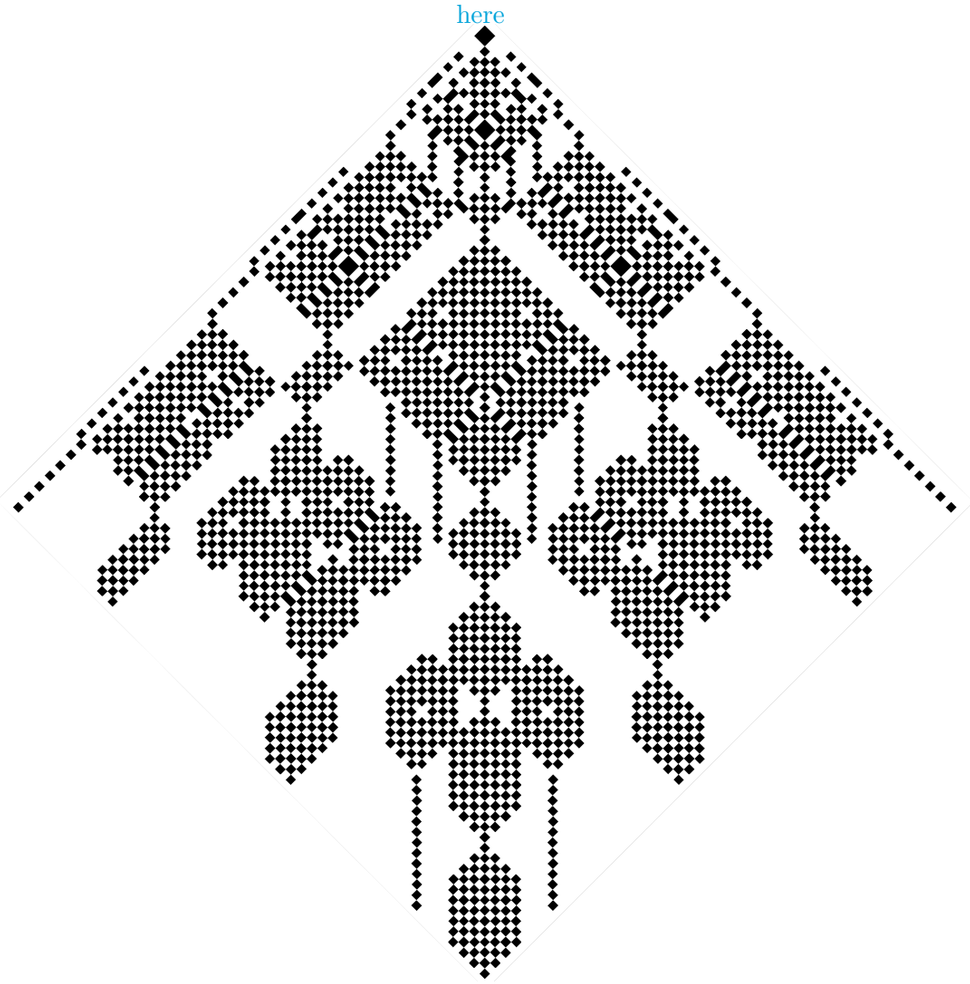

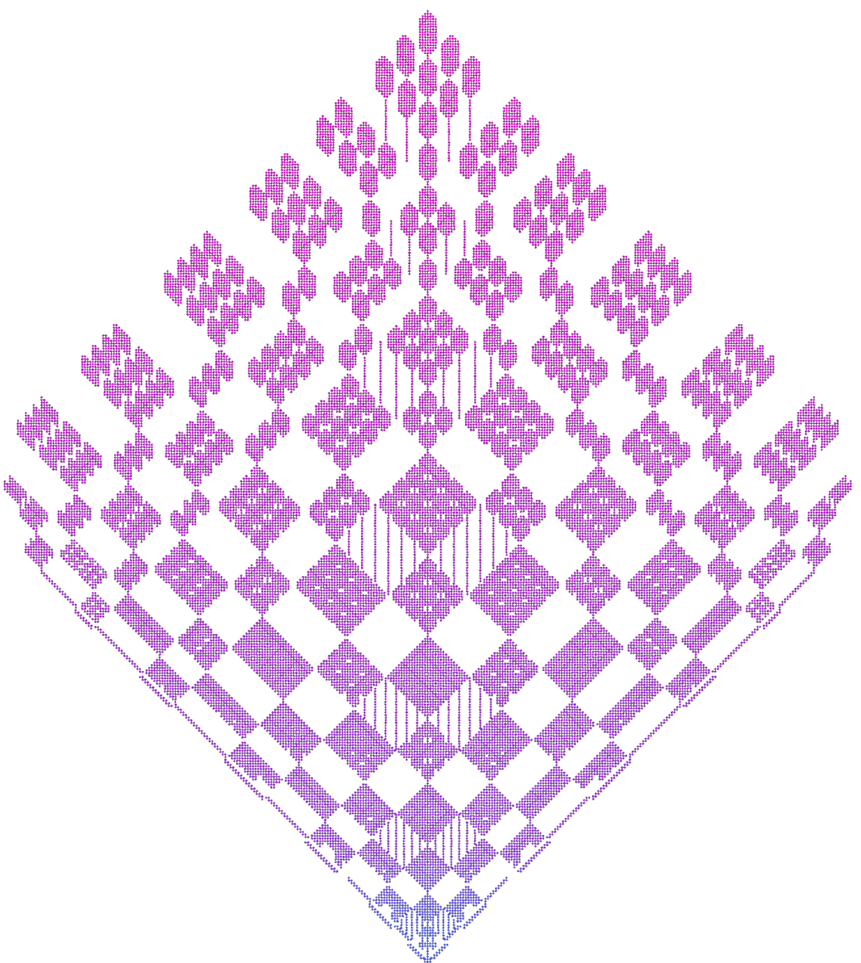

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qlanth is a tool that can be used to estimate the electronic structure of lanthanide ions in crystals. I uses an effective Hamiltonian limited to a single-configuration with included configuration-interaction corrections. This Hamiltonian aims to describe the observed properties of ions embedded in solids in a picture that imagines them as free-ions modified by the influence of the lattice in which they find themselves in.

This picture of lanthanide ions is one that developed and mostly matured in the second half of the last century by the efforts of John Slater,[1] Giulio Racah,[2] Brian Judd,[3] Gerhard Dieke,[4] Hannah Crosswhite,[5] Robert Cowan,[6] Michael Reid,[7] William Carnall,[8] Clyde Morrison,[9] Richard Leavitt,[10] Brian Wybourne,[11] Richard Trees,[12] and Katherine Rajnak[13] among others. The goal of this tool is to provide a modern implementation of the methods that resulted from their work. This code is written in Wolfram language.

Separate to their specific use in this code, **qlanth** also includes data that might be of use to those interested in the single-configuration description of lanthanide ions. These data include the coefficients of fractional parentage (as calculated by Velkov and parsed here), and reduced matrix elements for all the operators in the effective Hamiltonian. These are provided as standard *Mathematica* associations that should be simple to use elsewhere. One feature of **qlanth** is that symbolic expressions are maintained up to the very last moment where numerical approximations are inevitable. As such, the symbolic expressions that result for the matrix representation of the Hamiltonian, result in linear combinations of the model parameters with symbolic coefficients.

The included *Mathematica* notebook `qlanth.nb` lists most of the functions included in **qlanth** and should be considered complementary to this document. The `/examples` folder includes notebooks containing the result of this description for most of the trivalent lanthanide ions in lanthanum fluoride. LaF_3 is remarkable in that it was one of the systems in which a systematic study [Car+89] of all of the trivalent lanthanide ions were studied.

This code was originally authored by Christopher Dodson and Rashid Zia for their research into magnetic dipole transitions in lanthanide ions [DZ12]. Here it has been rewritten and expanded by David Lizarazo. It has also benefited from conversations with Tharnier Puel at the University of Iowa.

This document has 17 sections. Section 1 gives an overview the semi-empirical Hamiltonian. Section 2 explains the details of the basis in which the semi-empirical Hamiltonian is evaluated, together with the method of fractional parentage, additional quantum numbers, Kramer's degeneracy, and the JJ' block structure of the semi-empirical Hamiltonian. Section 3 gives a detailed explanation of each of the interactions include in the semi-empirical Hamiltonian. Section 4 gives explains the implicit assumptions in the orientation of the coordinate system. Section 5 gives an overview of the attendant experimental setups and considerations about uncertainty. Section 6 is about the calculation of magnetic and forced electric dipole transitions. Section 7 explain certain constraints often used for the parameters in the semi-empirical Hamiltonian.

Section 8 explains the details of fitting the Hamiltonian to experimental data.Section 9 lists included auxiliary *Mathematica* notebooks. Section 11 explains the details of an abbreviated Python extension to **qlanth**. Section 12 explains some of the included experimental data. Section 13 contains a few assorted details on running **qlanth**. Section 14 has a brief comment on units. Section 15 and Section 16 include a summary of notation and definitions used throughout this document. Finally, Section 17 contains a printout of the code included in **qlanth**.

Besides being a fully functional code that works out of the box, **qlanth** is unique in that it also includes computational routines that can generate from scratch (or close to scratch) the necessary reduced matrix elements which in other codes are simply loaded from other vintages. Great care was taken to comment every loop, variable, procedure, and data provenance. To highlight this, the code relevant to the different functions has been interspersed in the parts where they are mentioned.

[1] [Sla29] [2] [Rac42a; Rac42b; Rac43; Rac49] [3] [Jud62; Jud63b; Jud63a; Jud66; Jud67; JCC68; CCJ68; Jud82; Jud83; JS84; JC84; Jud85; Jud86; Jud88; Jud89; JL93; Jud96; Jud05] [4] [DC63; PDC67; Die68] [5] [CCJ68; Cro71; Cro+76; Cro+77; DC63; JCC68; JC84] [6] [Cow81] [7] [Rei81] [8] [CFW65; Car+89; Car92; CFR68b; CFR68e; CFR68c; CFR68d; CFR68a; Car+70; Car+76; GW+91] [9] [MWK76; ML79; MW94; Mor80; MT87; MKW77b; MKW77a; ML82; Mor+83] [10] [Lea87; Lea82; LM80; ML79; ML82] [11] [CFW65; CW63; RW63; RW64b; RW64a; Wyb64a; Wyb64b; Wyb65; Wyb70; WS07] [12] [Tre52; Tre51; Tre58] [13] [RW63; RW64b; RW64a; Raj65]

Contents

1 The semi-empirical Hamiltonian

Electrons in a multi-electron ion are subject to a number of interactions. They are attracted to the nucleus about which they orbit. Being bundled together with other electrons, they experience repulsion from all of them. Having spin, they are also subject to various magnetic interactions. The spin of each electron interacts with the magnetic field generated by its own orbital angular momentum and of other electrons. And between pairs of electrons, the spin of one can influence the others spin through the interaction of their respective magnetic dipoles.

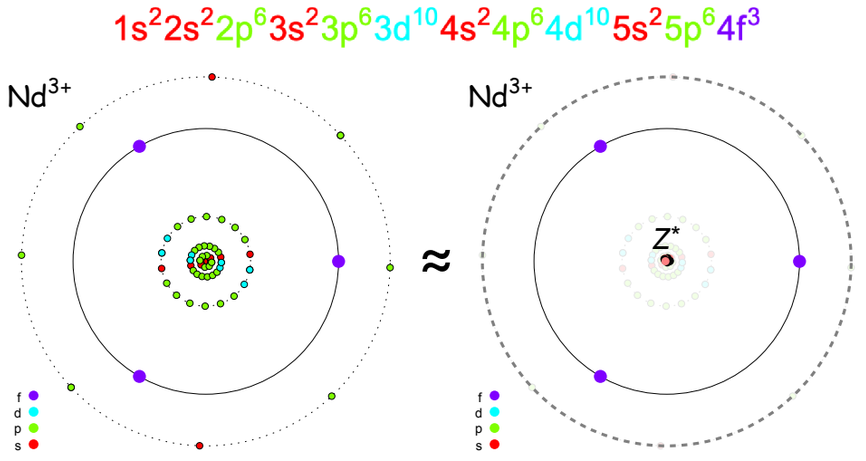

Figure 1: The trivalent neodymium ion shielded by $5s^2$ and $5p^6$ closed shells.

To describe the effect of the charges in the lattice surrounding the ion, the crystal field is introduced. In the simplest of embodiments, the crystal field is simply seen as the electrostatic field due to surrounding charges. This model is of limited applicability if taken too literally; however, if only symmetry considerations are assumed, the model is seen to have greater validity but a somewhat less clear physical origin.

The Hilbert space of a multi-electron ion is a vast stage. In principle, a basis for it should have a countable infinity of bound states and an uncountable infinity of unbound states. This is clearly too much to handle, but thankfully, this large stage can be put in some order thanks to the exclusion principle. The exclusion principle (together with that graceful tendency of things to drift downwards the energetic wells) provides the shell structure. This shell structure, in turn, makes it possible that an atom with many electrons, can be described effectively as an aggregate of an inert core, and a fewer active valence electrons.

Take for instance a triply ionized (or trivalent) neodymium atom, as depicted in $\mathcal{F}$ig-1. In principle, this gives us the daunting task of dealing with the enormous Hilbert space of 57 electrons. However, 54 of them arrange themselves in a xenon core, so that we are only left to deal with only three. Three are still a challenging task, but much less so than 57. Furthermore, the exclusion principle also guides us in what type of orbital we could possibly place these three electrons, in the case of the lanthanide ions, this being the 4f orbitals. But not really, there are many more unoccupied orbitals outside of the xenon core, two of these electrons, if they are willing to pay the energetic price, they could find themselves in a 5d or a 6s orbital.

Here we shall assume a single-configuration description. Meaning that all the valence electrons in the ions that we study will all be considered to be located in f-orbitals, or what is the same, that they are described by $\underline{f}^n$ wavefunctions. Table 2 shows the (ground) configuration for the trivalent lanthanide ions. This is, however, a harsh approximation, but thankfully one can make some corrections to it. The effects that arise in the single configuration description because of omitting all the other possible orbitals where the

58	59	60	61	62	63	64	65	66	67	68	69	70
Ce^{3+}	Pr^{3+}	Nd^{3+}	Pm^{3+}	Sm^{3+}	Eu^{3+}	Gd^{3+}	Tb^{3+}	Dy^{3+}	Ho^{3+}	Er^{3+}	Tm^{3+}	Yb^{3+}
[Xe] f^1	[Xe] f^2	[Xe] f^3	[Xe] f^4	[Xe] f^5	[Xe] f^6	[Xe] f^7	[Xe] f^8	[Xe] f^9	[Xe] f^{10}	[Xe] f^{11}	[Xe] f^{12}	[Xe] f^{13}
Cerium	Praseodymium	Neodymium	Promethium	Samarium	Europium	Gadolinium	Terbium	Dysprosium	Holmium	Erbium	Thulium	Ytterbium

Figure 2: The trivalent lanthanide row and their ground configurations.

electrons might find themselves, this is what is called *configuration-interaction*.

These effects can be brought within the simplified description through perturbation theory. The task not the usual one of correcting for the energies/eigenvectors given an added perturbation, but rather to consider the effects of using a truncated Hilbert space due to a known interaction. For a detailed analysis of this, see Rudzikas' book [Rud07] on theoretical atomic spectroscopy or this article [Lin74] by Lindgren. What results from this analysis are operators that now act solely within the single configuration but with a coefficient that depends on overlap integrals between different configurations. It is from *configuration-interaction* that the parameters $\alpha, \beta, \gamma, P^{(k)}, T^{(k)}$ enter into the description.

$$\hat{\mathcal{H}} = \underbrace{\hat{\mathcal{H}}_{\text{k}}}_{\text{kinetic}} + \underbrace{\hat{\mathcal{H}}_{\text{e:sn}}}_{\text{e:shielded nuc}} + \overbrace{\underbrace{\hat{\mathcal{H}}_{\text{s:o}}}_{\text{spin-orbit}} + \underbrace{\hat{\mathcal{H}}_{\text{s:s}}}_{\substack{\text{spin:spin} \\ \text{and spin:other-orbit}}} + \underbrace{\hat{\mathcal{H}}_{\text{s:oo}\oplus\text{ecs:o}}}_{\substack{\text{spin:other-orbit} \\ \text{ec-correlated-spin:orbit}}}}^{\text{magnetic interactions}} + \underbrace{\hat{\mathcal{H}}_{\text{Z}}}_{\text{Zeeman}}$$
$$+ \overbrace{\underbrace{\hat{\mathcal{H}}_{\text{e:e}}}_{\text{e:e}} + \underbrace{\hat{\mathcal{H}}_{\mathcal{SO}(3)}}_{\text{Trees effective op}} + \underbrace{\hat{\mathcal{H}}_{\mathcal{G}_2}}_{\text{G}_2\text{ effective op}} + \underbrace{\hat{\mathcal{H}}_{\mathcal{SO}(7)}}_{\mathcal{SO}(7)\text{ effective op}} + \underbrace{\hat{\mathcal{H}}_{\text{A}}}_{\substack{\text{effective} \\ \text{three-body}}} + \underbrace{\hat{\mathcal{H}}_{\text{cf}}}_{\text{crystal field}}}^{\text{electric interactions}} \tag{1}$$

$$\hat{\mathcal{H}}_{\text{k}} = -\frac{\hbar^2}{2m_e}\sum_{i=1}^{n}\nabla_i^2 \text{ (kinetic energy of } n \text{ valence electrons)} \tag{2}$$

$$\hat{\mathcal{H}}_{\text{e:sn}} = \sum_{i=1}^{n} V_{\text{sn}}(r_i) \text{ (valence-electrons interaction with shielded nuc. charge)} \tag{3}$$

$$\hat{\mathcal{H}}_{\text{s:o}} = \begin{cases} \sum_{i=1}^{n}\xi(r_i)\left(\hat{\underline{s}}_i\cdot\hat{\underline{\ell}}_i\right) & \text{with } \xi(r_i) = \frac{\hbar^2}{2m^2c^2r_i}\frac{\mathrm{d}V_{\text{sn}}(r_i)}{\mathrm{d}r_i} \\ \sum_{i=1}^{n}\zeta\left(\hat{\underline{s}}_i\cdot\hat{\underline{\ell}}_i\right) & \substack{\text{with } \zeta \text{ the radial average of } \xi(r_i) \\ \text{or used as phenomenological parameter}} \end{cases} \tag{4}$$

$$\hat{\mathcal{H}}_{\text{s:s}} = \sum_{k=0,2,4} m^{(k)}\hat{m}_k{}^{ss} \tag{5}$$

$$\hat{\mathcal{H}}_{\text{s:oo}\oplus\text{ecs:o}} = \sum_{k=2,4,6} P^{(k)}\hat{p}_k + \sum_{k=0,2,4} m^{(k)}\hat{m}_k \tag{6}$$

$$\hat{\mathcal{H}}_{\text{Z}} = -\vec{B}\cdot\hat{\mu} = \mu_B\vec{B}\cdot\left(\hat{L} + g_s\hat{S}\right) \text{ (interaction with a magnetic field)} \tag{7}$$

$$\hat{\mathcal{H}}_{\text{e:e}} = \sum_{i>j}^{n,n}\frac{e^2}{\|\vec{r}_i - \vec{r}_j\|} = \sum_{k=0,2,4,6} F^{(k)}\hat{f}_k \text{ (repulsion between valence electrons)} \tag{8}$$

Let $\hat{C}(\mathcal{G}) \coloneqq$ The Casimir operator of group $\mathcal{G}$.

$$\hat{\mathcal{H}}_{\mathcal{SO}(3)} = \alpha\,\hat{C}(\mathcal{SO}(3)) = \alpha\hat{L}^2 \text{ (Trees effective operator)} \tag{9}$$

$$\hat{\mathcal{H}}_{\mathcal{G}_2} = \beta\,\hat{C}(\mathcal{G}_2) \tag{10}$$

$$\hat{\mathcal{H}}_{\mathcal{SO}(7)} = \gamma\,\hat{C}(\mathcal{SO}(7)) \tag{11}$$

$$\hat{\mathcal{H}}_{\text{A}} = T'^{(2)}\hat{t}_2{}' + T'^{(11)}\hat{t}_{11}{}' + \sum_{\substack{k=2,3,4,6,7,8, \\ 11,12,14,15, \\ 16,17,18,19}} T^{(k)}\hat{t}_k \text{ (effective 3-body int.)} \tag{12}$$

$$\hat{\mathcal{H}}_{\text{cf}} = \sum_{i=1}^{n} V_{\text{CF}}(\hat{r}_i) = \sum_{i=1}^{n}\sum_{k=2,4,6}\sum_{q=-k}^{k}\mathcal{B}_q^{(k)}\mathcal{C}_q^{(k)}(i) \quad \substack{\text{(crystal field interaction} \\ \text{with surroudings)}} \tag{13}$$

One could try to evaluate the coefficients that result in the Hamiltonian. However, within the **semi-empirical** approach, these parameters are left to be fitted against experimental data, or at times approximated through Hartree-Fock analysis. This approach is only *semi* empirical in the sense that the model parameters are fitted from experimental data, but the semi-empirical Hamiltonian that is fitted is based on a clear physical picture inherited from atomic physics.

Putting all of this together leads to the following effective Hamiltonian as show in &qn-1, where "v-electrons" is shorthand for valence electrons. It is important to note that the eigenstates that we'll end up with have shoved under the rug all the radial dependence of the wavefunctions. This dependence having been integrated in the parameters of the effective Hamiltonian. The resulting wavefunctions being solely concerned with the angular

dependence of the wavefunctions, but modulated by the effects of the radial dependence.

Once all the parameters in this semi-empirical Hamiltonian have been fitted to experimental datal what results is a Hamiltonian such as the one for Pr^{3+} in LaF_3 shown in $\mathcal{F}$ig-3. Before we go on to explain in some detail each of the terms included in this Hamiltonian, let us continue to explain the basis used in calculations.

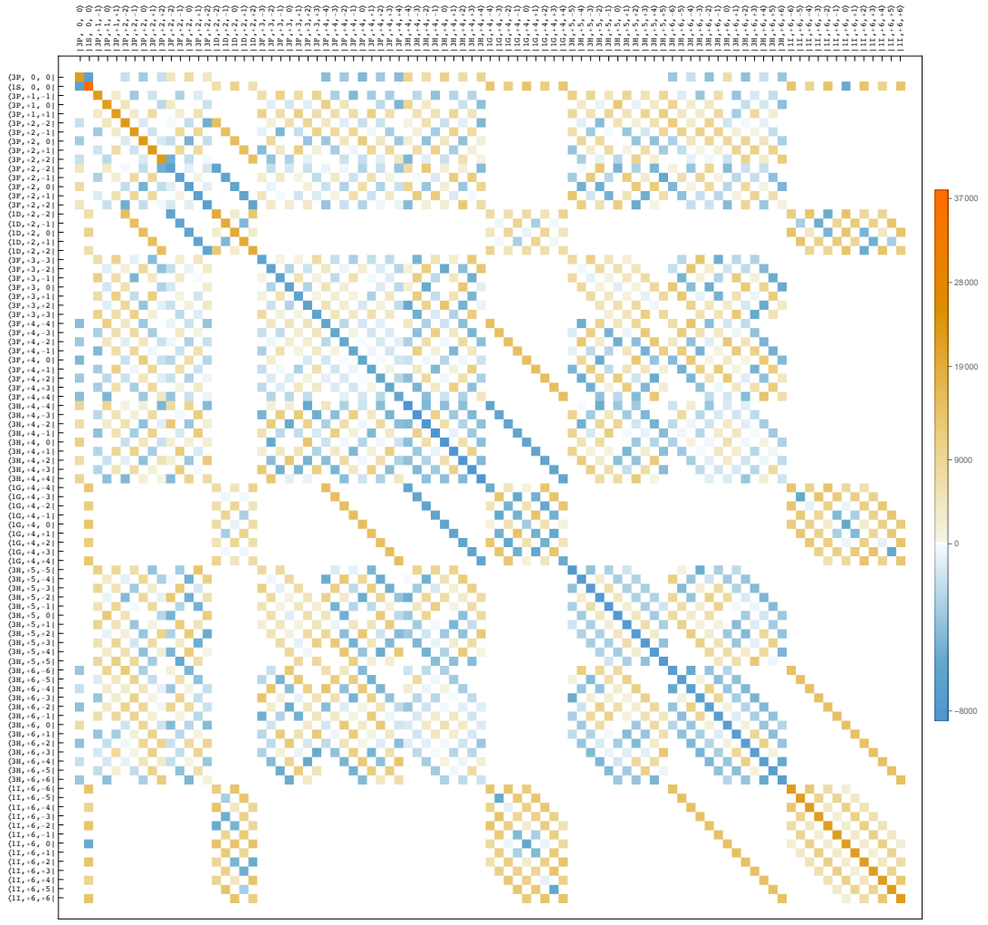

Figure 3: The matrix representation of $\hat{\mathcal{H}}$ for Pr^{3+} in LaF_3 in the $|LSJM_J\rangle$ basis.

2 LS coupling basis

In choosing a coupling scheme (or equivalently, choosing a basis in which to represent the Hamiltonian), there are a myriad options; all of them legitimate in their own right. The art of choosing a useful coupling scheme is that of proposing a basis for the angular part of the wavefunctions that will be close to the actual eigenstates of the system. It being necessary to calculate the matrix elements of the relevant operators, choosing a coupling scheme may also be justified by the easy by which these can be calculated.

qlanth uses LS coupling for its calculations. In LS coupling all the orbital angular momenta are added to form the total orbital angular momentum L, all the spin angular momenta are added to form the total spin angular momentum S, and finally these two angular momenta are then added together to form the total angular momentum J. The exclusion principle is taken into account in limiting the possible LS terms, and demands no further restrictions. Finally this total angular momentum J is complemented with the quantum number[14] M_J describing the projection of J along the z-axis.

It is worthwhile remembering here the spectroscopic hierarchy of descriptive elements: **terms** correspond to $|LS\rangle$ (also noted as $^{2S+1}$L), **levels** correspond to $|LSJ\rangle$ (also noted as $^{2S+1}L_J$), and **states** correspond to $|LSJM_J\rangle$ (also noted as $^{2S+1}L_{J,M_J}$). $\mathcal{F}$ig-4 shows an example of the relationship between a term and its associated levels and states.

In principle the $|LSJM_J\rangle$ description is the primordial one, the $|LSJ\rangle$ resulting from neglecting all parts of the Hamiltonian that have no spherical symmetry, and the $|LS\rangle$

[14] A *good* quantum number is any eigenvalue of an operator that commutes with the Hamiltonian; in other words, they are conserved quantities.

resulting from further neglecting all terms that couple the spin and orbital angular momenta. Note that a *state* is not an *eigen*-state; all of these are assumed to be basis vectors in the type of description attached to them.

Whereas all four quantum numbers $|LSJM_J\rangle$ are required to specify a state, one may, however, use two simpler descriptions as the situation merits. When all the parts of the Hamiltonian without spherical symmetry are excluded, then a description in terms of $|LSJ\rangle$ levels is sufficient, the M_J quantum numbers being redundant and with J being a good quantum number. In a second scenario, when in addition to neglecting all parts without spherical symmetry, one also neglects all parts of the Hamiltonian that couple the spin and orbital degrees of freedom, then the $|LS\rangle$ terms constitute the most parsimonious description, with L and S being separately conserved quantities.

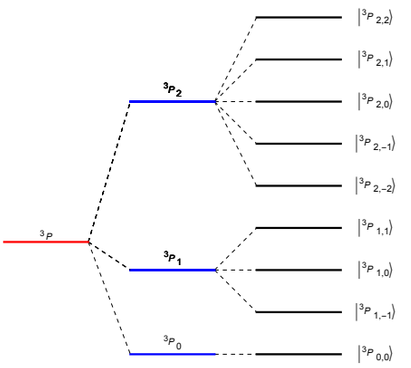

Figure 4: Levels and states associated with the ^3P term in $\underline{\mathrm{f}}^2$.

When a certain level of description has been adopted one can then assume (at one's own peril) that single states, levels, or terms are actual *eigen*-states/levels/terms of the system at hand. This assumption results in simple transition rules between states/levels/terms. One may, however, within each level of description, take an alternate route, the *intermediate coupling* route, of seeing how the different states/levels/terms mix in the eigenstates found by diagonalizing the appropriate Hamiltonian. This results in a more detailed description at the cost of increased complexity.

2.1 $|LSJM_J\rangle$ states

The basis vectors of the $|LSJM_J\rangle$ basis are common eigenvectors of the operators $\hat{L}^2$, $\hat{S}^2$, $\hat{J}^2$, and $\hat{J}_z$. They are formed starting from the allowed LS terms in a given configuration, and are then completed with attendant J and M_J quantum numbers. The LS terms allowed in each configuration $\underline{\mathrm{f}}^n$ are obtained from tables that originate from the original work by Nielson and Koster [NK63]. In **qlanth** these terms are parsed from the file `B1F_-ALL.TXT` which is part of the doctoral research of Dobromir Velkov (under the advisory of Brian Judd) [Vel00] in which he calculated anew the coefficients of fractional parentage.

One of the facts that have to be accounted for in a basis that uses L and S as quantum numbers, is that there might be several linearly independent paths to couple the electron spin and orbital momenta to add up to given total L and total S. For this reason additional labels are necessary to distinguish between these different terms. The simplest way of doing this dates back to the tables of Nielson and Koster [NK63], and consists in assigning consecutive integers to degenerate LS terms, with no further meaning to these integers, except that of discriminating between degenerate terms.

The following are all the LS terms in the $\underline{\mathrm{f}}^n$ configurations. In the notation used, the superscript index before the letter notes the spin multiplicity $2S+1$, the roman letter indicates the value of L in spectroscopic notation ($S\to1, P\to2, D\to3, F\to4, G\to5, H\to6, I\to7, K\to8, L\to9, M\to10, N\to11, O\to12, Q\to3, R\to14, T\to15, U\to16, V\to17$), and the final integer (if present) is the label that discriminates between several degenerate LS and must not be confused with a value of J. This last index we frequently label in the equations contained in this document with the greek letter α (sadly, for historical reasons, we prepend it, rather than append it).

$\underline{\mathrm{f}}^0$ (1 LS term)
^{1}S

$\underline{\mathrm{f}}^1$ (1 LS term)
^{2}F

$\underline{f}^2$
(7 LS terms)

---

^{3}P, ^{3}F, ^{3}H, ^{1}S, ^{1}D, ^{1}G, ^{1}I

$\underline{f}^3$
(17 LS terms)

---

^{4}S, ^{4}D, ^{4}F, ^{4}G, ^{4}I, ^{2}P, ^{2}D1, ^{2}D2, ^{2}F1, ^{2}F2, ^{2}G1, ^{2}G2, ^{2}H1, ^{2}H2, ^{2}I, ^{2}K, ^{2}L

$\underline{f}^4$
(47 LS terms)

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^{5}S, ^{5}D, ^{5}F, ^{5}G, ^{5}I, ^{3}P1, ^{3}P2, ^{3}P3, ^{3}D1, ^{3}D2, ^{3}F1, ^{3}F2, ^{3}F3, ^{3}F4, ^{3}G1, ^{3}G2, ^{3}G3, ^{3}H1, ^{3}H2, ^{3}H3, ^{3}H4, ^{3}I1, ^{3}I2, ^{3}K1, ^{3}K2, ^{3}L, ^{3}M, ^{1}S1, ^{1}S2, ^{1}D1, ^{1}D2, ^{1}D3, ^{1}D4, ^{1}F, ^{1}G1, ^{1}G2, ^{1}G3, ^{1}G4, ^{1}H1, ^{1}H2, ^{1}I1, ^{1}I2, ^{1}I3, ^{1}K, ^{1}L1, ^{1}L2, ^{1}N

$\underline{f}^5$
(73 LS terms)

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^{6}P, ^{6}F, ^{6}H, ^{4}S, ^{4}P1, ^{4}P2, ^{4}D1, ^{4}D2, ^{4}D3, ^{4}F1, ^{4}F2, ^{4}F3, ^{4}F4, ^{4}G1, ^{4}G2, ^{4}G3, ^{4}G4, ^{4}H1, ^{4}H2, ^{4}H3, ^{4}I1, ^{4}I2, ^{4}I3, ^{4}K1, ^{4}K2, ^{4}L, ^{4}M, ^{2}P1, ^{2}P2, ^{2}P3, ^{2}P4, ^{2}D1, ^{2}D2, ^{2}D3, ^{2}D4, ^{2}D5, ^{2}F1, ^{2}F2, ^{2}F3, ^{2}F4, ^{2}F5, ^{2}F6, ^{2}F7, ^{2}G1, ^{2}G2, ^{2}G3, ^{2}G4, ^{2}G5, ^{2}G6, ^{2}H1, ^{2}H2, ^{2}H3, ^{2}H4, ^{2}H5, ^{2}H6, ^{2}H7, ^{2}I1, ^{2}I2, ^{2}I3, ^{2}I4, ^{2}I5, ^{2}K1, ^{2}K2, ^{2}K3, ^{2}K4, ^{2}K5, ^{2}L1, ^{2}L2, ^{2}L3, ^{2}M1, ^{2}M2, ^{2}N, ^{2}O

$\underline{f}^6$
(119 LS terms)

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^{7}F, ^{5}S, ^{5}P, ^{5}D1, ^{5}D2, ^{5}D3, ^{5}F1, ^{5}F2, ^{5}G1, ^{5}G2, ^{5}G3, ^{5}H1, ^{5}H2, ^{5}I1, ^{5}I2, ^{5}K, ^{5}L, ^{3}P1, ^{3}P2, ^{3}P3, ^{3}P4, ^{3}P5, ^{3}P6, ^{3}D1, ^{3}D2, ^{3}D3, ^{3}D4, ^{3}D5, ^{3}F1, ^{3}F2, ^{3}F3, ^{3}F4, ^{3}F5, ^{3}F6, ^{3}F7, ^{3}F8, ^{3}F9, ^{3}G1, ^{3}G2, ^{3}G3, ^{3}G4, ^{3}G5, ^{3}G6, ^{3}G7, ^{3}H1, ^{3}H2, ^{3}H3, ^{3}H4, ^{3}H5, ^{3}H6, ^{3}H7, ^{3}H8, ^{3}H9, ^{3}I1, ^{3}I2, ^{3}I3, ^{3}I4, ^{3}I5, ^{3}I6, ^{3}K1, ^{3}K2, ^{3}K3, ^{3}K4, ^{3}K5, ^{3}K6, ^{3}L1, ^{3}L2, ^{3}L3, ^{3}M1, ^{3}M2, ^{3}M3, ^{3}N, ^{3}O, ^{1}S1, ^{1}S2, ^{1}S3, ^{1}S4, ^{1}P, ^{1}D1, ^{1}D2, ^{1}D3, ^{1}D4, ^{1}D5, ^{1}D6, ^{1}F1, ^{1}F2, ^{1}F3, ^{1}F4, ^{1}G1, ^{1}G2, ^{1}G3, ^{1}G4, ^{1}G5, ^{1}G6, ^{1}G7, ^{1}G8, ^{1}H1, ^{1}H2, ^{1}H3, ^{1}H4, ^{1}I1, ^{1}I2, ^{1}I3, ^{1}I4, ^{1}I5, ^{1}I6, ^{1}I7, ^{1}K1, ^{1}K2, ^{1}K3, ^{1}L1, ^{1}L2, ^{1}L3, ^{1}L4, ^{1}M1, ^{1}M2, ^{1}N1, ^{1}N2, ^{1}Q

$\underline{f}^7$
(119 LS terms)

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^{8}S, ^{6}P, ^{6}D, ^{6}F, ^{6}G, ^{6}H, ^{6}I, ^{4}S1, ^{4}S2, ^{4}P1, ^{4}P2, ^{4}D1, ^{4}D2, ^{4}D3, ^{4}D4, ^{4}D5, ^{4}D6, ^{4}F1, ^{4}F2, ^{4}F3, ^{4}F4, ^{4}F5, ^{4}G1, ^{4}G2, ^{4}G3, ^{4}G4, ^{4}G5, ^{4}G6, ^{4}G7, ^{4}H1, ^{4}H2, ^{4}H3, ^{4}H4, ^{4}H5, ^{4}I1, ^{4}I2, ^{4}I3, ^{4}I4, ^{4}I5, ^{4}K1, ^{4}K2, ^{4}K3, ^{4}L1, ^{4}L2, ^{4}L3, ^{4}M, ^{4}N, ^{2}S1, ^{2}S2, ^{2}P1, ^{2}P2, ^{2}P3, ^{2}P4, ^{2}P5, ^{2}D1, ^{2}D2, ^{2}D3, ^{2}D4, ^{2}D5, ^{2}D6, ^{2}D7, ^{2}F1, ^{2}F2, ^{2}F3, ^{2}F4, ^{2}F5, ^{2}F6, ^{2}F7, ^{2}F8, ^{2}F9, ^{2}F10, ^{2}G1, ^{2}G2, ^{2}G3, ^{2}G4, ^{2}G5, ^{2}G6, ^{2}G7, ^{2}G8, ^{2}G9, ^{2}G10, ^{2}H1, ^{2}H2, ^{2}H3, ^{2}H4, ^{2}H5, ^{2}H6, ^{2}H7, ^{2}H8, ^{2}H9, ^{2}I1, ^{2}I2, ^{2}I3, ^{2}I4, ^{2}I5, ^{2}I6, ^{2}I7, ^{2}I8, ^{2}I9, ^{2}K1, ^{2}K2, ^{2}K3, ^{2}K4, ^{2}K5, ^{2}K6, ^{2}K7, ^{2}L1, ^{2}L2, ^{2}L3, ^{2}L4, ^{2}L5, ^{2}M1, ^{2}M2, ^{2}M3, ^{2}M4, ^{2}N1, ^{2}N2, ^{2}O, ^{2}Q

$\underline{f}^8$
(119 LS terms)

---

^{7}F, ^{5}S, ^{5}P, ^{5}D1, ^{5}D2, ^{5}D3, ^{5}F1, ^{5}F2, ^{5}G1, ^{5}G2, ^{5}G3, ^{5}H1, ^{5}H2, ^{5}I1, ^{5}I2, ^{5}K, ^{5}L, ^{3}P1, ^{3}P2, ^{3}P3, ^{3}P4, ^{3}P5, ^{3}P6, ^{3}D1, ^{3}D2, ^{3}D3, ^{3}D4, ^{3}D5, ^{3}F1, ^{3}F2, ^{3}F3, ^{3}F4, ^{3}F5, ^{3}F6, ^{3}F7, ^{3}F8, ^{3}F9, ^{3}G1, ^{3}G2, ^{3}G3, ^{3}G4, ^{3}G5, ^{3}G6, ^{3}G7, ^{3}H1, ^{3}H2, ^{3}H3, ^{3}H4, ^{3}H5, ^{3}H6, ^{3}H7, ^{3}H8, ^{3}H9, ^{3}I1, ^{3}I2, ^{3}I3, ^{3}I4, ^{3}I5, ^{3}I6, ^{3}K1, ^{3}K2, ^{3}K3, ^{3}K4, ^{3}K5, ^{3}K6, ^{3}L1, ^{3}L2, ^{3}L3, ^{3}M1, ^{3}M2, ^{3}M3, ^{3}N, ^{3}O, ^{1}S1, ^{1}S2, ^{1}S3, ^{1}S4, ^{1}P, ^{1}D1, ^{1}D2, ^{1}D3, ^{1}D4, ^{1}D5, ^{1}D6, ^{1}F1, ^{1}F2, ^{1}F3, ^{1}F4,

^{1}G1, ^{1}G2, ^{1}G3, ^{1}G4, ^{1}G5, ^{1}G6, ^{1}G7, ^{1}G8, ^{1}H1, ^{1}H2, ^{1}H3, ^{1}H4, ^{1}I1, ^{1}I2, ^{1}I3, ^{1}I4, ^{1}I5, ^{1}I6, ^{1}I7, ^{1}K1, ^{1}K2, ^{1}K3, ^{1}L1, ^{1}L2, ^{1}L3, ^{1}L4, ^{1}M1, ^{1}M2, ^{1}N1, ^{1}N2, ^{1}Q

$\underline{\mathrm{f}}^9$ (73 LS terms)
^{6}P, ^{6}F, ^{6}H, ^{4}S, ^{4}P1, ^{4}P2, ^{4}D1, ^{4}D2, ^{4}D3, ^{4}F1, ^{4}F2, ^{4}F3, ^{4}F4, ^{4}G1, ^{4}G2, ^{4}G3, ^{4}G4, ^{4}H1, ^{4}H2, ^{4}H3, ^{4}I1, ^{4}I2, ^{4}I3, ^{4}K1, ^{4}K2, ^{4}L, ^{4}M, ^{2}P1, ^{2}P2, ^{2}P3, ^{2}P4, ^{2}D1, ^{2}D2, ^{2}D3, ^{2}D4, ^{2}D5, ^{2}F1, ^{2}F2, ^{2}F3, ^{2}F4, ^{2}F5, ^{2}F6, ^{2}F7, ^{2}G1, ^{2}G2, ^{2}G3, ^{2}G4, ^{2}G5, ^{2}G6, ^{2}H1, ^{2}H2, ^{2}H3, ^{2}H4, ^{2}H5, ^{2}H6, ^{2}H7, ^{2}I1, ^{2}I2, ^{2}I3, ^{2}I4, ^{2}I5, ^{2}K1, ^{2}K2, ^{2}K3, ^{2}K4, ^{2}K5, ^{2}L1, ^{2}L2, ^{2}L3, ^{2}M1, ^{2}M2, ^{2}N, ^{2}O

$\underline{\mathrm{f}}^{10}$ (47 LS terms)
^{5}S, ^{5}D, ^{5}F, ^{5}G, ^{5}I, ^{3}P1, ^{3}P2, ^{3}P3, ^{3}D1, ^{3}D2, ^{3}F1, ^{3}F2, ^{3}F3, ^{3}F4, ^{3}G1, ^{3}G2, ^{3}G3, ^{3}H1, ^{3}H2, ^{3}H3, ^{3}H4, ^{3}I1, ^{3}I2, ^{3}K1, ^{3}K2, ^{3}L, ^{3}M, ^{1}S1, ^{1}S2, ^{1}D1, ^{1}D2, ^{1}D3, ^{1}D4, ^{1}F, ^{1}G1, ^{1}G2, ^{1}G3, ^{1}G4, ^{1}H1, ^{1}H2, ^{1}I1, ^{1}I2, ^{1}I3, ^{1}K, ^{1}L1, ^{1}L2, ^{1}N

$\underline{\mathrm{f}}^{11}$ (17 LS terms)
^{4}S, ^{4}D, ^{4}F, ^{4}G, ^{4}I, ^{2}P, ^{2}D1, ^{2}D2, ^{2}F1, ^{2}F2, ^{2}G1, ^{2}G2, ^{2}H1, ^{2}H2, ^{2}I, ^{2}K, ^{2}L

$\underline{\mathrm{f}}^{12}$ (7 LS terms)
^{3}P, ^{3}F, ^{3}H, ^{1}S, ^{1}D, ^{1}G, ^{1}I

$\underline{\mathrm{f}}^{13}$ (1 LS term)
^{2}F

$\underline{\mathrm{f}}^{14}$ (1 LS term)
^{1}S

In **qlanth** these terms may be queried through the function `AllowedNKSLTerms`.

```
AllowedNKSLTerms::usage = "AllowedNKSLTerms[numE] returns a list with
    the allowed terms in the f^numE configuration, the terms are
   given as strings in spectroscopic notation. The integers in the
   last positions are used to distinguish cases with degeneracy.";
AllowedNKSLTerms[numE_] := Map[First, CFPTerms[Min[numE, 14-numE]]];
AllowedNKSLTerms[0]  = {"1S"};
AllowedNKSLTerms[14] = {"1S"};
```

In addition to LS, the $|LSJM_J\rangle$ basis states are also specified by the total angular momentum J (which may go from $|L-S|$ to $|L+S|$). Then for each J, there are $2J+1$ projections on the z-axis. For example, the ordered $|LSJM_J\rangle$ basis for $\underline{\mathrm{f}}^2$ is shown below, where the first element is the LS term given as a string, the second equal to J, and the third one equal to M_J:

$(J=0)$ (2 states)
$\lvert^3\mathrm{P}_{0,0}\rangle$, $\lvert^1\mathrm{S}_{0,0}\rangle$

$(J = 1)$
(3 states)

$|^3\mathrm{P}_{1,-1}\rangle$, $|^3\mathrm{P}_{1,0}\rangle$, $|^3\mathrm{P}_{1,1}\rangle$

$(J = 2)$
(15 states)

$|^3\mathrm{P}_{2,-2}\rangle$, $|^3\mathrm{P}_{2,-1}\rangle$, $|^3\mathrm{P}_{2,0}\rangle$, $|^3\mathrm{P}_{2,1}\rangle$, $|^3\mathrm{P}_{2,2}\rangle$, $|^3\mathrm{F}_{2,-2}\rangle$, $|^3\mathrm{F}_{2,-1}\rangle$, $|^3\mathrm{F}_{2,0}\rangle$, $|^3\mathrm{F}_{2,1}\rangle$, $|^3\mathrm{F}_{2,2}\rangle$, $|^1\mathrm{D}_{2,-2}\rangle$, $|^1\mathrm{D}_{2,-1}\rangle$, $|^1\mathrm{D}_{2,0}\rangle$, $|^1\mathrm{D}_{2,1}\rangle$, $|^1\mathrm{D}_{2,2}\rangle$

$(J = 3)$
(7 states)

$|^3\mathrm{F}_{3,-3}\rangle$, $|^3\mathrm{F}_{3,-2}\rangle$, $|^3\mathrm{F}_{3,-1}\rangle$, $|^3\mathrm{F}_{3,0}\rangle$, $|^3\mathrm{F}_{3,1}\rangle$, $|^3\mathrm{F}_{3,2}\rangle$, $|^3\mathrm{F}_{3,3}\rangle$

$(J = 4)$
(27 states)

$|^3\mathrm{F}_{4,-4}\rangle$, $|^3\mathrm{F}_{4,-3}\rangle$, $|^3\mathrm{F}_{4,-2}\rangle$, $|^3\mathrm{F}_{4,-1}\rangle$, $|^3\mathrm{F}_{4,0}\rangle$, $|^3\mathrm{F}_{4,1}\rangle$, $|^3\mathrm{F}_{4,2}\rangle$, $|^3\mathrm{F}_{4,3}\rangle$, $|^3\mathrm{F}_{4,4}\rangle$, $|^3\mathrm{H}_{4,-4}\rangle$, $|^3\mathrm{H}_{4,-3}\rangle$, $|^3\mathrm{H}_{4,-2}\rangle$, $|^3\mathrm{H}_{4,-1}\rangle$, $|^3\mathrm{H}_{4,0}\rangle$, $|^3\mathrm{H}_{4,1}\rangle$, $|^3\mathrm{H}_{4,2}\rangle$, $|^3\mathrm{H}_{4,3}\rangle$, $|^3\mathrm{H}_{4,4}\rangle$, $|^1\mathrm{G}_{4,-4}\rangle$, $|^1\mathrm{G}_{4,-3}\rangle$, $|^1\mathrm{G}_{4,-2}\rangle$, $|^1\mathrm{G}_{4,-1}\rangle$, $|^1\mathrm{G}_{4,0}\rangle$, $|^1\mathrm{G}_{4,1}\rangle$, $|^1\mathrm{G}_{4,2}\rangle$, $|^1\mathrm{G}_{4,3}\rangle$, $|^1\mathrm{G}_{4,4}\rangle$

$(J = 5)$
(11 states)

$|^3\mathrm{H}_{5,-5}\rangle$, $|^3\mathrm{H}_{5,-4}\rangle$, $|^3\mathrm{H}_{5,-3}\rangle$, $|^3\mathrm{H}_{5,-2}\rangle$, $|^3\mathrm{H}_{5,-1}\rangle$, $|^3\mathrm{H}_{5,0}\rangle$, $|^3\mathrm{H}_{5,1}\rangle$, $|^3\mathrm{H}_{5,2}\rangle$, $|^3\mathrm{H}_{5,3}\rangle$, $|^3\mathrm{H}_{5,4}\rangle$, $|^3\mathrm{H}_{5,5}\rangle$

$(J = 6)$
(26 states)

$|^3\mathrm{H}_{6,-6}\rangle$, $|^3\mathrm{H}_{6,-5}\rangle$, $|^3\mathrm{H}_{6,-4}\rangle$, $|^3\mathrm{H}_{6,-3}\rangle$, $|^3\mathrm{H}_{6,-2}\rangle$, $|^3\mathrm{H}_{6,-1}\rangle$, $|^3\mathrm{H}_{6,0}\rangle$, $|^3\mathrm{H}_{6,1}\rangle$, $|^3\mathrm{H}_{6,2}\rangle$, $|^3\mathrm{H}_{6,3}\rangle$, $|^3\mathrm{H}_{6,4}\rangle$, $|^3\mathrm{H}_{6,5}\rangle$, $|^3\mathrm{H}_{6,6}\rangle$, $|^1\mathrm{I}_{6,-6}\rangle$, $|^1\mathrm{I}_{6,-5}\rangle$, $|^1\mathrm{I}_{6,-4}\rangle$, $|^1\mathrm{I}_{6,-3}\rangle$, $|^1\mathrm{I}_{6,-2}\rangle$, $|^1\mathrm{I}_{6,-1}\rangle$, $|^1\mathrm{I}_{6,0}\rangle$, $|^1\mathrm{I}_{6,1}\rangle$, $|^1\mathrm{I}_{6,2}\rangle$, $|^1\mathrm{I}_{6,3}\rangle$, $|^1\mathrm{I}_{6,4}\rangle$, $|^1\mathrm{I}_{6,5}\rangle$, $|^1\mathrm{I}_{6,6}\rangle$

The order above is an example of the bases ordering used in **qlanth**. Notice how the basis vectors are sorted in order of increasing J, so that for instance not all of the basis states associated with the ^{3}P LS term are contiguous. Within each group for a given J the basis kets are then ordered in decreasing S, then ordered in increasing L, and then according to M_J.

In **qlanth** the ordered basis used for a given $\underline{\mathrm{f}}^n$ is provided by `BasisLSJMJ` which provides a list with $\binom{14}{n}$ elements.

```
BasisLSJMJ::usage = "BasisLSJMJ[numE] returns the ordered basis in L-S-J-MJ with the total orbital angular momentum L and total spin angular momentum S coupled together to form J. The function returns a list with each element representing the quantum numbers for each basis vector. Each element is of the form {SL (string in spectroscopic notation),J, MJ}.
The option ''AsAssociation'' can be set to True to return the basis as an association with the keys corresponding to values of J and the values lists with the corresponding {L, S, J, MJ} list. The default of this option is False.
";
Options[BasisLSJMJ] = {"AsAssociation" -> False};
BasisLSJMJ[numE_, OptionsPattern[]] := Module[
  {energyStatesTable, basis, idx1},
  (
    energyStatesTable = BasisTableGenerator[numE];
```

```
    basis = Table[
      energyStatesTable[{numE, AllowedJ[numE][[idx1]]}],
      {idx1, 1, Length[AllowedJ[numE]]}];
    basis = Flatten[basis, 1];
    If[OptionValue["AsAssociation"],
      (
        Js    = AllowedJ[numE];
        basis = Table[(J -> Select[basis, #[[2]] == J &]), {J, Js}];
        basis = Association[basis];
      )
    ];
    Return[basis]
  )
];
```

2.2 More quantum numbers

Besides using an integer which solves the problem of discriminating between degenerate LS terms by enumerating them, it is also possible to add more useful labels that reflect additional symmetries that the f-electron basis states have in the groups $\mathcal{SO}(7)$ (the Lie group of rotations in seven dimensions) and $\mathcal{G}_2$ (the rank-2 exceptional simple Lie group).

2.2.1 Seniority ν

The seniority number connects different LS terms between configurations, so that a term below can be seen as the *senior* of a term above. To determine the seniority of a given term in configuration $\underline{\mathrm{f}}^n$, one must first find the configuration $\underline{\mathrm{f}}^{\tilde{n}}$ in which this term appeared. For example, $\underline{\mathrm{f}}^5$ contains six degenerate ${}^2G$ terms. The first time this term appeared was in $\underline{\mathrm{f}}^3$, where it had a degeneracy of 2. The 2 degenerate terms in $\underline{\mathrm{f}}^3$ would then both have a seniority of $\boldsymbol{\nu} = 3$ since they first appeared in $\underline{\mathrm{f}}^3$. In consequence two of the six degenerate terms in $\underline{\mathrm{f}}^5$ would have the same degeneracy those two in $\underline{\mathrm{f}}^3$, and are therefore linked to those previous two. The four remaining ones, are considered to be *born* in $\underline{\mathrm{f}}^5$, and therefore have a seniority $\boldsymbol{\nu} = 5$.

These rules seem to be ad-hoc, but they are useful in dealing with the degeneracies in the LS terms as they arrive going up the configurations. It provides a useful way of tracking what happens to each *branch* of the coupling tree as it grows and withers with increasing number of electrons.

There is, however, a deeper meaning to the seniority number. It can be shown that the seniority number (more exactly a quantity related to it) is a sort of spin, a *quasi*-spin, where the spin projections along the 'z-axis' correspond to different number of electrons in $\underline{\mathrm{f}}^n$ configurations [Jud67]. This is a consequence of the exclusion principle. It is also useful to relate matrix elements of operators in one configuration to those in another, through the use of the Wigner-Eckart theorem. This is an interesting and useful theoretical construct, but the method of fractional parentage (which is what is implemented in **qlanth**) is equally adequate, albeit being somewhat less parsimonious than what the quasi-spin view that seniority can provide. As such **qlanth** does not use the seniority numbers that are associated with each LS term[15]. However, in **qlanth** the seniority of a given LS term can be obtained using the function `Seniority`.

```
Seniority::usage = "Seniority[LS] returns the seniority of the given
    term.";
Seniority[LS_] := FindNKLSTerm[LS][[1, 2]];
```

2.2.2 $\mathcal{U}$ and $\mathcal{W}$

Much as L tells us how a rotation acts on an L wavefunction by mixing different M_L components, these other two quantum numbers specify how the wavefunctions transform under the operations of two other two groups. The $\mathcal{W}$ label determines how a wavefunction transforms under a rotation in 7-dimensional space, and $\mathcal{U}$ how they transform under an operator of group $\mathcal{G}_2$. Without going into the group theoretical details, the irreducible representations of $\mathcal{SO}(7)$ can be represented by triples of integer numbers, and those of $\mathcal{G}_2$ as pairs of two integers.

[15] Except for calculating the coefficients of fractional parentage beyond $\underline{\mathrm{f}}^7$, which are useful, but not essential to the calculations of **qlanth**.

In **qlanth** the $\mathcal{W}$ and $\mathcal{U}$ are used in order to determine the matrix elements of the $\hat{\mathcal{C}}(\mathcal{SO}(7))$ and $\hat{\mathcal{C}}(\mathcal{G}_2)$ Casimir operators. These labels can be retrieved, for a given LS string, using the function `FindNKLSTerm`.

```
FindNKLSTerm::usage = "Given the string LS FindNKLSTerm[SL] returns
    all the terms that are compatible with it. This is only for f^n
    configurations. The provided terms might belong to more than one
    configuration. The function returns a list with elements of the
    form {LS, seniority, W, U}.";
FindNKLSTerm[SL_] := Module[
  {NKterms, n},
  (
    n = 7;
    NKterms = {{}};
    Map[
      If[! StringFreeQ[First[#], SL],
        If[ToExpression[Part[#, 2]] <= n,
          NKterms = Join[NKterms, {#}, 1]
        ]
      ] &,
    fnTermLabels
    ];
    NKterms = DeleteCases[NKterms, {}];
    NKterms
  )
];
```

2.3 $|LSJ\rangle$ levels

When the Hamiltonian only includes spherically symmetric terms (or what is the same, when the crystal field is neglected) then the M_J quantum numbers in the $|LSJM_J\rangle$ basis states are redundant. This permits a simplified description in terms of $|LSJ\rangle$ levels. The following are the different $^{2S+1}\mathrm{L}_J$ levels that span the eigenvectors that result from diagonalizing the Hamiltonian in the level description, these may also be termed *mutiplets*. (In these we have excluded the indices that distinguish between degenerate LS terms)

$\underline{\mathrm{f}}^1$ (2 LSJ levels)
$^2\mathrm{F}_{5/2}$, $^2\mathrm{F}_{7/2}$

$\underline{\mathrm{f}}^2$ (13 LSJ levels)
$^3\mathrm{P}_0$, $^1\mathrm{S}_0$, $^3\mathrm{P}_1$, $^3\mathrm{P}_2$, $^3\mathrm{F}_2$, $^1\mathrm{D}_2$, $^3\mathrm{F}_3$, $^3\mathrm{F}_4$, $^3\mathrm{H}_4$, $^1\mathrm{G}_4$, $^3\mathrm{H}_5$, $^3\mathrm{H}_6$, $^1\mathrm{I}_6$

$\underline{\mathrm{f}}^3$ (41 LSJ levels)
$^4\mathrm{D}_{1/2}$, $^2\mathrm{P}_{1/2}$, $^4\mathrm{S}_{3/2}$, $^4\mathrm{D}_{3/2}$, $^4\mathrm{F}_{3/2}$, $^2\mathrm{P}_{3/2}$, $^2\mathrm{D}_{3/2}$, $^2\mathrm{D}_{3/2}$, $^4\mathrm{D}_{5/2}$, $^4\mathrm{F}_{5/2}$, $^4\mathrm{G}_{5/2}$, $^2\mathrm{D}_{5/2}$, $^2\mathrm{D}_{5/2}$, $^2\mathrm{F}_{5/2}$, $^2\mathrm{F}_{5/2}$, $^4\mathrm{D}_{7/2}$, $^4\mathrm{F}_{7/2}$, $^4\mathrm{G}_{7/2}$, $^2\mathrm{F}_{7/2}$, $^2\mathrm{F}_{7/2}$, $^2\mathrm{G}_{7/2}$, $^2\mathrm{G}_{7/2}$, $^4\mathrm{F}_{9/2}$, $^4\mathrm{G}_{9/2}$, $^4\mathrm{I}_{9/2}$, $^2\mathrm{G}_{9/2}$, $^2\mathrm{G}_{9/2}$, $^2\mathrm{H}_{9/2}$, $^2\mathrm{H}_{9/2}$, $^4\mathrm{G}_{11/2}$, $^4\mathrm{I}_{11/2}$, $^2\mathrm{H}_{11/2}$, $^2\mathrm{H}_{11/2}$, $^2\mathrm{I}_{11/2}$, $^4\mathrm{I}_{13/2}$, $^2\mathrm{I}_{13/2}$, $^2\mathrm{K}_{13/2}$, $^4\mathrm{I}_{15/2}$, $^2\mathrm{K}_{15/2}$, $^2\mathrm{L}_{15/2}$, $^2\mathrm{L}_{17/2}$

$\underline{\mathrm{f}}^4$ (107 LSJ levels)
$^5\mathrm{D}_0$, $^3\mathrm{P}_0$, $^3\mathrm{P}_0$, $^3\mathrm{P}_0$, $^1\mathrm{S}_0$, $^1\mathrm{S}_0$, $^5\mathrm{D}_1$, $^5\mathrm{F}_1$, $^3\mathrm{P}_1$, $^3\mathrm{P}_1$, $^3\mathrm{P}_1$, $^3\mathrm{D}_1$, $^3\mathrm{D}_1$, $^5\mathrm{S}_2$, $^5\mathrm{D}_2$, $^5\mathrm{F}_2$, $^5\mathrm{G}_2$, $^3\mathrm{P}_2$, $^3\mathrm{P}_2$, $^3\mathrm{P}_2$, $^3\mathrm{D}_2$, $^3\mathrm{D}_2$, $^3\mathrm{F}_2$, $^3\mathrm{F}_2$, $^3\mathrm{F}_2$, $^3\mathrm{F}_2$, $^1\mathrm{D}_2$, $^1\mathrm{D}_2$, $^1\mathrm{D}_2$, $^1\mathrm{D}_2$, $^5\mathrm{D}_3$, $^5\mathrm{F}_3$, $^5\mathrm{G}_3$, $^3\mathrm{D}_3$, $^3\mathrm{D}_3$, $^3\mathrm{F}_3$, $^3\mathrm{F}_3$, $^3\mathrm{F}_3$, $^3\mathrm{F}_3$, $^3\mathrm{G}_3$, $^3\mathrm{G}_3$, $^3\mathrm{G}_3$, $^1\mathrm{F}_3$, $^5\mathrm{D}_4$, $^5\mathrm{F}_4$, $^5\mathrm{G}_4$, $^5\mathrm{I}_4$, $^3\mathrm{F}_4$, $^3\mathrm{F}_4$, $^3\mathrm{F}_4$, $^3\mathrm{F}_4$, $^3\mathrm{G}_4$, $^3\mathrm{G}_4$, $^3\mathrm{G}_4$, $^3\mathrm{H}_4$, $^3\mathrm{H}_4$, $^3\mathrm{H}_4$, $^3\mathrm{H}_4$, $^1\mathrm{G}_4$, $^1\mathrm{G}_4$, $^1\mathrm{G}_4$, $^1\mathrm{G}_4$, $^5\mathrm{F}_5$, $^5\mathrm{G}_5$, $^5\mathrm{I}_5$, $^3\mathrm{G}_5$, $^3\mathrm{G}_5$, $^3\mathrm{G}_5$, $^3\mathrm{H}_5$, $^3\mathrm{H}_5$, $^3\mathrm{H}_5$, $^3\mathrm{H}_5$, $^3\mathrm{I}_5$, $^3\mathrm{I}_5$, $^1\mathrm{H}_5$, $^1\mathrm{H}_5$, $^5\mathrm{G}_6$, $^5\mathrm{I}_6$, $^3\mathrm{H}_6$, $^3\mathrm{H}_6$, $^3\mathrm{H}_6$, $^3\mathrm{H}_6$, $^3\mathrm{I}_6$, $^3\mathrm{I}_6$, $^3\mathrm{K}_6$, $^3\mathrm{K}_6$, $^1\mathrm{I}_6$, $^1\mathrm{I}_6$, $^1\mathrm{I}_6$, $^5\mathrm{I}_7$, $^3\mathrm{I}_7$, $^3\mathrm{I}_7$, $^3\mathrm{K}_7$, $^3\mathrm{K}_7$, $^3\mathrm{L}_7$, $^1\mathrm{K}_7$, $^5\mathrm{I}_8$, $^3\mathrm{K}_8$, $^3\mathrm{K}_8$, $^3\mathrm{L}_8$, $^3\mathrm{M}_8$, $^1\mathrm{L}_8$, $^1\mathrm{L}_8$, $^3\mathrm{L}_9$, $^3\mathrm{M}_9$, $^3\mathrm{M}_{10}$, $^1\mathrm{N}_{10}$

$\underline{\mathrm{f}}^5$ (198 LSJ levels)
$^6\mathrm{F}_{1/2}$, $^4\mathrm{P}_{1/2}$, $^4\mathrm{P}_{1/2}$, $^4\mathrm{D}_{1/2}$, $^4\mathrm{D}_{1/2}$, $^4\mathrm{D}_{1/2}$, $^2\mathrm{P}_{1/2}$, $^2\mathrm{P}_{1/2}$, $^2\mathrm{P}_{1/2}$, $^2\mathrm{P}_{1/2}$, $^6\mathrm{P}_{3/2}$, $^6\mathrm{F}_{3/2}$, $^4\mathrm{S}_{3/2}$, $^4\mathrm{P}_{3/2}$, $^4\mathrm{P}_{3/2}$, $^4\mathrm{D}_{3/2}$, $^4\mathrm{D}_{3/2}$, $^4\mathrm{D}_{3/2}$, $^4\mathrm{F}_{3/2}$, $^4\mathrm{F}_{3/2}$, $^4\mathrm{F}_{3/2}$, $^4\mathrm{F}_{3/2}$, $^2\mathrm{P}_{3/2}$, $^2\mathrm{P}_{3/2}$, $^2\mathrm{P}_{3/2}$, $^2\mathrm{P}_{3/2}$,

$^{2}D_{3/2}$, $^{2}D_{3/2}$, $^{2}D_{3/2}$, $^{2}D_{3/2}$, $^{2}D_{3/2}$, $^{6}P_{5/2}$, $^{6}F_{5/2}$, $^{6}H_{5/2}$, $^{4}P_{5/2}$, $^{4}P_{5/2}$, $^{4}D_{5/2}$, $^{4}D_{5/2}$, $^{4}D_{5/2}$, $^{4}F_{5/2}$, $^{4}F_{5/2}$, $^{4}F_{5/2}$, $^{4}F_{5/2}$, $^{4}G_{5/2}$, $^{4}G_{5/2}$, $^{4}G_{5/2}$, $^{4}G_{5/2}$, $^{2}D_{5/2}$, $^{2}D_{5/2}$, $^{2}D_{5/2}$, $^{2}D_{5/2}$, $^{2}D_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{6}P_{7/2}$, $^{6}F_{7/2}$, $^{6}H_{7/2}$, $^{4}D_{7/2}$, $^{4}D_{7/2}$, $^{4}D_{7/2}$, $^{4}F_{7/2}$, $^{4}F_{7/2}$, $^{4}F_{7/2}$, $^{4}F_{7/2}$, $^{4}G_{7/2}$, $^{4}G_{7/2}$, $^{4}G_{7/2}$, $^{4}G_{7/2}$, $^{4}H_{7/2}$, $^{4}H_{7/2}$, $^{4}H_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{6}F_{9/2}$, $^{6}H_{9/2}$, $^{4}F_{9/2}$, $^{4}F_{9/2}$, $^{4}F_{9/2}$, $^{4}F_{9/2}$, $^{4}G_{9/2}$, $^{4}G_{9/2}$, $^{4}G_{9/2}$, $^{4}G_{9/2}$, $^{4}H_{9/2}$, $^{4}H_{9/2}$, $^{4}H_{9/2}$, $^{4}I_{9/2}$, $^{4}I_{9/2}$, $^{4}I_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{6}F_{11/2}$, $^{6}H_{11/2}$, $^{4}G_{11/2}$, $^{4}G_{11/2}$, $^{4}G_{11/2}$, $^{4}G_{11/2}$, $^{4}H_{11/2}$, $^{4}H_{11/2}$, $^{4}H_{11/2}$, $^{4}I_{11/2}$, $^{4}I_{11/2}$, $^{4}I_{11/2}$, $^{4}I_{11/2}$, $^{4}K_{11/2}$, $^{4}K_{11/2}$, $^{2}H_{11/2}$, $^{2}H_{11/2}$, $^{2}H_{11/2}$, $^{2}H_{11/2}$, $^{2}H_{11/2}$, $^{2}H_{11/2}$, $^{2}H_{11/2}$, $^{2}I_{11/2}$, $^{2}I_{11/2}$, $^{2}I_{11/2}$, $^{2}I_{11/2}$, $^{2}I_{11/2}$, $^{6}H_{13/2}$, $^{4}H_{13/2}$, $^{4}H_{13/2}$, $^{4}H_{13/2}$, $^{4}I_{13/2}$, $^{4}I_{13/2}$, $^{4}I_{13/2}$, $^{4}K_{13/2}$, $^{4}K_{13/2}$, $^{4}L_{13/2}$, $^{2}I_{13/2}$, $^{2}I_{13/2}$, $^{2}I_{13/2}$, $^{2}I_{13/2}$, $^{2}I_{13/2}$, $^{2}K_{13/2}$, $^{2}K_{13/2}$, $^{2}K_{13/2}$, $^{2}K_{13/2}$, $^{2}K_{13/2}$, $^{6}H_{15/2}$, $^{4}I_{15/2}$, $^{4}I_{15/2}$, $^{4}I_{15/2}$, $^{4}K_{15/2}$, $^{4}K_{15/2}$, $^{4}L_{15/2}$, $^{4}M_{15/2}$, $^{2}K_{15/2}$, $^{2}K_{15/2}$, $^{2}K_{15/2}$, $^{2}K_{15/2}$, $^{2}K_{15/2}$, $^{2}L_{15/2}$, $^{2}L_{15/2}$, $^{2}L_{15/2}$, $^{4}K_{17/2}$, $^{4}K_{17/2}$, $^{4}L_{17/2}$, $^{4}M_{17/2}$, $^{2}L_{17/2}$, $^{2}L_{17/2}$, $^{2}L_{17/2}$, $^{2}M_{17/2}$, $^{2}M_{17/2}$, $^{4}L_{19/2}$, $^{4}M_{19/2}$, $^{2}M_{19/2}$, $^{2}M_{19/2}$, $^{2}N_{19/2}$, $^{4}M_{21/2}$, $^{2}N_{21/2}$, $^{2}O_{21/2}$, $^{2}O_{23/2}$

$\underline{f}^{6}$ (295 LSJ levels)

$^{7}F_{0}$, $^{5}D_{0}$, $^{5}D_{0}$, $^{5}D_{0}$, $^{3}P_{0}$, $^{3}P_{0}$, $^{3}P_{0}$, $^{3}P_{0}$, $^{3}P_{0}$, $^{3}P_{0}$, $^{1}S_{0}$, $^{1}S_{0}$, $^{1}S_{0}$, $^{1}S_{0}$, $^{7}F_{1}$, $^{5}P_{1}$, $^{5}D_{1}$, $^{5}D_{1}$, $^{5}D_{1}$, $^{5}F_{1}$, $^{5}F_{1}$, $^{3}P_{1}$, $^{3}P_{1}$, $^{3}P_{1}$, $^{3}P_{1}$, $^{3}P_{1}$, $^{3}P_{1}$, $^{3}D_{1}$, $^{3}D_{1}$, $^{3}D_{1}$, $^{3}D_{1}$, $^{3}D_{1}$, $^{1}P_{1}$, $^{7}F_{2}$, $^{5}S_{2}$, $^{5}P_{2}$, $^{5}D_{2}$, $^{5}D_{2}$, $^{5}D_{2}$, $^{5}F_{2}$, $^{5}F_{2}$, $^{5}G_{2}$, $^{5}G_{2}$, $^{5}G_{2}$, $^{3}P_{2}$, $^{3}P_{2}$, $^{3}P_{2}$, $^{3}P_{2}$, $^{3}P_{2}$, $^{3}P_{2}$, $^{3}D_{2}$, $^{3}D_{2}$, $^{3}D_{2}$, $^{3}D_{2}$, $^{3}D_{2}$, $^{3}D_{2}$, $^{3}F_{2}$, $^{3}F_{2}$, $^{3}F_{2}$, $^{3}F_{2}$, $^{3}F_{2}$, $^{3}F_{2}$, $^{3}F_{2}$, $^{3}F_{2}$, $^{3}F_{2}$, $^{1}D_{2}$, $^{1}D_{2}$, $^{1}D_{2}$, $^{1}D_{2}$, $^{1}D_{2}$, $^{1}D_{2}$, $^{7}F_{3}$, $^{5}P_{3}$, $^{5}D_{3}$, $^{5}D_{3}$, $^{5}D_{3}$, $^{5}F_{3}$, $^{5}F_{3}$, $^{5}G_{3}$, $^{5}G_{3}$, $^{5}G_{3}$, $^{5}H_{3}$, $^{5}H_{3}$, $^{3}D_{3}$, $^{3}D_{3}$, $^{3}D_{3}$, $^{3}D_{3}$, $^{3}D_{3}$, $^{3}F_{3}$, $^{3}F_{3}$, $^{3}F_{3}$, $^{3}F_{3}$, $^{3}F_{3}$, $^{3}F_{3}$, $^{3}F_{3}$, $^{3}F_{3}$, $^{3}G_{3}$, $^{3}G_{3}$, $^{3}G_{3}$, $^{3}G_{3}$, $^{3}G_{3}$, $^{3}G_{3}$, $^{3}G_{3}$, $^{1}F_{3}$, $^{1}F_{3}$, $^{1}F_{3}$, $^{1}F_{3}$, $^{7}F_{4}$, $^{5}D_{4}$, $^{5}D_{4}$, $^{5}D_{4}$, $^{5}F_{4}$, $^{5}F_{4}$, $^{5}G_{4}$, $^{5}G_{4}$, $^{5}G_{4}$, $^{5}H_{4}$, $^{5}H_{4}$, $^{5}I_{4}$, $^{5}I_{4}$, $^{3}F_{4}$, $^{3}F_{4}$, $^{3}F_{4}$, $^{3}F_{4}$, $^{3}F_{4}$, $^{3}F_{4}$, $^{3}F_{4}$, $^{3}F_{4}$, $^{3}F_{4}$, $^{3}G_{4}$, $^{3}G_{4}$, $^{3}G_{4}$, $^{3}G_{4}$, $^{3}G_{4}$, $^{3}G_{4}$, $^{3}G_{4}$, $^{3}H_{4}$, $^{3}H_{4}$, $^{3}H_{4}$, $^{3}H_{4}$, $^{3}H_{4}$, $^{3}H_{4}$, $^{3}H_{4}$, $^{3}H_{4}$, $^{3}H_{4}$, $^{1}G_{4}$, $^{1}G_{4}$, $^{1}G_{4}$, $^{1}G_{4}$, $^{1}G_{4}$, $^{1}G_{4}$, $^{1}G_{4}$, $^{7}F_{5}$, $^{5}F_{5}$, $^{5}F_{5}$, $^{5}G_{5}$, $^{5}G_{5}$, $^{5}G_{5}$, $^{5}H_{5}$, $^{5}H_{5}$, $^{5}I_{5}$, $^{5}I_{5}$, $^{5}K_{5}$, $^{3}G_{5}$, $^{3}G_{5}$, $^{3}G_{5}$, $^{3}G_{5}$, $^{3}G_{5}$, $^{3}G_{5}$, $^{3}G_{5}$, $^{3}H_{5}$, $^{3}H_{5}$, $^{3}H_{5}$, $^{3}H_{5}$, $^{3}H_{5}$, $^{3}H_{5}$, $^{3}H_{5}$, $^{3}H_{5}$, $^{3}H_{5}$, $^{3}I_{5}$, $^{3}I_{5}$, $^{3}I_{5}$, $^{3}I_{5}$, $^{3}I_{5}$, $^{3}I_{5}$, $^{1}H_{5}$, $^{1}H_{5}$, $^{1}H_{5}$, $^{1}H_{5}$, $^{7}F_{6}$, $^{5}G_{6}$, $^{5}G_{6}$, $^{5}G_{6}$, $^{5}H_{6}$, $^{5}H_{6}$, $^{5}I_{6}$, $^{5}I_{6}$, $^{5}K_{6}$, $^{5}L_{6}$, $^{3}H_{6}$, $^{3}H_{6}$, $^{3}H_{6}$, $^{3}H_{6}$, $^{3}H_{6}$, $^{3}H_{6}$, $^{3}H_{6}$, $^{3}H_{6}$, $^{3}H_{6}$, $^{3}I_{6}$, $^{3}I_{6}$, $^{3}I_{6}$, $^{3}I_{6}$, $^{3}I_{6}$, $^{3}I_{6}$, $^{3}K_{6}$, $^{3}K_{6}$, $^{3}K_{6}$, $^{3}K_{6}$, $^{3}K_{6}$, $^{3}K_{6}$, $^{1}I_{6}$, $^{1}I_{6}$, $^{1}I_{6}$, $^{1}I_{6}$, $^{1}I_{6}$, $^{1}I_{6}$, $^{1}I_{6}$, $^{5}H_{7}$, $^{5}H_{7}$, $^{5}I_{7}$, $^{5}I_{7}$, $^{5}K_{7}$, $^{5}L_{7}$, $^{3}I_{7}$, $^{3}I_{7}$, $^{3}I_{7}$, $^{3}I_{7}$, $^{3}I_{7}$, $^{3}I_{7}$, $^{3}K_{7}$, $^{3}K_{7}$, $^{3}K_{7}$, $^{3}K_{7}$, $^{3}K_{7}$, $^{3}K_{7}$, $^{3}L_{7}$, $^{3}L_{7}$, $^{3}L_{7}$, $^{1}K_{7}$, $^{1}K_{7}$, $^{1}K_{7}$, $^{5}I_{8}$, $^{5}I_{8}$, $^{5}K_{8}$, $^{5}L_{8}$, $^{3}K_{8}$, $^{3}K_{8}$, $^{3}K_{8}$, $^{3}K_{8}$, $^{3}K_{8}$, $^{3}K_{8}$, $^{3}L_{8}$, $^{3}L_{8}$, $^{3}L_{8}$, $^{3}M_{8}$, $^{3}M_{8}$, $^{3}M_{8}$, $^{1}L_{8}$, $^{1}L_{8}$, $^{1}L_{8}$, $^{1}L_{8}$, $^{5}K_{9}$, $^{5}L_{9}$, $^{3}L_{9}$, $^{3}L_{9}$, $^{3}L_{9}$, $^{3}M_{9}$, $^{3}M_{9}$, $^{3}M_{9}$, $^{3}N_{9}$, $^{1}M_{9}$, $^{1}M_{9}$, $^{5}L_{10}$, $^{3}M_{10}$, $^{3}M_{10}$, $^{3}M_{10}$, $^{3}N_{10}$, $^{3}O_{10}$, $^{1}N_{10}$, $^{1}N_{10}$, $^{3}N_{11}$, $^{3}O_{11}$, $^{3}O_{12}$, $^{1}Q_{12}$

$\underline{f}^{7}$ (327 LSJ levels)

$^{6}D_{1/2}$, $^{6}F_{1/2}$, $^{4}P_{1/2}$, $^{4}P_{1/2}$, $^{4}D_{1/2}$, $^{4}D_{1/2}$, $^{4}D_{1/2}$, $^{4}D_{1/2}$, $^{4}D_{1/2}$, $^{4}D_{1/2}$, $^{2}S_{1/2}$, $^{2}S_{1/2}$, $^{2}P_{1/2}$, $^{2}P_{1/2}$, $^{2}P_{1/2}$, $^{2}P_{1/2}$, $^{2}P_{1/2}$, $^{6}P_{3/2}$, $^{6}D_{3/2}$, $^{6}F_{3/2}$, $^{6}G_{3/2}$, $^{4}S_{3/2}$, $^{4}S_{3/2}$, $^{4}P_{3/2}$, $^{4}P_{3/2}$, $^{4}D_{3/2}$, $^{4}D_{3/2}$, $^{4}D_{3/2}$, $^{4}D_{3/2}$, $^{4}D_{3/2}$, $^{4}F_{3/2}$, $^{4}F_{3/2}$, $^{4}F_{3/2}$, $^{4}F_{3/2}$, $^{4}F_{3/2}$, $^{2}P_{3/2}$, $^{2}P_{3/2}$, $^{2}P_{3/2}$, $^{2}P_{3/2}$, $^{2}P_{3/2}$, $^{2}D_{3/2}$, $^{2}D_{3/2}$, $^{2}D_{3/2}$, $^{2}D_{3/2}$, $^{2}D_{3/2}$, $^{2}D_{3/2}$, $^{6}P_{5/2}$, $^{6}D_{5/2}$, $^{6}F_{5/2}$, $^{6}G_{5/2}$, $^{6}H_{5/2}$, $^{4}P_{5/2}$, $^{4}P_{5/2}$, $^{4}D_{5/2}$, $^{4}D_{5/2}$, $^{4}D_{5/2}$, $^{4}D_{5/2}$, $^{4}D_{5/2}$, $^{4}D_{5/2}$, $^{4}F_{5/2}$, $^{4}F_{5/2}$, $^{4}F_{5/2}$, $^{4}F_{5/2}$, $^{4}F_{5/2}$, $^{4}G_{5/2}$, $^{4}G_{5/2}$, $^{4}G_{5/2}$, $^{4}G_{5/2}$, $^{4}G_{5/2}$, $^{4}G_{5/2}$, $^{4}G_{5/2}$, $^{2}D_{5/2}$, $^{2}D_{5/2}$, $^{2}D_{5/2}$, $^{2}D_{5/2}$, $^{2}D_{5/2}$, $^{2}D_{5/2}$, $^{2}D_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{2}F_{5/2}$, $^{8}S_{7/2}$, $^{6}P_{7/2}$, $^{6}D_{7/2}$, $^{6}F_{7/2}$, $^{6}G_{7/2}$, $^{6}H_{7/2}$, $^{6}I_{7/2}$, $^{4}D_{7/2}$, $^{4}D_{7/2}$, $^{4}D_{7/2}$, $^{4}D_{7/2}$, $^{4}D_{7/2}$, $^{4}D_{7/2}$, $^{4}F_{7/2}$, $^{4}F_{7/2}$, $^{4}F_{7/2}$, $^{4}F_{7/2}$, $^{4}F_{7/2}$, $^{4}G_{7/2}$, $^{4}G_{7/2}$, $^{4}G_{7/2}$, $^{4}G_{7/2}$, $^{4}G_{7/2}$, $^{4}G_{7/2}$, $^{4}G_{7/2}$, $^{4}H_{7/2}$, $^{4}H_{7/2}$, $^{4}H_{7/2}$, $^{4}H_{7/2}$, $^{4}H_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}F_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{2}G_{7/2}$, $^{6}D_{9/2}$, $^{6}F_{9/2}$, $^{6}G_{9/2}$, $^{6}H_{9/2}$, $^{6}I_{9/2}$, $^{4}F_{9/2}$, $^{4}F_{9/2}$, $^{4}F_{9/2}$, $^{4}F_{9/2}$, $^{4}F_{9/2}$, $^{4}G_{9/2}$, $^{4}G_{9/2}$, $^{4}G_{9/2}$, $^{4}G_{9/2}$, $^{4}G_{9/2}$, $^{4}G_{9/2}$, $^{4}G_{9/2}$, $^{4}H_{9/2}$, $^{4}H_{9/2}$, $^{4}H_{9/2}$, $^{4}H_{9/2}$, $^{4}H_{9/2}$, $^{4}I_{9/2}$, $^{4}I_{9/2}$, $^{4}I_{9/2}$, $^{4}I_{9/2}$, $^{4}I_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}G_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{2}H_{9/2}$, $^{6}F_{11/2}$, $^{6}G_{11/2}$, $^{6}H_{11/2}$, $^{6}I_{11/2}$, $^{4}G_{11/2}$, $^{4}G_{11/2}$, $^{4}G_{11/2}$, $^{4}G_{11/2}$, $^{4}G_{11/2}$, $^{4}G_{11/2}$, $^{4}H_{11/2}$, $^{4}H_{11/2}$, $^{4}H_{11/2}$, $^{4}H_{11/2}$, $^{4}H_{11/2}$, $^{4}I_{11/2}$, $^{4}I_{11/2}$, $^{4}I_{11/2}$, $^{4}I_{11/2}$, $^{4}I_{11/2}$, $^{4}K_{11/2}$, $^{4}K_{11/2}$, $^{4}K_{11/2}$, $^{2}H_{11/2}$, $^{2}H_{11/2}$, $^{2}H_{11/2}$, $^{2}H_{11/2}$, $^{2}H_{11/2}$, $^{2}H_{11/2}$, $^{2}H_{11/2}$, $^{2}H_{11/2}$, $^{2}I_{11/2}$, $^{2}I_{11/2}$, $^{2}I_{11/2}$, $^{2}I_{11/2}$, $^{2}I_{11/2}$, $^{2}I_{11/2}$, $^{2}I_{11/2}$, $^{2}I_{11/2}$, $^{2}I_{11/2}$, $^{6}G_{13/2}$, $^{6}H_{13/2}$, $^{6}I_{13/2}$, $^{4}H_{13/2}$, $^{4}H_{13/2}$, $^{4}H_{13/2}$, $^{4}H_{13/2}$, $^{4}H_{13/2}$, $^{4}I_{13/2}$, $^{4}I_{13/2}$, $^{4}I_{13/2}$, $^{4}I_{13/2}$, $^{4}I_{13/2}$, $^{4}K_{13/2}$, $^{4}K_{13/2}$, $^{4}K_{13/2}$, $^{4}L_{13/2}$, $^{4}L_{13/2}$, $^{4}L_{13/2}$, $^{2}I_{13/2}$, $^{2}I_{13/2}$, $^{2}I_{13/2}$, $^{2}I_{13/2}$, $^{2}I_{13/2}$, $^{2}I_{13/2}$, $^{2}I_{13/2}$, $^{2}I_{13/2}$, $^{2}K_{13/2}$, $^{2}K_{13/2}$, $^{2}K_{13/2}$, $^{2}K_{13/2}$, $^{2}K_{13/2}$, $^{2}K_{13/2}$, $^{2}K_{13/2}$, $^{6}H_{15/2}$, $^{6}I_{15/2}$, $^{4}I_{15/2}$, $^{4}I_{15/2}$, $^{4}I_{15/2}$, $^{4}I_{15/2}$, $^{4}I_{15/2}$, $^{4}K_{15/2}$, $^{4}K_{15/2}$,

$^4K_{15/2}$, $^4L_{15/2}$, $^4L_{15/2}$, $^4L_{15/2}$, $^4M_{15/2}$, $^2K_{15/2}$, $^2K_{15/2}$, $^2K_{15/2}$, $^2K_{15/2}$, $^2K_{15/2}$, $^2K_{15/2}$, $^2K_{15/2}$, $^2L_{15/2}$, $^2L_{15/2}$, $^2L_{15/2}$, $^2L_{15/2}$, $^6I_{17/2}$, $^4K_{17/2}$, $^4K_{17/2}$, $^4K_{17/2}$, $^4L_{17/2}$, $^4L_{17/2}$, $^4L_{17/2}$, $^4M_{17/2}$, $^4N_{17/2}$, $^2L_{17/2}$, $^2L_{17/2}$, $^2L_{17/2}$, $^2L_{17/2}$, $^2L_{17/2}$, $^2M_{17/2}$, $^2M_{17/2}$, $^2M_{17/2}$, $^2M_{17/2}$, $^4L_{19/2}$, $^4L_{19/2}$, $^4L_{19/2}$, $^4M_{19/2}$, $^4N_{19/2}$, $^2M_{19/2}$, $^2M_{19/2}$, $^2M_{19/2}$, $^2M_{19/2}$, $^2N_{19/2}$, $^2N_{19/2}$, $^4M_{21/2}$, $^4N_{21/2}$, $^2N_{21/2}$, $^2N_{21/2}$, $^2O_{21/2}$, $^4N_{23/2}$, $^2O_{23/2}$, $^2Q_{23/2}$, $^2Q_{25/2}$

$\underline{f}^8$ (295 LSJ levels)

$^7F_0$, $^5D_0$, $^5D_0$, $^5D_0$, $^3P_0$, $^3P_0$, $^3P_0$, $^3P_0$, $^3P_0$, $^3P_0$, $^1S_0$, $^1S_0$, $^1S_0$, $^1S_0$, $^7F_1$, $^5P_1$, $^5D_1$, $^5D_1$, $^5D_1$, $^5F_1$, $^5F_1$, $^3P_1$, $^3P_1$, $^3P_1$, $^3P_1$, $^3P_1$, $^3P_1$, $^3D_1$, $^3D_1$, $^3D_1$, $^3D_1$, $^3D_1$, $^1P_1$, $^7F_2$, $^5S_2$, $^5P_2$, $^5D_2$, $^5D_2$, $^5D_2$, $^5F_2$, $^5F_2$, $^5G_2$, $^5G_2$, $^5G_2$, $^3P_2$, $^3P_2$, $^3P_2$, $^3P_2$, $^3P_2$, $^3P_2$, $^3D_2$, $^3D_2$, $^3D_2$, $^3D_2$, $^3D_2$, $^3F_2$, $^3F_2$, $^3F_2$, $^3F_2$, $^3F_2$, $^3F_2$, $^3F_2$, $^3F_2$, $^3F_2$, $^1D_2$, $^1D_2$, $^1D_2$, $^1D_2$, $^1D_2$, $^1D_2$, $^7F_3$, $^5P_3$, $^5D_3$, $^5D_3$, $^5D_3$, $^5F_3$, $^5F_3$, $^5G_3$, $^5G_3$, $^5G_3$, $^5H_3$, $^5H_3$, $^3D_3$, $^3D_3$, $^3D_3$, $^3D_3$, $^3D_3$, $^3F_3$, $^3F_3$, $^3F_3$, $^3F_3$, $^3F_3$, $^3F_3$, $^3F_3$, $^3F_3$, $^3G_3$, $^3G_3$, $^3G_3$, $^3G_3$, $^3G_3$, $^3G_3$, $^3G_3$, $^1F_3$, $^1F_3$, $^1F_3$, $^1F_3$, $^7F_4$, $^5D_4$, $^5D_4$, $^5D_4$, $^5F_4$, $^5F_4$, $^5G_4$, $^5G_4$, $^5G_4$, $^5H_4$, $^5H_4$, $^5I_4$, $^5I_4$, $^3F_4$, $^3F_4$, $^3F_4$, $^3F_4$, $^3F_4$, $^3F_4$, $^3F_4$, $^3F_4$, $^3F_4$, $^3G_4$, $^3G_4$, $^3G_4$, $^3G_4$, $^3G_4$, $^3G_4$, $^3G_4$, $^3H_4$, $^3H_4$, $^3H_4$, $^3H_4$, $^3H_4$, $^3H_4$, $^3H_4$, $^3H_4$, $^3H_4$, $^1G_4$, $^1G_4$, $^1G_4$, $^1G_4$, $^1G_4$, $^1G_4$, $^1G_4$, $^7F_5$, $^5F_5$, $^5F_5$, $^5G_5$, $^5G_5$, $^5G_5$, $^5H_5$, $^5H_5$, $^5I_5$, $^5I_5$, $^5K_5$, $^3G_5$, $^3G_5$, $^3G_5$, $^3G_5$, $^3G_5$, $^3G_5$, $^3H_5$, $^3H_5$, $^3H_5$, $^3H_5$, $^3H_5$, $^3H_5$, $^3H_5$, $^3H_5$, $^3I_5$, $^3I_5$, $^3I_5$, $^3I_5$, $^3I_5$, $^1H_5$, $^1H_5$, $^1H_5$, $^1H_5$, $^7F_6$, $^5G_6$, $^5G_6$, $^5G_6$, $^5H_6$, $^5H_6$, $^5I_6$, $^5I_6$, $^5K_6$, $^5L_6$, $^3H_6$, $^3H_6$, $^3H_6$, $^3H_6$, $^3H_6$, $^3H_6$, $^3H_6$, $^3H_6$, $^3H_6$, $^3I_6$, $^3I_6$, $^3I_6$, $^3I_6$, $^3I_6$, $^3I_6$, $^3K_6$, $^3K_6$, $^3K_6$, $^3K_6$, $^3K_6$, $^3K_6$, $^1I_6$, $^1I_6$, $^1I_6$, $^1I_6$, $^1I_6$, $^1I_6$, $^1I_6$, $^5H_7$, $^5H_7$, $^5I_7$, $^5I_7$, $^5K_7$, $^5L_7$, $^3I_7$, $^3I_7$, $^3I_7$, $^3I_7$, $^3I_7$, $^3I_7$, $^3K_7$, $^3K_7$, $^3K_7$, $^3K_7$, $^3K_7$, $^3K_7$, $^3L_7$, $^3L_7$, $^3L_7$, $^1K_7$, $^1K_7$, $^1K_7$, $^5I_8$, $^5I_8$, $^5K_8$, $^5L_8$, $^3K_8$, $^3K_8$, $^3K_8$, $^3K_8$, $^3K_8$, $^3K_8$, $^3L_8$, $^3L_8$, $^3L_8$, $^3M_8$, $^3M_8$, $^3M_8$, $^1L_8$, $^1L_8$, $^1L_8$, $^1L_8$, $^5K_9$, $^5L_9$, $^3L_9$, $^3L_9$, $^3L_9$, $^3M_9$, $^3M_9$, $^3M_9$, $^3N_9$, $^1M_9$, $^1M_9$, $^5L_{10}$, $^3M_{10}$, $^3M_{10}$, $^3M_{10}$, $^3N_{10}$, $^3O_{10}$, $^1N_{10}$, $^1N_{10}$, $^3N_{11}$, $^3O_{11}$, $^3O_{12}$, $^1Q_{12}$

$\underline{f}^9$ (198 LSJ levels)

$^6F_{1/2}$, $^4P_{1/2}$, $^4P_{1/2}$, $^4D_{1/2}$, $^4D_{1/2}$, $^4D_{1/2}$, $^2P_{1/2}$, $^2P_{1/2}$, $^2P_{1/2}$, $^2P_{1/2}$, $^6P_{3/2}$, $^6F_{3/2}$, $^4S_{3/2}$, $^4P_{3/2}$, $^4P_{3/2}$, $^4D_{3/2}$, $^4D_{3/2}$, $^4D_{3/2}$, $^4F_{3/2}$, $^4F_{3/2}$, $^4F_{3/2}$, $^4F_{3/2}$, $^2P_{3/2}$, $^2P_{3/2}$, $^2P_{3/2}$, $^2P_{3/2}$, $^2D_{3/2}$, $^2D_{3/2}$, $^2D_{3/2}$, $^2D_{3/2}$, $^2D_{3/2}$, $^6P_{5/2}$, $^6F_{5/2}$, $^6H_{5/2}$, $^4P_{5/2}$, $^4P_{5/2}$, $^4D_{5/2}$, $^4D_{5/2}$, $^4D_{5/2}$, $^4F_{5/2}$, $^4F_{5/2}$, $^4F_{5/2}$, $^4F_{5/2}$, $^4G_{5/2}$, $^4G_{5/2}$, $^4G_{5/2}$, $^4G_{5/2}$, $^2D_{5/2}$, $^2D_{5/2}$, $^2D_{5/2}$, $^2D_{5/2}$, $^2D_{5/2}$, $^2F_{5/2}$, $^2F_{5/2}$, $^2F_{5/2}$, $^2F_{5/2}$, $^2F_{5/2}$, $^2F_{5/2}$, $^2F_{5/2}$, $^6P_{7/2}$, $^6F_{7/2}$, $^6H_{7/2}$, $^4D_{7/2}$, $^4D_{7/2}$, $^4D_{7/2}$, $^4F_{7/2}$, $^4F_{7/2}$, $^4F_{7/2}$, $^4F_{7/2}$, $^4G_{7/2}$, $^4G_{7/2}$, $^4G_{7/2}$, $^4G_{7/2}$, $^4H_{7/2}$, $^4H_{7/2}$, $^4H_{7/2}$, $^2F_{7/2}$, $^2F_{7/2}$, $^2F_{7/2}$, $^2F_{7/2}$, $^2F_{7/2}$, $^2F_{7/2}$, $^2F_{7/2}$, $^2G_{7/2}$, $^2G_{7/2}$, $^2G_{7/2}$, $^2G_{7/2}$, $^2G_{7/2}$, $^2G_{7/2}$, $^6F_{9/2}$, $^6H_{9/2}$, $^4F_{9/2}$, $^4F_{9/2}$, $^4F_{9/2}$, $^4F_{9/2}$, $^4G_{9/2}$, $^4G_{9/2}$, $^4G_{9/2}$, $^4G_{9/2}$, $^4H_{9/2}$, $^4H_{9/2}$, $^4H_{9/2}$, $^4I_{9/2}$, $^4I_{9/2}$, $^4I_{9/2}$, $^2G_{9/2}$, $^2G_{9/2}$, $^2G_{9/2}$, $^2G_{9/2}$, $^2G_{9/2}$, $^2G_{9/2}$, $^2H_{9/2}$, $^2H_{9/2}$, $^2H_{9/2}$, $^2H_{9/2}$, $^2H_{9/2}$, $^2H_{9/2}$, $^2H_{9/2}$, $^6F_{11/2}$, $^6H_{11/2}$, $^4G_{11/2}$, $^4G_{11/2}$, $^4G_{11/2}$, $^4G_{11/2}$, $^4H_{11/2}$, $^4H_{11/2}$, $^4H_{11/2}$, $^4I_{11/2}$, $^4I_{11/2}$, $^4I_{11/2}$, $^4K_{11/2}$, $^4K_{11/2}$, $^2H_{11/2}$, $^2H_{11/2}$, $^2H_{11/2}$, $^2H_{11/2}$, $^2H_{11/2}$, $^2H_{11/2}$, $^2H_{11/2}$, $^2I_{11/2}$, $^2I_{11/2}$, $^2I_{11/2}$, $^2I_{11/2}$, $^2I_{11/2}$, $^6H_{13/2}$, $^4H_{13/2}$, $^4H_{13/2}$, $^4H_{13/2}$, $^4I_{13/2}$, $^4I_{13/2}$, $^4I_{13/2}$, $^4K_{13/2}$, $^4K_{13/2}$, $^4L_{13/2}$, $^2I_{13/2}$, $^2I_{13/2}$, $^2I_{13/2}$, $^2I_{13/2}$, $^2I_{13/2}$, $^2K_{13/2}$, $^2K_{13/2}$, $^2K_{13/2}$, $^2K_{13/2}$, $^2K_{13/2}$, $^6H_{15/2}$, $^4I_{15/2}$, $^4I_{15/2}$, $^4I_{15/2}$, $^4K_{15/2}$, $^4K_{15/2}$, $^4L_{15/2}$, $^4M_{15/2}$, $^2K_{15/2}$, $^2K_{15/2}$, $^2K_{15/2}$, $^2K_{15/2}$, $^2K_{15/2}$, $^2L_{15/2}$, $^2L_{15/2}$, $^2L_{15/2}$, $^4K_{17/2}$, $^4K_{17/2}$, $^4L_{17/2}$, $^4M_{17/2}$, $^2L_{17/2}$, $^2L_{17/2}$, $^2L_{17/2}$, $^2M_{17/2}$, $^2M_{17/2}$, $^4L_{19/2}$, $^4M_{19/2}$, $^2M_{19/2}$, $^2M_{19/2}$, $^2N_{19/2}$, $^4M_{21/2}$, $^2N_{21/2}$, $^2O_{21/2}$, $^2O_{23/2}$

$\underline{f}^{10}$ (107 LSJ levels)

$^5D_0$, $^3P_0$, $^3P_0$, $^3P_0$, $^1S_0$, $^1S_0$, $^5D_1$, $^5F_1$, $^3P_1$, $^3P_1$, $^3P_1$, $^3D_1$, $^3D_1$, $^5S_2$, $^5D_2$, $^5F_2$, $^5G_2$, $^3P_2$, $^3P_2$, $^3P_2$, $^3D_2$, $^3D_2$, $^3F_2$, $^3F_2$, $^3F_2$, $^3F_2$, $^1D_2$, $^1D_2$, $^1D_2$, $^1D_2$, $^5D_3$, $^5F_3$, $^5G_3$, $^3D_3$, $^3D_3$, $^3F_3$, $^3F_3$, $^3F_3$, $^3F_3$, $^3G_3$, $^3G_3$, $^3G_3$, $^1F_3$, $^5D_4$, $^5F_4$, $^5G_4$, $^5I_4$, $^3F_4$, $^3F_4$, $^3F_4$, $^3F_4$, $^3G_4$, $^3G_4$, $^3G_4$, $^3H_4$, $^3H_4$, $^3H_4$, $^3H_4$, $^1G_4$, $^1G_4$, $^1G_4$, $^1G_4$, $^5F_5$, $^5G_5$, $^5I_5$, $^3G_5$, $^3G_5$, $^3G_5$, $^3H_5$, $^3H_5$, $^3H_5$, $^3H_5$, $^3I_5$, $^3I_5$, $^1H_5$, $^1H_5$, $^5G_6$, $^5I_6$, $^3H_6$, $^3H_6$, $^3H_6$, $^3H_6$, $^3I_6$, $^3I_6$, $^3K_6$, $^3K_6$, $^1I_6$, $^1I_6$, $^1I_6$, $^5I_7$, $^3I_7$, $^3I_7$, $^3K_7$, $^3K_7$, $^3L_7$, $^1K_7$, $^5I_8$, $^3K_8$, $^3K_8$, $^3L_8$, $^3M_8$, $^1L_8$, $^1L_8$, $^3L_9$, $^3M_9$, $^3M_{10}$, $^1N_{10}$

$\underline{f}^{11}$ (41 LSJ levels)

$^4D_{1/2}$, $^2P_{1/2}$, $^4S_{3/2}$, $^4D_{3/2}$, $^4F_{3/2}$, $^2P_{3/2}$, $^2D_{3/2}$, $^2D_{3/2}$, $^4D_{5/2}$, $^4F_{5/2}$, $^4G_{5/2}$, $^2D_{5/2}$, $^2D_{5/2}$, $^2F_{5/2}$, $^2F_{5/2}$, $^4D_{7/2}$, $^4F_{7/2}$, $^4G_{7/2}$, $^2F_{7/2}$, $^2F_{7/2}$, $^2G_{7/2}$, $^2G_{7/2}$, $^4F_{9/2}$, $^4G_{9/2}$, $^4I_{9/2}$, $^2G_{9/2}$,

$^2G_{9/2}$, $^2H_{9/2}$, $^2H_{9/2}$, $^4G_{11/2}$, $^4I_{11/2}$, $^2H_{11/2}$, $^2H_{11/2}$, $^2I_{11/2}$, $^4I_{13/2}$, $^2I_{13/2}$, $^2K_{13/2}$, $^4I_{15/2}$, $^2K_{15/2}$, $^2L_{15/2}$, $^2L_{17/2}$

$\underline{f}^{12}$ (13 LSJ levels)
$^3P_0$, $^1S_0$, $^3P_1$, $^3P_2$, $^3F_2$, $^1D_2$, $^3F_3$, $^3F_4$, $^3H_4$, $^1G_4$, $^3H_5$, $^3H_6$, $^1I_6$

$\underline{f}^{13}$ (2 LSJ levels)
$^2F_{5/2}$, $^2F_{7/2}$

The level picture is a much more frugal description of the eigenstates. Not only are the number of basis elements that need to be considered much less than otherwise, but also the diagonalization is more efficient since it can be carried out within subspaces of shared J. One needs, however, to use adequate degeneracy factors in the relevant calculations.

In **qlanth** the function BasisLSJ can be used to retrieve the ordered basis that is used for the intermediate coupling description in terms of levels.

```
BasisLSJ::usage = "BasisLSJ[numE] returns the level basis LSJ. The function returns a list with each element representing the quantum numbers for each basis vector. Each element is of the form {SL (string in spectroscopic notation), J}.
The option ''AsAssociation'' can be set to True to return the basis as an association with the keys being the allowed J values. The default is False.
  ";
Options[BasisLSJ]={"AsAssociation"->False};
BasisLSJ[numE_,OptionsPattern[]] := Module[
  {Js, basis},
  (
    Js    = AllowedJ[numE];
    basis = BasisLSJMJ[numE,"AsAssociation"->False];
    basis = DeleteDuplicates[{#[[1]],#[[2]]}& /@ basis];
    If[OptionValue["AsAssociation"],
      (
        basis = Association @ Table[(J->Select[basis,#[[2]]==J&]),{J, Js}]
      )
    ];
    Return[basis];
  )
];
```

To obtain the blocks (indexed by J) representing the Hamiltonian in the level description, the function LevelSimplerSymbolicHamMatrix is provided in **qlanth**.

```
LevelSimplerSymbolicHamMatrix::usage = "LevelSimplerSymbolicHamMatrix[numE] is a variation of HamMatrixAssembly that returns the diagonal JJ Hamiltonian blocks applying a simplifier and with simplifications adequate for the level description. The keys of the given association correspond to the different values of J that are possible for f^numE, the values are sparse array that are meant to be interpreted in the basis provided by BasisLSJ.
The option ''Simplifier'' is a list of symbols that are set to zero. At a minimum this has to include the crystal field parameters. By default this includes everything except the Slater parameters Fk and the spin orbit coupling ζ.
The option ''Export'' controls whether the resulting association is saved to disk, the default is True and the resulting file is saved to the ./hams/ folder. A hash is appended to the filename that corresponds to the simplifier used in the resulting expression. If the option ''Overwrite'' is set to False then these files may be used to quickly retrieve a previously computed case. The file is saved both in .m and .mx format.
The option ''PrependToFilename'' can be used to append a string to the filename to which the function may export to.
The option ''Return'' can be used to choose whether the function returns the matrix or not.
The option ''Overwrite'' can be used to overwrite the file if it already exists.";
Options[LevelSimplerSymbolicHamMatrix] = {
```

```
  "Export" -> True,
  "PrependToFilename" -> "",
  "Overwrite" -> False,
  "Return" -> True,
  "Simplifier" -> Join[
    {F0,\[Sigma]SS},
    cfSymbols,
    TSymbols,
    casimirSymbols,
    pseudoMagneticSymbols,
    marvinSymbols,
    DeleteCases[magneticSymbols,ζ]
  ]
};
LevelSimplerSymbolicHamMatrix[numE_Integer, OptionsPattern[]] :=
   Module[
  {thisHamAssoc, Js, fname,
  fnamemx, hash, simplifier},
  (
    simplifier = (#->0)&/@Sort[OptionValue["Simplifier"]];
    hash       = Hash[simplifier];
    If[Not[ValueQ[ElectrostaticTable]],LoadElectrostatic[];
    If[Not[ValueQ[SOOandECSOTable]],LoadSOOandECSO[];
    If[Not[ValueQ[SpinOrbitTable]],LoadSpinOrbit[];
    If[Not[ValueQ[SpinSpinTable]],LoadSpinSpin[];
    If[Not[ValueQ[ThreeBodyTable]],LoadThreeBody[];
    fname   = FileNameJoin[{moduleDir,"hams",OptionValue["
    PrependToFilename"]<>"Level-SymbolicMatrix-f"<>ToString[numE]<>"-"
    <>ToString[hash]<>".m"}];
    fnamemx = FileNameJoin[{moduleDir,"hams",OptionValue["
    PrependToFilename"]<>"Level-SymbolicMatrix-f"<>ToString[numE]<>"-"
    <>ToString[hash]<>".mx"}];
    If[Or[FileExistsQ[fname],FileExistsQ[fnamemx]]&&Not[OptionValue["
    Overwrite"]],
    (
      If[OptionValue["Return"],
      (
        Which[FileExistsQ[fnamemx],
        (
          Print["File ",fnamemx," already exists, and option ''
    Overwrite'' is set to False, loading file ..."];
          thisHamAssoc=Import[fnamemx];
          Return[thisHamAssoc];
        ),
        FileExistsQ[fname],
        (
          Print["File ",fname," already exists, and option ''
    Overwrite'' is set to False, loading file ..."];
          thisHamAssoc=Import[fname];
          Print["Exporting to file ",fnamemx," for quicker loading."
    ];
          Export[fnamemx,thisHamAssoc];
          Return[thisHamAssoc];
        )
        ]
      ),
      (
        Print["File ",fname," already exists, skipping ..."];
        Return[Null];
      )
      ]
    )
    ];
    Js           = AllowedJ[numE];
    thisHamAssoc = HamMatrixAssembly[numE,
      "Set t2Switch"->True,
      "IncludeZeeman"->False,
      "ReturnInBlocks"->True
    ];
    thisHamAssoc = Diagonal[thisHamAssoc];
    thisHamAssoc = Map[SparseArray[ReplaceInSparseArray[#,simplifier
    ]]&,thisHamAssoc,{1}];
    thisHamAssoc = FreeHam[thisHamAssoc,numE];
    thisHamAssoc = AssociationThread[Js->thisHamAssoc];
    If[OptionValue["Export"],
      (
```

```
        Print["Exporting to file ",fname," and to ",fnamemx];
        Export[fname,thisHamAssoc];
        Export[fnamemx,thisHamAssoc];
      )
    ];
    If[OptionValue["Return"],
      Return[thisHamAssoc],
      Return[Null]
    ];
  )
];
```

Whereas this description may be calculated without ever making explicit reference to M_J, in **qlanth** what is done is that the more verbose description associated with the $|LSJM_J\rangle$ basis is "downsized" to obtain the description in terms of levels. For this aim the following functions in **qlanth** are instrumental: `EigenLever`, `FreeHam`, `ListRepeater`, and `ListLever`.

The function `LevelSolver` can be used to calculate the level structure for given values of the parameters that one wishes to keep for the level description, which is often simply termed the *free-ion* part of the Hamiltonian.

```
LevelSolver::usage = "LevelSolver[numE, params] puts together (or
    retrieves from disk) the symbolic level Hamiltonian for the f^numE
     configuration and solves it for the given params returning the
    resultant energies and eigenstates.
If the option ''Return as states'' is set to False, then the function
     returns an association whose keys are values for J in f^numE, and
     whose values are lists with two elements. The first element being
     equal to the ordered basis for the corresponding subpsace, given
    as a list of lists of the form {LS string, J}. The second element
    being another list of two elements, the first element being equal
    to the energies and the second being equal to the corresponding
    normalized eigenvectors. The energies given have been subtracted
    the energy of the ground state.
If the option ''Return as states'' is set to True, then the function
    returns a list with three elements. The first element is the
    global level basis for the f^numE configuration, given as a list
    of lists of the form {LS string, J}. The second element are the
    mayor LSJ components in the returned eigenstates. The third
    element is a list of lists with three elements, in each list the
    first element being equal to the energy, the second being equal to
     the value of J, and the third being equal to the corresponding
    normalized eigenvector (given as a row). The energies given have
    been subtracted the energy of the ground state, and the states
    have been sorted in order of increasing energy.
The following options are admitted:
  - ''Overwrite Hamiltonian'', if set to True the function will
    overwrite the symbolic Hamiltonian. Default is False.
  - ''Return as states'', see description above. Default is True.
  - ''Simplifier'', this is a list with symbols that are set to zero
    for defining the parameters kept in the level  description.
";
Options[LevelSolver] = {
  "Overwrite Hamiltonian" -> False,
  "Return as states" -> True,
  "Simplifier" -> Join[
    cfSymbols,
    TSymbols,
    casimirSymbols,
    pseudoMagneticSymbols,
    marvinSymbols,
    DeleteCases[magneticSymbols,ζ]
  ],
  "PrintFun" -> PrintTemporary
};
LevelSolver[numE_Integer, params0_Association, OptionsPattern[]] :=
    Module[
  {ln, simplifier, simpleHam, basis,
  numHam, eigensys, startTime, endTime,
  diagonalTime, params=params0, globalBasis,
  eigenVectors, eigenEnergies, eigenJs,
  states, groundEnergy, allEnergies, PrintFun},
  (
    ln          = theLanthanides[[numE]];
    basis       = BasisLSJ[numE, "AsAssociation"->True];
```

```
    simplifier = OptionValue["Simplifier"];
    PrintFun   = OptionValue["PrintFun"];
    PrintFun["> LevelSolver for ",ln," with ",numE," f-electrons."];
    PrintFun["> Loading the symbolic level Hamiltonian ..."];
    simpleHam  = LevelSimplerSymbolicHamMatrix[numE,
      "Simplifier" -> simplifier,
      "Overwrite"  -> OptionValue["Overwrite Hamiltonian"]
    ];
    (* Everything that is not given is set to zero *)
    PrintFun["> Setting to zero every parameter not given ..."];
    params     = ParamPad[params, "PrintFun" -> PrintFun];
    PrintFun[params];
    (* Create the numeric hamiltonian *)
    PrintFun["> Replacing parameters in the J-blocks of the
    Hamiltonian to produce numeric arrays ..."];
    numHam     = N /@ Map[ReplaceInSparseArray[#, params]&, simpleHam
    ];
    Clear[simpleHam];
    (* Eigensolver *)
    PrintFun["> Diagonalizing the numerical Hamiltonian within each
    separate J-subspace ..."];
    startTime    = Now;
    eigensys     = Eigensystem /@ numHam;
    endTime      = Now;
    diagonalTime = QuantityMagnitude[endTime-startTime,"Seconds"];
    allEnergies  = Flatten[First/@Values[eigensys]];
    groundEnergy = Min[allEnergies];
    eigensys     = Map[Chop[{#[[1]]-groundEnergy,#[[2]]}]&,eigensys];
    eigensys     = Association@KeyValueMap[#1->{basis[#1],#2} &,
    eigensys];
    PrintFun[">> Diagonalization took ",diagonalTime," seconds."];
    If[OptionValue["Return as states"],
      (
        PrintFun["> Padding the eigenvectors to correspond to the
    level basis ..."];
        eigenVectors  = SparseArray @ BlockDiagonalMatrix[Values
    [#[[2, 2]] & /@ eigensys]];
        globalBasis   = Flatten[Values[basis], 1];
        eigenEnergies = Flatten[Values[#[[2, 1]] & /@ eigensys]];
        eigenJs       = Flatten[KeyValueMap[ConstantArray[#1, Length
    [#2[[2, 2]]]] &, eigensys]];
        states        = Transpose[{eigenEnergies, eigenJs,
    eigenVectors}];
        states        = SortBy[states, First];
        eigenVectors  = Last /@ states;
        LSJmultiplets = (RemoveTrailingDigits[#[[1]]] <> ToString[
    InputForm[#[[2]]]]) & /@ globalBasis;
        majorComponentIndices = Ordering[Abs[#]][[-1]] & /@
    eigenVectors;
        levelLabels           = LSJmultiplets[[majorComponentIndices
    ]];
        Return[{globalBasis, levelLabels, states}];
      ),
      Return[{basis, eigensys}]
    ];
  )
];
```

2.4 The coefficients of fractional parentage

In the 1920s and 1930s, when spectroscopic evidence was being studied to inform the emergent quantum mechanics, one conceptual tool that was put forward for the analysis of the complex spectra of ions [BG34] involved using the spectrum of an ion at one stage of ionization to understand another stage. For instance, using the fourth spectrum of oxygen (OIV) in order to understand the third spectrum (OIII) of the same element.

In 1943 Giulio Racah [Rac43] provided a useful extension to this idea. In addition of using the energies of one spectrum to span the energies of another, Racah extended this idea to the wavefunctions themselves, such that from configuration $\underline{\mathrm{f}}^{n-1}$ one can create the wavefunctions for $\underline{\mathrm{f}}^{n}$ with all the required antisymmetry and normalization conditions. In this approach, a given *daughter* term in $\underline{\mathrm{f}}^{n}$ has a number of *parent* terms in $\underline{\mathrm{f}}^{n-1}$, with the coefficients of fractional parentage determining how much of each parent is in the daughter

as a sum over parents

$$|\underline{\ell}^n \alpha LS\rangle = \sum_{\bar{\alpha}\bar{L}\bar{S}} \underbrace{\left(\underline{\ell}^{n-1}\bar{\alpha}\bar{L}\bar{S}\right\}\underline{\ell}^n \alpha LS\right)}_{\substack{\text{How much of parent } |\underline{\ell}^{n-1}\bar{\alpha}\bar{L}\bar{S}\rangle \\ \text{is in daughter} |\underline{\ell}^n \alpha LS\rangle}} \underbrace{|\left(\underline{\ell}^{n-1}\bar{\alpha}\bar{L}\bar{S}, \underline{\ell}\right) \alpha LS\rangle}_{\substack{\text{Couple an additional } \underline{\ell} \\ \text{to the parent } |\underline{\ell}^{n-1}\bar{\alpha}\bar{L}\bar{S}\rangle}} . \tag{14}$$

More importantly for **qlanth**, the coefficients of fractional parentage can be used to evaluate matrix elements of operators, such as in &qn-29, &qn-51, &qn-65, and &qn-41. These formulas realize a convenient calculation advantage: if one knows matrix elements in one configuration, then one can immediately calculate them in any other configuration.

In principle all the data that is needed in order to evaluate the matrix elements that **qlanth** uses can all be derived from coefficients of fractional parentage, tables of 6-j and 3-j coefficients, the LSUW labels for the terms in the $\underline{f}^n$ configurations, reduced matrix elements in $\underline{f}^3$ for the three-body operators, and reduced matrix elements in $\underline{f}^2$ for the magnetic interactions.

The data for the coefficients of fractional parentage we owe to [Vel00] from which the file `B1F_all.txt` originates, and which we use here to extract this useful "escalator" up the $\underline{f}^n$ configurations.

In **qlanth** the function `GenerateCFPTable` is used to parse the data contained in this file. From this data the association `CFP` is generated. This association has keys that represent LS terms for a configuration $\underline{f}^n$ and values that are lists which contain all the parent terms, together with the corresponding coefficients of fractional parentage.

```
GenerateCFPTable::usage = "GenerateCFPTable[] generates the table for the coefficients of fractional parentage. If the optional parameter ''Export'' is set to True then the resulting data is saved to ./data/CFPTable.m.
The data being parsed here is the file attachment B1F_ALL.TXT which comes from Velkov's thesis.";
Options[GenerateCFPTable] = {"Export" -> True};
GenerateCFPTable[OptionsPattern[]] := Module[
  {rawText, rawLines, leadChar, configIndex, line, daughter,
  lineParts, numberCode, parsedNumber, toAppend, CFPTablefname},
  (
    CleanWhitespace[string_]      := StringReplace[string, RegularExpression["\\s+"]->" "];
    AddSpaceBeforeMinus[string_] := StringReplace[string, RegularExpression["(?<!\\s)-"]->" -"];
    ToIntegerOrString[list_]      := Map[If[StringMatchQ[#, NumberString], ToExpression[#], #] &, list];
    CFPTable       = ConstantArray[{},7];
    CFPTable[[1]] = {{"2F",{"1S",1}}};

    (* Cleaning before processing is useful *)
    rawText  = Import[FileNameJoin[{moduleDir, "data", "B1F_ALL.TXT"}]];
    rawLines = StringTrim/@StringSplit[rawText,"\n"];
    rawLines = Select[rawLines,#!=""&];
    rawLines = CleanWhitespace/@rawLines;
    rawLines = AddSpaceBeforeMinus/@rawLines;

    Do[(
      (* the first character can be used to identify the start of a block *)
      leadChar=StringTake[line,{1}];
      (* ..FN, N is at position 50 in that line *)
      If[leadChar=="[",
      (
        configIndex=ToExpression[StringTake[line,{50}]];
        Continue[];
      )
      ];
      (* Identify which daughter term is being listed *)
      If[StringContainsQ[line,"[DAUGHTER TERM]"],
        daughter=StringSplit[line,"["][[1]];
        CFPTable[[configIndex]]=Append[CFPTable[[configIndex]],{daughter}];
        Continue[];
      ];
      (* Once we get here we are already parsing a row with coefficient data *)
      lineParts     = StringSplit[line," "];
```

```
      parent       = lineParts[[1]];
      numberCode   = ToIntegerOrString[lineParts[[3;;]]];
      parsedNumber = SquarePrimeToNormal[numberCode];
      toAppend     = {parent,parsedNumber};
      CFPTable[[configIndex]][[-1]] = Append[CFPTable[[configIndex
    ]][[-1]], toAppend]
    ),
    {line,rawLines}];
    If[OptionValue["Export"],
      (
      CFPTablefname = FileNameJoin[{moduleDir, "data", "CFPTable.m"
    }];
      Export[CFPTablefname, CFPTable];
      )
    ];
    Return[CFPTable];
  )
];
```

The coefficients of fractional parentage are traditionally only provided up to $\underline{\mathrm{f}}^7$ (such is the case in `B1f_all.txt`), tabulating these beyond $\underline{\mathrm{f}}^7$ would be redundant since the coefficients of fractional parentage beyond $\underline{\mathrm{f}}^7$ satisfy relationships with those below $\underline{\mathrm{f}}^7$. According to [NK63]

$$\left(\underline{\ell}^{(14-n)-1}\bar{\alpha}\bar{L}\bar{S}\right\}\underline{\ell}^{(14-n)}\alpha LS\Big) = \xi(-1)^{S+\bar{S}+L+\bar{L}-7/2}\sqrt{\frac{(n+1)[\![\bar{S}]\!][\![\bar{L}]\!]}{(14-n)[\![S]\!][\![L]\!]}}\left(\underline{\ell}^{n-1}\alpha LS\right\}\underline{\ell}^{n}\bar{\alpha}\bar{L}\bar{S}\Big)$$

$$\text{with } \xi = \begin{cases} 1 & \text{if } n \neq 6 \\ (-1)^{(\bar{\nu}-1)/2} & \text{if } n = 6 \end{cases}, \text{ and where } \bar{\boldsymbol{\nu}} \text{ is the seniority of } |\bar{\alpha}\bar{L}\bar{S}\rangle. \quad (15)$$

Under this relationship and phase convention, the matrix elements of operators pick up a global phase which depends on the rank of the operator, namely [NK63]:

$$\langle \underline{\mathrm{f}}^{14-n}\alpha SL\|\hat{U}^{(K)}\|\underline{\mathrm{f}}^{14-n}\alpha' S'L'\rangle = -(-1)^K\langle \underline{\mathrm{f}}^{n}\alpha SL\|\hat{U}^{(K)}\|\underline{\mathrm{f}}^{n}\alpha' S'L'\rangle \quad (16)$$

for a single tensor operator $\hat{U}^{(K)}$ of rank K, and

$$\langle \underline{\mathrm{f}}^{14-n}\alpha SL\|\hat{V}^{(1K)}\|\underline{\mathrm{f}}^{14-n}\alpha' S'L'\rangle = (-1)^K\langle \underline{\mathrm{f}}^{n}\alpha SL\|\hat{V}^{(1K)}\|\underline{\mathrm{f}}^{n}\alpha' S'L'\rangle \quad (17)$$

for a double tensor operator $\hat{V}^{(1K)}$ of rank 1 for spin and rank K for orbit.

2.5 Going beyond $\underline{\mathrm{f}}^7$

In most cases all matrix elements in **qlanth** are only calculated up to and including $\underline{\mathrm{f}}^7$. Beyond $\underline{\mathrm{f}}^7$ adequate changes of sign are enforced to take into account the equivalence that can be made between $\underline{\mathrm{f}}^n$ and $\underline{\mathrm{f}}^{14-n}$ as given by &qn-17 and &qn-16.

This is enforced when the function `HamMatrixAssembly` is called. In there `HoleElectronConjugation` is the function responsible for enforcing a global sign flip for the following operators (or alternatively, to their accompanying coefficients):

$$\begin{aligned} &\zeta, B_q^{(k)} \\ &T^{(2)\prime}, T^{(3)}, T^{(4)}, T^{(6)}, T^{(7)}, T^{(8)} \\ &T^{(11)\prime}, T^{(12)}, T^{(13)}, T^{(14)}, T^{(15)}, T^{(16)}, T^{(17)}, T^{(18)}, T^{(19)}, \end{aligned} \quad (18)$$

$T^{(2)}$ and $T^{(11)}$ must be treated separately since they have a part that changes sign, and another that doesn't.

```
HoleElectronConjugation::usage = "HoleElectronConjugation[params]
    takes the parameters (as an association) that define a
    configuration and converts them so that they may be interpreted as
     corresponding to a complentary hole configuration. Some of this
    can be simply done by changing the sign of the model parameters.
    In the case of the effective three body interaction the
    relationship is more complex and is controlled by the value of the
     t2Switch variable.";
HoleElectronConjugation[params_] := Module[
  {newparams = params},
  (
    flipSignsOf = Join[{ζ} , cfSymbols, TSymbols];
    flipped = Table[
```

```
      (
        flipper -> - newparams[flipper]
      ),
      {flipper, flipSignsOf}
      ];
    nonflipped = Table[
      (
        flipper -> newparams[flipper]
      ),
      {flipper, Complement[Keys[newparams], flipSignsOf]}
      ];
    flippedParams = Association[Join[nonflipped, flipped]];
    flippedParams = Select[flippedParams, FreeQ[#, Missing]&];
    Return[flippedParams];
  )
];
```

2.6 The J-J' block structure

Now that we know how the bases are ordered, we can already understand the structure of how the final Hamiltonian matrix representation in the $|LSJM_J\rangle$ basis is put together.

For a given configuration $\underline{\mathrm{f}}^n$ and for each term $\hat{h}$ in the Hamiltonian, **qlanth** first calculates the matrix elements $\langle \alpha LSJM_J|\hat{h}|\alpha' L'S'J'M'_J\rangle$ so that for each interaction an association with keys of the form $\{J, J'\}$ is created. The values being rectangular arrays.

Fig-5 shows roughly this block structure for $\underline{\mathrm{f}}^2$. In that figure, the shape of the rectangular blocks is determined by the fact that for $J = 0, 1, 2, 3, 4, 5, 6$ there are (2, 3, 15, 7, 27, 11, 26) corresponding basis states. As such, for example, the first row of blocks consists of blocks of size (2×2), (2×3), (2×15), (2×7), (2×27), (2×11), and (2×26).

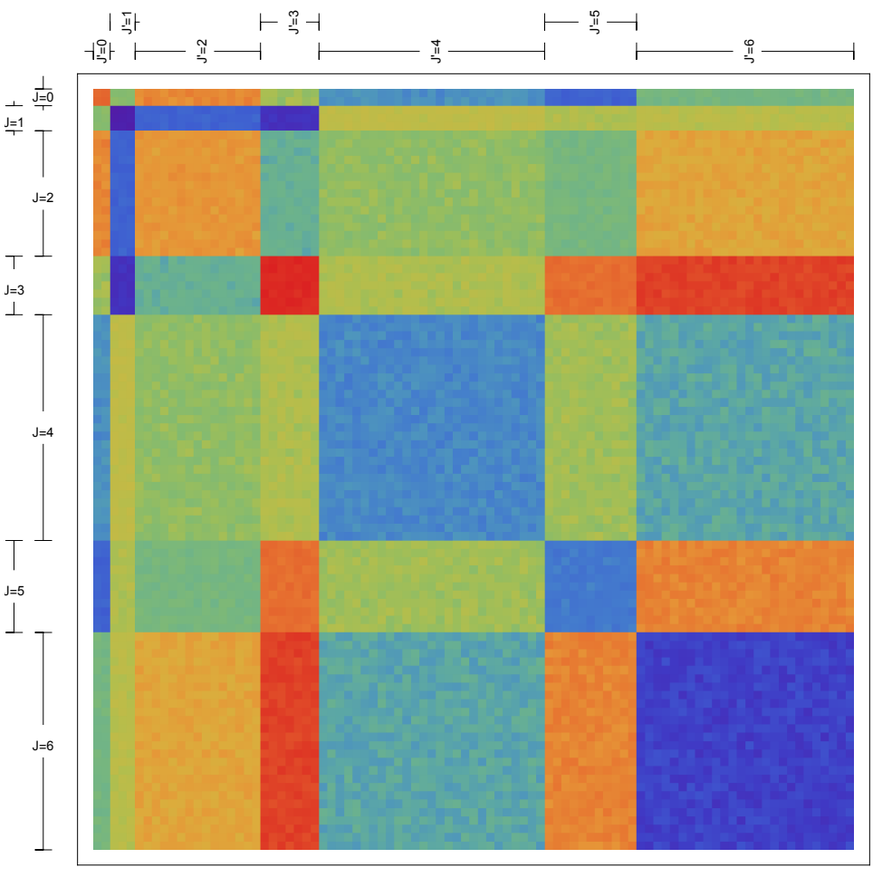


Figure 5: The J-J' block structure for $\underline{\mathrm{f}}^2$

In **qlanth** these blocks are put together by the function JJBlockMatrix which adds together the contributions from the different terms in the Hamiltonian.

```
JJBlockMatrix::usage = "For given J, J' in the f^n configuration
    JJBlockMatrix[numE, J, J'] determines all the SL S'L' terms that
    may contribute to them and using those it provides the matrix
    elements <J, LS | H | J', LS'>. H having contributions from the
    following interactions: Coulomb, spin-orbit, spin-other-orbit,
```

```
    electrostatically-correlated-spin-orbit, spin-spin, three-body
    interactions, and crystal-field.";
Options[JJBlockMatrix] = {"Sparse"->True, "ChenDeltas"->False};
JJBlockMatrix[numE_, J_, Jp_, CFTable_, OptionsPattern[]] := Module[
  {NKSLJMs, NKSLJMps, NKSLJM, NKSLJMp,
  SLterm, SpLpterm,
  MJ, MJp,
  subKron, matValue, eMatrix},
  (
    NKSLJMs  = AllowedNKSLJMforJTerms[numE, J];
    NKSLJMps = AllowedNKSLJMforJTerms[numE, Jp];
    eMatrix =
      Table[
        (*Condition for a scalar matrix op*)
        SLterm   = NKSLJM[[1]];
        SpLpterm = NKSLJMp[[1]];
        MJ       = NKSLJM[[3]];
        MJp      = NKSLJMp[[3]];
        subKron  = (
            KroneckerDelta[J, Jp] *
            KroneckerDelta[MJ, MJp]
                );
        matValue =
          If[subKron==0,
            0,
              (
                ElectrostaticTable[{numE, SLterm, SpLpterm}] +
                ElectrostaticConfigInteraction[{SLterm, SpLpterm}] +
                SpinOrbitTable[{numE, SLterm, SpLpterm, J}] +
                MagneticInteractions[{numE, SLterm, SpLpterm, J},
                  "ChenDeltas" -> OptionValue["ChenDeltas"]] +
                ThreeBodyTable[{numE, SLterm, SpLpterm}]
              )
          ];
        matValue += CFTable[{numE, SLterm, J, MJ, SpLpterm, Jp, MJp
    }];
        matValue,
      {NKSLJMp, NKSLJMps},
      {NKSLJM , NKSLJMs}
      ];
  If[OptionValue["Sparse"],
    eMatrix = SparseArray[eMatrix]
  ];
  Return[eMatrix]
)
];
```

Once these blocks have been calculated and saved to disk (in the folder `./hams/`) the function `HamMatrixAssembly` takes them, assembles the arrays in block form, and finally flattens them to provide a sparse rank-2 array. These are the arrays that are finally diagonalized to find energies and eigenstates. Through options, this function can also return the Hamiltonian in block form, which is useful for the level description of the eigenstates.

```
HamMatrixAssembly::usage = "HamMatrixAssembly[numE] returns the
    Hamiltonian matrix for the f^n_i configuration. The matrix is
    returned as a SparseArray.
The function admits an optional parameter ''FilenameAppendix'' which
    can be used to modify the filename to which the resulting array is
     exported to.
It also admits an optional parameter ''IncludeZeeman'' which can be
    used to include the Zeeman interaction. The default is False.
The option ''Set t2Switch'' can be used to toggle on or off setting
    the t2 selector automatically or not, the default is True, which
    replaces the parameter according to numE.
The option ''ReturnInBlocks'' can be used to return the matrix in
    block or flattened form. The default is to return it in flattened
    form.";
Options[HamMatrixAssembly] = {
      "FilenameAppendix" -> "",
      "IncludeZeeman"    -> False,
      "Set t2Switch"     -> True,
      "ReturnInBlocks"   -> False,
      "OperatorBasis"    -> "Legacy"};
HamMatrixAssembly[nf_, OptionsPattern[]] := Module[
  {numE, ii, jj, howManyJs, Js, blockHam, opBasis},
```

```
(
  (*####################################*)
  ImportFun = ImportMZip;
  opBasis = OptionValue["OperatorBasis"];
  If[Not[MemberQ[{"Legacy", "MostlyOrthogonal", "Orthogonal"},
  opBasis]],
    Print["Operator basis ", opBasis, " not recognized, using ''
  Legacy'' basis."];
    opBasis = "Legacy";
  ];
  If[opBasis == "Orthogonal",
    Print["Operator basis ''Orthogonal'' not implemented yet,
  aborting ..."];
    Return[Null];
  ];
  (*####################################*)
  If[opBasis == "MostlyOrthogonal",
    (
      blockHam = HamMatrixAssembly[nf,
      "OperatorBasis" -> "Legacy",
      "FilenameAppendix" -> OptionValue["FilenameAppendix"],
      "IncludeZeeman" -> OptionValue["IncludeZeeman"],
      "Set t2Switch" -> OptionValue["Set t2Switch"],
      "ReturnInBlocks" -> OptionValue["ReturnInBlocks"]];
      paramChanger = Which[
        nf < 7,
        <|
          F0 -> 1/91 (54 E1p+91 E0p+78 γp),
          F2 -> (15/392 *
            (
              140   E1p  +
              20020 E2p +
              1540  E3p  +
              770 αp -
              70 γp  +
              22 Sqrt[2] T2p -
              11 Sqrt[2] nf T2p t2Switch -
              11 Sqrt[2] (14-nf) T2p (1 - t2Switch)
            )
          ),
          F4 -> (99/490 *
            (
              70   E1p -
              9100 E2p +
              280  E3p +
              140 αp -
              35 γp  +
              4 Sqrt[2] T2p -
              2 Sqrt[2] nf T2p t2Switch -
              2 Sqrt[2] (14-nf) T2p (1-t2Switch)
            )
            ),
          F6 -> (5577/7000 *
            (
              20  E1p +
              700 E2p -
              140 E3p -
              70 αp -
              10 γp -
              2 Sqrt[2] T2p +
              Sqrt[2] nf T2p t2Switch +
              Sqrt[2] (14-nf) T2p (1-t2Switch)
            )
            ),
          ζ  -> ζ,
          α -> (5 αp)/4,
          β  -> -6  (5 αp + βp),
          γ -> 5/2 (2 βp + 5 γp),
          T2 -> 0
        |>,
      nf >= 7,
        <|
          F0 -> 1/91 (54 E1p+91 E0p+78 γp),
          F2 -> (15/392 *
            (
              140   E1p  +
```

```
87                20020 E2p +
88                1540  E3p  +
89                770 αp -
90                70 γp  +
91                22 Sqrt[2] T2p -
92                11 Sqrt[2] nf T2p
93              )
94            ),
95            F4 -> (99/490 *
96              (
97                70   E1p -
98                9100 E2p +
99                280  E3p +
100               140 αp -
101               35 γp  +
102               4 Sqrt[2] T2p -
103               2 Sqrt[2] nf T2p
104             )
105             ),
106           F6 -> (5577/7000 *
107             (
108               20  E1p +
109               700 E2p -
110               140 E3p -
111               70 αp -
112               10 γp -
113               2 Sqrt[2] T2p +
114               Sqrt[2] nf T2p
115             )
116             ),
117           ζ  -> ζ,
118           α -> (5 αp)/4,
119           β  -> -6  (5 αp + βp),
120           γ -> 5/2 (2 βp + 5 γp),
121           T2 -> 0
122         |>
123       ];
124       blockHamM0 = Which[
125         OptionValue["ReturnInBlocks"] == False,
126         ReplaceInSparseArray[blockHam, paramChanger],
127         OptionValue["ReturnInBlocks"] == True,
128         Map[ReplaceInSparseArray[#, paramChanger]&, blockHam, {2}]
129       ];
130       Return[blockHamM0];
131     )
132   ];
133   (*######################################*)
134
135   (*hole-particle equivalence enforcement*)
136   numE = nf;
137   allVars = {E0, E1, E2, E3, ζ, F0, F2, F4, F6, M0, M2, M4, T2, T2p
      ,
138     T3, T4, T6, T7, T8, P0, P2, P4, P6, gs,
139     α, β, γ, B02, B04, B06, B12, B14, B16,
140     B22, B24, B26, B34, B36, B44, B46, B56, B66, S12, S14, S16, S22
      ,
141     S24, S26, S34, S36, S44, S46, S56, S66, T11p, T12, T14, T15,
     T16,
142     T17, T18, T19, Bx, By, Bz};
143   params0 = AssociationThread[allVars, allVars];
144   If[nf > 7,
145     (
146       numE = 14 - nf;
147       params = HoleElectronConjugation[params0];
148       If[OptionValue["Set t2Switch"], params[t2Switch] = 0];
149     ),
150     params = params0;
151     If[OptionValue["Set t2Switch"], params[t2Switch] = 1];
152   ];
153   (* Load symbolic expressions for LS,J,J' energy sub-matrices. *)
154   emFname = JJBlockMatrixFileName[numE, "FilenameAppendix" ->
     OptionValue["FilenameAppendix"]];
155   JJBlockMatrixTable = ImportFun[emFname];
156   (*Patch together the entire matrix representation using J,J'
     blocks.*)
157   PrintTemporary["Patching JJ blocks ..."];
```

```
158     Js         = AllowedJ[numE];
159     howManyJs  = Length[Js];
160     blockHam   = ConstantArray[0, {howManyJs, howManyJs}];
161     Do[
162       blockHam[[jj, ii]] = JJBlockMatrixTable[{numE, Js[[ii]], Js[[jj
        ]]}];,
163     {ii, 1, howManyJs},
164     {jj, 1, howManyJs}
165     ];
166
167     (* Once the block form is created flatten it *)
168     If[Not[OptionValue["ReturnInBlocks"]],
169       (blockHam = ArrayFlatten[blockHam];
170       blockHam  = ReplaceInSparseArray[blockHam, params];
171       ),
172       (blockHam  = Map[ReplaceInSparseArray[#, params]&,blockHam
        ,{2}];)
173     ];
174
175     If[OptionValue["IncludeZeeman"],
176       (
177         PrintTemporary["Including Zeeman terms ..."];
178         {magx, magy, magz} = MagDipoleMatrixAssembly[numE, "
        ReturnInBlocks" -> OptionValue["ReturnInBlocks"]];
179         blockHam += - teslaToKayser * (Bx * magx + By * magy + Bz *
        magz);
180       )
181     ];
182     Return[blockHam];
183   )
184 ];
```

In **qlanth** the reduced matrix elements of all operators, and the subsequent matrix elements of $\hat{\mathcal{H}}$ are calculated exactly. This is in contrast to what is done in older alternatives to **qlanth** such as linuxemp, in which calculations of reduced matrix elements were obtained from tables calculated with finite precision. To underscore this fact, Eqn-2.6 shows an example of a J-J block as contained in **qlanth**.

2.7 Kramers' degeneracy

In the odd-electron cases, every energy is at least doubly degenerate. In **qlanth**, except in the case of the experimental data compiled for LaF_3, Kramers' degeneracy is given/expected explicitly.

3 Interactions

3.1 $\hat{\mathcal{H}}_{\mathrm{k}}$: kinetic energy

$$\hat{\mathcal{H}}_{\mathrm{k}} = -\frac{\hbar^2}{2m}\sum_{i=1}^{N}\nabla_i^2 \text{ (kinetic energy of N v-electrons)} \tag{20}$$

Since our description is limited to a single configuration, the kinetic energy simply contributes a constant energy shift, and since all we care about are energy differences, then this term can be omitted from the analysis.

To interpret the range of energies that result from diagonalizing the semi-empirical Hamiltonian, it might be instructive, however, to note that this term imparts an energy of about $10\,\mathrm{eV} \approx 10^6\mathcal{K}$[16] to each electron.

3.2 $\hat{\mathcal{H}}_{\mathrm{e:sn}}$: the central field potential

$$\hat{\mathcal{H}}_{\mathrm{e:sn}} = -e^2\sum_{i=1}^{Z}\frac{1}{r_i} + e^2\underbrace{\sum_{i=1}^{n}\sum_{j=1}^{Z-n}\frac{1}{r_{ij}}}_{\substack{\text{Repulsion between valence}\\ \text{and inner shell}\\ \text{electrons}}} \approx \sum_{i=1}^{n}V_{\mathrm{sn}}(r_i) \text{ (with Z = atomic No.)} \tag{21}$$

In principle, the sum over the Coulomb potential should extend over the nuclear charge and over all the electrons in the atom (not just the valence electrons). However, given the

[16] Note, (Kayser) $\mathcal{K} \equiv \mathrm{cm}^{-1}$, see section on units.

	$\vert^4\mathrm{F}_{1,-1}\rangle$	$\vert^4\mathrm{F}_{1,0}\rangle$	$\vert^4\mathrm{F}_{1,1}\rangle$
$\langle^4\mathrm{F}_{1,-1}\vert$	$2\alpha+\beta-\frac{B_0^{(2)}}{10}+\gamma$ $-\frac{\zeta}{2}+\frac{14F^{(0)}}{13}+\frac{43F^{(2)}}{195}+\frac{19F^{(4)}}{429}$ $-\frac{875F^{(6)}}{5577}+2m^{(0)}\sigma_{\mathrm{SS}}+\frac{61m^{(0)}}{12}$ $+4m^{(2)}\sigma_{\mathrm{SS}}+\frac{145m^{(2)}}{12}+\frac{50m^{(4)}\sigma_{\mathrm{SS}}}{11}$ $+\frac{1805m^{(4)}}{132}+\frac{43P^{(2)}}{1080}+\frac{19P^{(4)}}{2376}$ $-\frac{875P^{(6)}}{30888}$	$-\frac{\sqrt{3}B_1^{(2)}}{10}-\frac{1}{10}i\sqrt{3}S_2^{(2)}$	$-\frac{1}{5}\sqrt{\frac{3}{2}}B_2^{(2)}-\frac{1}{5}i\sqrt{\frac{3}{2}}S_2^{(2)}$
$\langle^4\mathrm{F}_{1,0}\vert$	$-\frac{\sqrt{3}B_1^{(2)}}{10}+\frac{1}{10}i\sqrt{3}S_2^{(2)}$	$2\alpha+\beta+\frac{B_0^{(2)}}{5}+\gamma$ $-\frac{\zeta}{2}+\frac{14F^{(0)}}{13}+\frac{43F^{(2)}}{195}+\frac{19F^{(4)}}{429}$ $-\frac{875F^{(6)}}{5577}+2m^{(0)}\sigma_{\mathrm{SS}}+\frac{61m^{(0)}}{12}$ $+4m^{(2)}\sigma_{\mathrm{SS}}+\frac{145m^{(2)}}{12}+\frac{50m^{(4)}\sigma_{\mathrm{SS}}}{11}$ $+\frac{1805m^{(4)}}{132}+\frac{43P^{(2)}}{1080}+\frac{19P^{(4)}}{2376}$ $-\frac{875P^{(6)}}{30888}$	$\frac{\sqrt{3}B_1^{(2)}}{10}+\frac{1}{10}i\sqrt{3}S_2^{(2)}$
$\langle^4\mathrm{F}_{1,1}\vert$	$-\frac{1}{5}\sqrt{\frac{3}{2}}B_2^{(2)}+\frac{1}{5}i\sqrt{\frac{3}{2}}S_2^{(2)}$	$\frac{\sqrt{3}B_1^{(2)}}{10}-\frac{1}{10}i\sqrt{3}S_2^{(2)}$	$2\alpha+\beta-\frac{B_0^{(2)}}{10}+\gamma$ $-\frac{\zeta}{2}+\frac{14F^{(0)}}{13}+\frac{43F^{(2)}}{195}+\frac{19F^{(4)}}{429}$ $-\frac{875F^{(6)}}{5577}+2m^{(0)}\sigma_{\mathrm{SS}}+\frac{61m^{(0)}}{12}$ $+4m^{(2)}\sigma_{\mathrm{SS}}+\frac{145m^{(2)}}{12}+\frac{50m^{(4)}\sigma_{\mathrm{SS}}}{11}$ $+\frac{1805m^{(4)}}{132}+\frac{43P^{(2)}}{1080}+\frac{19P^{(4)}}{2376}$ $-\frac{875P^{(6)}}{30888}$

(19)

shell structure of the atom, the lanthanide ions "see" the nuclear charge as shielded by a xenon core. Since every closed shell is a singlet, having spherical symmetry, these shields are like spherical shells surrounding the nucleus.

The precise form of $V_{\text{sn}}(r_i)$ is not of our concern here; all that matters is that we assume that it is spherically symmetric so that we can justify the separation of radial and angular parts of the wavefunctions.

3.3 $\hat{\mathcal{H}}_{\text{e:e}}$: e:e repulsion

$$\hat{\mathcal{H}}_{\text{e:e}} = \sum_{i>j}^{n,n} \frac{e^2}{\|\vec{r}_i - \vec{r}_j\|} = \sum_{k=0,2,4,6} F^{(k)} \hat{f}_k = \sum_{k=0,1,2,3} E_k \hat{e}_k \tag{22}$$

This term is the first we will not discard. Calculating this term for the $\underline{\text{f}}^n$ configurations was one of the contributions from Slater, as such the parameters we use to write it up are called *Slater integrals*. After the analysis from Slater, Giulio Racah contributed further to the analysis of this term [Rac49]. The insight that Racah had was that if in a given operator one identifies the parts in it that transform accordingly to the different symmetry groups present in the problem, then calculating the necessary matrix element in all $\underline{\text{f}}^n$ configurations can be greatly simplified.

The functions used in `qlanth` to compute these LS-reduced matrix elements [17] are `Electrostatic` and `fsubk`. In addition to these, the LS-reduced matrix elements of the tensor operators $\hat{C}^{(k)}$ and $\hat{U}^{(k)}$ are also needed. These functions are based in equations 12.16 and 12.17 from [Cow81] as specialized for the case of electrons belonging to a single $\underline{\text{f}}^n$ configuration. By default this term is computed in terms of $F^{(k)}$ Slater integrals, but it can also be computed in terms of the E_k Racah parameters, the functions `EtoF` and `FtoE` are useful for going from one representation to the other.

$$\langle \underline{\text{f}}^n \alpha^{2S+1}\text{L} \| \hat{\mathcal{H}}_{\text{e:e}} \| \underline{\text{f}}^n \alpha'^{2S'+1}\text{L'} \rangle = \sum_{k=0,2,4,6} {F^{(f)}}_k(n, \alpha LS, \alpha' L' S') \tag{23}$$

where

$$f_k(n, \alpha LS, \alpha' L' S') = \frac{1}{2}\delta(S,S')\delta(L,L')\langle \underline{\text{f}} \| \hat{C}^{(k)} \| \underline{\text{f}} \rangle^2 \times$$
$$\left\{ \frac{1}{2L+1} \sum_{\alpha'' L''} \langle \underline{\text{f}}^n \alpha'' L'' S \| \hat{U}^{(k)} \| \underline{\text{f}}^n \alpha LS \rangle \langle \underline{\text{f}}^n \alpha'' L'' S \| \hat{U}^{(k)} \| \underline{\text{f}}^n \alpha' LS \rangle - \delta(\alpha, \alpha') \frac{n\,(4\underline{\text{f}} + 2 - n)}{(2\underline{\text{f}}+1)(4\underline{\text{f}}+1)} \right\}. \tag{24}$$

```
Electrostatic::usage = "Electrostatic[{numE, NKSL, NKSLp}] returns
    the LS reduced matrix element for repulsion matrix element for
    equivalent electrons. See equation 2-79 in Wybourne (1965). The
    option ''Coefficients'' can be set to ''Slater'' or ''Racah''. If
    set to ''Racah'' then E_k parameters and e^k operators are assumed
    , otherwise the Slater integrals F^k and operators f_k. The
    default is ''Slater''.";
Options[Electrostatic] = {"Coefficients" -> "Slater"};
Electrostatic[{numE_, NKSL_, NKSLp_}, OptionsPattern[]] := Module[
  {fsub0, fsub2, fsub4, fsub6,
   esub0, esub1, esub2, esub3,
   fsup0, fsup2, fsup4, fsup6,
   eMatrixVal, orbital},
  (
    orbital = 3;
    Which[
      OptionValue["Coefficients"] == "Slater",
      (
        fsub0 = fsubk[numE, orbital, NKSL, NKSLp, 0];
        fsub2 = fsubk[numE, orbital, NKSL, NKSLp, 2];
        fsub4 = fsubk[numE, orbital, NKSL, NKSLp, 4];
        fsub6 = fsubk[numE, orbital, NKSL, NKSLp, 6];
        eMatrixVal = fsub0*F0 + fsub2*F2 + fsub4*F4 + fsub6*F6;
      ),
      OptionValue["Coefficients"] == "Racah",
      (
        fsup0 = fsupk[numE, orbital, NKSL, NKSLp, 0];
        fsup2 = fsupk[numE, orbital, NKSL, NKSLp, 2];
        fsup4 = fsupk[numE, orbital, NKSL, NKSLp, 4];
        fsup6 = fsupk[numE, orbital, NKSL, NKSLp, 6];
```

[17] An LS-reduced matrix element is ...

```
        esub0 = fsup0;
        esub1 = 9/7*fsup0 +   1/42*fsup2 +   1/77*fsup4 +  1/462*
    fsup6;
        esub2 =              143/42*fsup2 - 130/77*fsup4 + 35/462*
    fsup6;
        esub3 =               11/42*fsup2 + 4/77*fsup4   -  7/462*
    fsup6;
        eMatrixVal = esub0*E0 + esub1*E1 + esub2*E2 + esub3*E3;
      )
    ];
    Return[eMatrixVal];
  )
];
```

```
fsubk::usage = "fsubk[numE, orbital, SL, SLp, k] gives the Slater
    integral f_k for the given configuration and pair of SL terms. See
     equation 12.17 in TASS.";
fsubk[numE_, orbital_, NKSL_, NKSLp_, k_] := Module[
  {terms, S, L, Sp, Lp,
  termsWithSameSpin, SL,
  fsubkVal, spinMultiplicity,
  prefactor, summand1, summand2},
  (
    {S, L}   = FindSL[NKSL];
    {Sp, Lp} = FindSL[NKSLp];
    terms    = AllowedNKSLTerms[numE];
    (* sum for summand1 is over terms with same spin *)
    spinMultiplicity  = 2*S + 1;
    termsWithSameSpin = StringCases[terms, ToString[spinMultiplicity]
     ~~ __];
    termsWithSameSpin = Flatten[termsWithSameSpin];
    If[Not[{S, L} == {Sp, Lp}],
      Return[0]
    ];
    prefactor = 1/2 * Abs[Ck[orbital, k]]^2;
    summand1 = Sum[(
        ReducedUkTable[{numE, orbital, SL, NKSL,  k}] *
        ReducedUkTable[{numE, orbital, SL, NKSLp, k}]
        ),
      {SL, termsWithSameSpin}
    ];
    summand1 = 1 / TP0[L] * summand1;
    summand2 = (
      KroneckerDelta[NKSL, NKSLp] *
        (numE *(4*orbital + 2 - numE)) /
        ((2*orbital + 1) * (4*orbital + 1))
      );
    fsubkVal = prefactor*(summand1 - summand2);
    Return[fsubkVal];
  )
];
```

```
EtoF::usage = "EtoF[E0, E1, E2, E3] calculates the Slater integral
    parameters {F0, F2, F4, F6} corresponding to the given Racah
    parameters {E0, E1, E2, E3}. This is the inverse of the FtoE
    function.";
EtoF[E0_, E1_, E2_, E3_] := Module[
  {F0, F2, F4, F6},
  (
    F0 = 1/7      (7 E0 + 9 E1);
    F2 = 75/14    (E1 + 143 E2 + 11 E3);
    F4 = 99/7     (E1 - 130 E2 + 4 E3);
    F6 = 5577/350 (E1 + 35 E2 - 7 E3);
    Return[{F0, F2, F4, F6}];
  )
];
```

```
FtoE::usage = "FtoE[F0, F2, F4, F6] calculates the Racah parameters {
    E0, E1, E2, E3} corresponding to the given Slater integrals.
See eqn. 2-80 in Wybourne.
Note that in that equation the subscripted Slater integrals are used
    but since this function assumes the the input values are
    superscripted Slater integrals, it is necessary to convert them
    using Dk.";
FtoE[F0_, F2_, F4_, F6_] := Module[
```

```
  {E0, E1, E2, E3},
  (
    E0 = (F0 - 10*F2/Dk[2] - 33*F4/Dk[4] - 286*F6/Dk[6]);
    E1 = (70*F2/Dk[2] + 231*F4/Dk[4] + 2002*F6/Dk[6])/9;
    E2 = (F2/Dk[2] - 3*F4/Dk[4] + 7*F6/Dk[6])/9;
    E3 = (5*F2/Dk[2] + 6*F4/Dk[4] - 91*F6/Dk[6])/3;
    Return[{E0, E1, E2, E3}];
  )
];
```

3.4 $\hat{\mathcal{H}}_{\text{s:o}}$: spin-orbit

The spin-orbit interaction arises from the interaction of the magnetic moment of the electron and the magnetic field that its orbital motion generates. In terms of the central potential $V_{\text{s:n}}$, the spin-orbit term for a single electron is

$$\hat{h}_{\text{s:o}} = \frac{\hbar^2}{2m_e^2c^2}\left(\frac{1}{r}\frac{\mathrm{d}V_{\text{s:n}}}{\mathrm{d}r}\right)\hat{l}\cdot\hat{s} \coloneqq \zeta(r)\hat{l}\cdot\hat{s}. \tag{25}$$

Adding this term for all the n valence electrons, and replacing $\zeta(r)$ by its radial average ζ then gives

$$\hat{\mathcal{H}}_{\text{s:o}} = \zeta\sum_i^n \hat{l}_i\cdot\hat{s}_i. \tag{26}$$

From equations 2-106 to 2-109 in Wybourne [Wyb63] the matrix elements we need are given by

$$\begin{aligned}\langle\alpha LSJM_J|\hat{\mathcal{H}}_{\text{s:o}}|\alpha' L'S'J'M_{J'}\rangle &= \zeta\delta(J,J')\delta(M_J,M_{J'})\langle\alpha LSJM_J|\sum_i^n \hat{l}_i\cdot\hat{s}_i|\alpha' L'S'JM_J\rangle\\ &= \zeta\delta(J,J')\delta(M_J,M_{J'})(-1)^{J+L+S'}\begin{Bmatrix}L & L' & 1\\ S' & S & J\end{Bmatrix}\langle\alpha LS|\sum_i^n \hat{l}_i\cdot\hat{s}_i|\alpha' L'S'\rangle\\ = \zeta\delta(J,J')\delta(M_J,M_{J'})(-1)^{J+L+S'}&\begin{Bmatrix}L & L' & 1\\ S' & S & J\end{Bmatrix}\sqrt{\underline{\ell}(\underline{\ell}+1)(2\underline{\ell}+1)}\langle\alpha LS\|\hat{V}^{(11)}\|\alpha' L'S'\rangle,\end{aligned} \tag{27}$$

where $\hat{V}^{(11)}$ is a double tensor operator of rank one over spin and orbital parts defined as

$$\hat{V}^{(11)} = \sum_{i=1}^n\left(\hat{s}\hat{u}^{(1)}\right)_i, \tag{28}$$

in which the rank on the spin operator $\hat{s}$ has been omitted, and the rank of the orbital tensor operator given explicitly as 1.

In **qlanth** the reduced matrix elements for this double tensor operator are calculated by `ReducedV1k` and stored in a static association called `ReducedV1kTable`. The reduced matrix elements of this operator are calculated using equation 2-101 from Wybourne [Wyb65]:

$$\begin{aligned}\langle\underline{\ell}^n\psi\|\hat{V}^{(1k)}\|\underline{\ell}^n\psi'\rangle = \langle\underline{\ell}^n\alpha LS\|\hat{V}^{(1k)}\|\underline{\ell}^n\alpha' L'S'\rangle = n\sqrt{\underline{\jmath}(\underline{\jmath}+1)(2\underline{\jmath}+1)}\sqrt{[S][L][S'][L']}\times\\ \sum_{\bar{\psi}}(-1)^{\bar{S}+\bar{L}+S+L+\underline{\ell}+\underline{\jmath}+k+1}\left(\psi\left\{\left|\bar{\psi}\right.\right)\left(\left.\bar{\psi}\right\}\right|\psi'\right)\begin{Bmatrix}S & S' & 1\\ \underline{\jmath} & \underline{\jmath} & \bar{S}\end{Bmatrix}\begin{Bmatrix}L & L' & k\\ \underline{\ell} & \underline{\ell} & \bar{L}\end{Bmatrix}\end{aligned} \tag{29}$$

In this expression the sum over $\bar{\psi}$ depends on (ψ,ψ') and is over all the states in $\underline{\ell}^{n-1}$ which are common parents to both ψ and ψ'. Also note that in the equation above, since our concern are $\underline{\text{f}}$-electron configurations, we have $\underline{\ell}=3$ and $\underline{\jmath}=\frac{1}{2}$.

```
ReducedV1k::usage = "ReducedV1k[n, l, SL, SpLp, k] gives the reduced
    matrix element of the spherical tensor operator V^(1k). See
    equation 2-101 in Wybourne 1965.";
ReducedV1k[numE_, SL_, SpLp_, k_] := Module[
  {Vk1, S, L, Sp, Lp,
  Sb, Lb, spin, orbital,
  cfpSL, cfpSpLp,
  SLparents, SpLpparents,
  commonParents, prefactor},
  (
    {spin, orbital} = {1/2, 3};
```

```
    {S, L}         = FindSL[SL];
    {Sp, Lp}       = FindSL[SpLp];
    cfpSL          = CFP[{numE, SL}];
    cfpSpLp        = CFP[{numE, SpLp}];
    SLparents      = First /@ Rest[cfpSL];
    SpLpparents    = First /@ Rest[cfpSpLp];
    commonParents = Intersection[SLparents, SpLpparents];
    Vk1 = Sum[(
        {Sb, Lb} = FindSL[\[Psi]b];
        Phaser[(Sb + Lb + S + L + orbital + k - spin)] *
        CFPAssoc[{numE, SL, \[Psi]b}] *
        CFPAssoc[{numE, SpLp, \[Psi]b}] *
        SixJay[{S, Sp, 1}, {spin, spin, Sb}] *
        SixJay[{L, Lp, k}, {orbital, orbital, Lb}]
      ),
    {\[Psi]b, commonParents}
    ];
    prefactor = numE * Sqrt[spin * (spin + 1) * TPO[spin, S, L, Sp, Lp] ];
    Return[prefactor * Vk1];
  )
];
```

These reduced matrix elements are then used by the function `SpinOrbit`.

```
SpinOrbit::usage = "SpinOrbit[numE, SL, SpLp, J] returns the LSJ reduced matrix element ζ <SL, J|L.S|SpLp, J>. These are given as a function of ζ. This function requires that the association ReducedV1kTable be defined.
See equations 2-106 and 2-109 in Wybourne (1965). Equivalently see eqn. 12.43 in TASS.";
SpinOrbit[numE_, SL_, SpLp_, J_] := Module[
  {S, L, Sp, Lp, orbital, sign, prefactor, val},
  (
    orbital    = 3;
    {S, L}     = FindSL[SL];
    {Sp, Lp}   = FindSL[SpLp];
    prefactor = Sqrt[orbital * (orbital+1) * (2*orbital+1)] *
                SixJay[{L, Lp, 1}, {Sp, S, J}];
    sign       = Phaser[J + L + Sp];
    val        = sign * prefactor * ζ * ReducedV1kTable[{numE, SL, SpLp, 1}];
    Return[val];
  )
];
```

3.5 $\hat{\mathcal{H}}_{\mathcal{SO}(3)}, \hat{\mathcal{H}}_{\mathcal{G}_2}, \hat{\mathcal{H}}_{\mathcal{SO}(7)}$: electrostatic configuration interaction

These are the first terms where we take into account the contributions from *configuration-interaction*. Rajnak and Wybourne [RW63] showed that *configuration-interaction* of the electrostatic interactions corresponding to two-electron excitations from $\underline{f}^n$ can be represented through the Casimir operators of the groups $\mathcal{SO}(3)$, $\mathcal{G}_2$, and $\mathcal{SO}(7)$. This borrowed from an earlier insight of Trees [Tre52], who realized that an addition of a term proportional to $L(L+1)$ improved the energy calculations for the second spectrum of manganese (Mn-II) and the third spectrum of iron (Fe-III).

One of these Casimir operators is the familiar $\hat{L}^2$ from $\mathcal{SO}(3)$. In analogy to $\hat{L}^2$ in which the quantum number L can be used to determine the eigenvalues, in the cases of $\hat{\mathcal{H}}_{\mathcal{G}_2}$ the necessary state label is the U label of the LS term, and in the case of $\hat{\mathcal{H}}_{\mathcal{SO}(7)}$ the necessary label is W. If $\Lambda_{\mathcal{G}_2}(U)$ is used to note the eigenvalue of the Casimir operator of $\mathcal{G}_2$ corresponding to label U, and $\Lambda_{\mathcal{SO}(7)}(W)$ the eigenvalue corresponding to state label W, then the matrix elements of $\hat{\mathcal{H}}_{\mathcal{SO}(3)}$, $\hat{\mathcal{H}}_{\mathcal{G}_2}$ and $\hat{\mathcal{H}}_{\mathcal{SO}(7)}$ are diagonal in all quantum numbers (see Rajnak and Wybourne [RW63]) and are given by

$$\langle \underline{\ell}^n \alpha S L J M_J | \hat{\mathcal{H}}_{\mathcal{SO}(3)} | \underline{\ell}^n \alpha' S' L' J' M'_J \rangle = \alpha \delta(\alpha S L J M_J, \alpha' S' L' J' M'_J) L(L+1) \tag{30}$$

$$\langle \underline{\ell}^n U \alpha S L J M_J | \hat{\mathcal{H}}_{\mathcal{G}_2} | \underline{\ell}^n U \alpha' S' L' J' M'_J \rangle = \beta \delta(\alpha S L J M_J, \alpha' S' L' J' M'_J) \Lambda_{\mathcal{G}_2}(U) \tag{31}$$

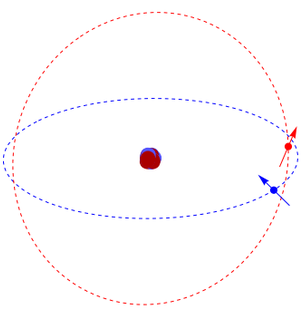
$$\langle \underline{\ell}^n W \alpha S L J M_J | \hat{\mathcal{H}}_{\mathcal{SO}(7)} | \underline{\ell}^n W \alpha' S' L' J' M_J \rangle = \gamma \delta(\alpha S L J M_J, \alpha' S' L' J' M'_J) \Lambda_{\mathcal{SO}(7)}(W) \tag{32}$$

In **qlanth** the role of $\Lambda_{\mathcal{SO}(7)}(W)$ is played by the function `GSO7W`, the role of $\Lambda_{\mathcal{G}_2}(U)$ by `GG2U`, and the role of $\Lambda_{\mathcal{SO}(3)}(L)$ by `CasimirSO3`. These are used by `CasimirG2`, `CasimirSO3`, and `CasimirSO7` which find the corresponding U, W, L labels to the LS terms provided to them. Finally, the function `ElectrostaticConfigInteraction` puts them together.

```
ElectrostaticConfigInteraction::usage = "
    ElectrostaticConfigInteraction[{SL, SpLp}] returns the matrix
    element for configuration interaction as approximated by the
    Casimir operators of the groups R3, G2, and R7. SL and SpLp are
    strings that represent terms under LS coupling.";
ElectrostaticConfigInteraction[{SL_, SpLp_}] := Module[
  {S, L, val},
  (
    {S, L} = FindSL[SL];
    val = (
      If[SL == SpLp,
        CasimirSO3[{SL, SL}] +
        CasimirSO7[{SL, SL}] +
        CasimirG2[{SL, SL}],
        0
      ]
      );
    ElectrostaticConfigInteraction[{S, L}] = val;
    Return[val];
  )
];
```

3.6 $\hat{\mathcal{H}}_{\text{s:s-s:oo}}$: *spin-spin* and *spin-other-orbit*

The calculation of the $\hat{\mathcal{H}}_{\text{s:s-s:oo}}$ is qualitatively different from the previous ones. The previous ones were self-contained in the sense that the reduced matrix elements that we require we also computed on our own. In the case of the interactions that follow from here, we use values from literature for reduced matrix elements either in $\underline{f}^2$ or in $\underline{f}^3$ and then we "pull" them up for all $\underline{f}^n$ configurations with help of the coefficients of fractional parentage.



The analysis of *spin-other-orbit*, and the *spin-spin* contributions used in **qlanth** is that of Judd, Crosswhite, and Crosswhite [JCC68]. Much as the spin-orbit effect can be extracted from the Dirac equation as a relativistic correction, the multi-electron spin-orbit effects can be derived from the Breit operator $\hat{\mathcal{H}}_B$ [BS57] which is a term added to the relativistic description of a many-particle system in order to account for retardation of the electromagnetic field

$$\hat{\mathcal{H}}_B = -\frac{1}{2}e^2 \sum_{i>j} \left[(\alpha_i \cdot \alpha_j) \frac{1}{r_{ij}} + (\alpha_i \cdot \vec{r}_{ij}) (\alpha_j \cdot \vec{r}_{ij}) \frac{1}{r_{ij}^3} \right]. \tag{33}$$

When this operator is expanded in powers of v/c, a number of non-relativistic inter-electron interactions result. Two of them are the *spin-other-orbit* and *spin-spin* interactions. As usual, the radial part of the Hamiltonian is averaged, which in this case gives appearance to the Marvin integrals

$$m^{(k)} := \frac{e^2\hbar^2}{8m^2c^2} \langle (nl)^2 | \frac{r_<^k}{r_>^{k+3}} | (nl)^2 \rangle. \tag{34}$$

With these, the expression for the *spin-spin* term within the single configuration description is [JCC68]

$$\hat{\mathcal{H}}_{s:s} = -2 \sum_{i\neq j} \sum_k m^{(k)} \sqrt{(k+1)(k+2)(2k+3)} \langle \underline{\ell} \| C^{(k)} \| \underline{\ell} \rangle \langle \underline{\ell} \| C^{(k+2)} \| \underline{\ell} \rangle \left\{ \hat{w}_i^{(1,k)} \hat{w}_j^{(1,k+2)} \right\}^{(2,2)0} \tag{35}$$

and the one for *spin-other-orbit*

$$\begin{aligned} \hat{\mathcal{H}}_{s:oo} = \sum_{i\neq j} \sum_k & \sqrt{(k+1)(2\underline{\ell}+k+2)(2\underline{\ell}-k)} \times \\ & \Big[\left\{ \hat{w}_i^{(0,k+1)} \hat{w}_j^{(1,k)} \right\}^{(11)0} \left\{ m^{(k-1)} \langle \underline{\ell} \| C^{(k+1)} \| \underline{\ell} \rangle^2 + 2 m^{(k)} \langle \underline{\ell} \| C^{(k)} \| \underline{\ell} \rangle^2 \right\} + \\ & \left\{ \hat{w}_i^{(0,k)} \hat{w}_j^{(1,k+1)} \right\}^{(11)0} \left\{ m^{(k)} \langle \underline{\ell} \| C^{(k)} \| \underline{\ell} \rangle^2 + 2 m^{(k-1)} \langle \underline{\ell} \| C^{(k+1)} \| \underline{\ell} \rangle^2 \right\} \Big]. \end{aligned} \tag{36}$$

In the expressions above $\hat{w}_i^{(\kappa,k)}$ is a double tensor operator of rank κ over spin, of rank k over orbit, and acting on electron i. It is defined by its reduced matrix elements as

$$\langle \underline{\ell} \| \hat{w}^{(\kappa,k)} \| \underline{\ell} \rangle = \sqrt{[\![\kappa]\!][\![k]\!]}. \tag{37}$$

The explicit complexity of the above expressions can be somewhat reduced by identifying them with the scalar part of two new double tensors $\hat{\mathcal{T}}_0^{(11)}$ and $\hat{\mathcal{T}}_0^{(22)}$ such that

$$\sqrt{5}\hat{\mathcal{T}}_0^{(22)} \coloneqq \hat{\mathcal{H}}_{\text{s:s}} \tag{38}$$

$$-\sqrt{3}\hat{\mathcal{T}}_0^{(11)} \coloneqq \hat{\mathcal{H}}_{\text{s:oo}}. \tag{39}$$

In terms of which the reduced matrix elements in the $|LSJ\rangle$ basis can be obtained by

$$\langle \gamma SLJ | \hat{\mathcal{H}} | \gamma' S' L' J' \rangle = \delta(J, J') \begin{Bmatrix} S' & L' & J \\ L & S & t \end{Bmatrix} \langle \gamma SL \| \hat{\mathcal{T}}^{(tt)} \| \gamma' S' L' \rangle. \tag{40}$$

This above relationship is the one effectively used in **qlanth** in the functions SpinSpin and SOOandECSO.

```
SpinSpin::usage = "SpinSpin[n, SL, SpLp, J] returns the matrix
    element <|SL,J|spin-spin|SpLp,J|> for the spin-spin operator
    within the configuration f^n. This matrix element is independent
    of MJ. This is obtained by querying the relevant reduced matrix
    element from the association T22Table, putting in the adequate
    phase, and 6-j symbol.
This is calculated according to equation (3) in ''Judd, BR, HM
    Crosswhite, and Hannah Crosswhite. ''Intra-Atomic Magnetic
    Interactions for f Electrons.'' Physical Review 169, no. 1 (1968):
     130.''
''.
";
SpinSpin[numE_, SL_, SpLp_, J_] := Module[
  {S, L, Sp, Lp, α, val},
  (
    α = 2;
    {S, L} = FindSL[SL];
    {Sp, Lp} = FindSL[SpLp];
    val = (
            Phaser[Sp + L + J] *
            SixJay[{Sp, Lp, J}, {L, S, α}] *
            T22Table[{numE, SL, SpLp}]
          );
    Return[val]
  )
];
```

```
SOOandECSO::usage = "SOOandECSO[n, SL, SpLp, J] returns the matrix
    element <|SL,J|spin-spin|SpLp,J|> for the combined effects of the
    spin-other-orbit interaction and the electrostatically-correlated-
    spin-orbit (which originates from configuration interaction
    effects) within the configuration f^n. This matrix element is
    independent of MJ. This is obtained by querying the relevant
    reduced matrix element by querying the association
    SOOandECSOLSTable and putting in the adequate phase and 6-j symbol
    . The SOOandECSOLSTable puts together the reduced matrix elements
    from three operators.
This is calculated according to equation (3) in ''Judd, BR, HM
    Crosswhite, and Hannah Crosswhite. ''Intra-Atomic Magnetic
    Interactions for f Electrons.'' Physical Review 169, no. 1 (1968):
     130.''.
";
SOOandECSO[numE_, SL_, SpLp_, J_] := Module[
  {S, Sp, L, Lp, α, val},
  (
    α = 1;
    {S, L}   = FindSL[SL];
    {Sp, Lp} = FindSL[SpLp];
    val = (
            Phaser[Sp + L + J] *
            SixJay[{Sp, Lp, J}, {L, S, α}] *
            SOOandECSOLSTable[{numE, SL, SpLp}]
          );
    Return[val];
  )
];
```

For two-electron operators such as these, the matrix elements in $\underline{\mathrm{f}}^n$ are related to those in $\underline{\mathrm{f}}^{n-1}$ by equation 4 in Judd *et al.* [JCC68]

$$\langle \underline{\mathrm{f}}^n \psi \| \hat{\mathcal{T}}^{(tt)} \| \underline{\mathrm{f}}^n \psi' \rangle = \frac{n}{n-2} \sum_{\bar{\psi},\bar{\psi}'} (-1)^{\bar{S}+\bar{L}+\underline{s}+\underline{\ell}+S'+L'} \sqrt{[\![S]\!][\![S']\!][\![L]\!][\![L']\!]} \times \left(\psi \{\!| \bar{\psi} \right) \left(\psi' \{\!| \bar{\psi}' \right) \begin{Bmatrix} S & t & S' \\ \bar{S}' & \underline{s} & \bar{S} \end{Bmatrix} \begin{Bmatrix} L & t & L' \\ \bar{L}' & \underline{\ell} & \bar{L} \end{Bmatrix} \langle \underline{\mathrm{f}}^{n-1} \bar{\psi} \| \hat{\mathcal{T}}^{(tt)} \| \underline{\mathrm{f}}^{n-1}] \bar{\psi}' \rangle, \quad (41)$$

where the sum runs over the terms $\bar{\psi}$ and $\bar{\psi}'$ in $\underline{\mathrm{f}}^{n-1}$ which are parents common to ψ and ψ'. Using these the matrix elements of $\hat{\mathcal{T}}^{(11)}$ and $\hat{\mathcal{T}}^{(22)}$ in $\underline{\mathrm{f}}^2$ can be used to compute all the reduced matrix elements in $\underline{\mathrm{f}}^n$. These could then be used together with &qn-40 to obtain the matrix elements of $\hat{\mathcal{H}}_{\mathrm{s:s}}$ and $\hat{\mathcal{H}}_{\mathrm{s:oo}}$. This is done for $\hat{\mathcal{H}}_{\mathrm{s:s}}$, but not for $\hat{\mathcal{H}}_{\mathrm{s:oo}}$, because this term is traditionally computed (with a slight modification) at the same time as the electrostically-correlated-spin-orbit (see next section).

These equations are implemented in **qlanth** through the following functions: `GenerateT22Table`, `ReducedT22infn`, `ReducedT22inf2`, `ReducedT11inf2`. Where `ReducedT22inf2` and `ReducedT11inf2` provide the reduced matrix elements for $\hat{\mathcal{T}}^{(11)}$ and $\hat{\mathcal{T}}^{(22)}$ in $\underline{\mathrm{f}}^2$ as provided in table II of [JCC68].

```
GenerateT22Table::usage = "GenerateT22Table[nmax] generates the LS
    reduced matrix elements for the double tensor operator T22 in f^n
    up to n=nmax. If the option ''Export'' is set to true then the
    resulting association is saved to the data folder. The values for
    n=1 and n=2 are taken from ''Judd, BR, HM Crosswhite, and Hannah
    Crosswhite. ''Intra-Atomic Magnetic Interactions for f Electrons
    .'' Physical Review 169, no. 1 (1968): 130.'', and the values for
    n>2 are calculated recursively using equation (4) of that same
    paper.
This is an intermediate step to the calculation of the reduced matrix
     elements of the spin-spin operator.";
Options[GenerateT22Table] = {"Export" -> True, "Progress" -> True};
GenerateT22Table[nmax_Integer, OptionsPattern[]] := (
  If[And[OptionValue["Progress"], frontEndAvailable],
    (
      numItersai = Association[Table[numE->Length[AllowedNKSLTerms[
    numE]]^2, {numE, 1, nmax}]];
      counters = Association[Table[numE->0, {numE, 1, nmax}]];
      totalIters = Total[Values[numItersai[[1;;nmax]]]];
      template1 = StringTemplate["Iteration `numiter` of `totaliter`"
    ];
      template2 = StringTemplate["`remtime` min remaining"];template3
     = StringTemplate["Iteration speed = `speed` ms/it"];
      template4 = StringTemplate["Time elapsed = `runtime` min"];
      progBar = PrintTemporary[
        Dynamic[
          Pane[
            Grid[{{Superscript["f", numE]},
                  {template1[<|"numiter"->numiter, "totaliter"->
    totalIters|>]},
                  {template4[<|"runtime"->Round[QuantityMagnitude[
    UnitConvert[(Now-startTime), "min"]], 0.1]|>]},
                  {template2[<|"remtime"->Round[QuantityMagnitude[
    UnitConvert[(Now-startTime)/(numiter)*(totalIters-numiter), "min"
    ]], 0.1]|>]},
                  {template3[<|"speed"->Round[QuantityMagnitude[Now-
    startTime, "ms"]/(numiter), 0.01]|>]},
                  {ProgressIndicator[Dynamic[numiter], {1, totalIters
    }]}},
                  Frame->All],
                  Full,
                  Alignment->Center]
              ]
            ];
    )
  ];
  T22Table = <||>;
  startTime = Now;
  numiter = 1;
  Do[
    (
      numiter+= 1;
      T22Table[{numE, SL, SpLp}] = Which[
        numE==1,
```

```
        0,
        numE==2,
        SimplifyFun[ReducedT22inf2[SL, SpLp]],
        True,
        SimplifyFun[ReducedT22infn[numE,  SL, SpLp]]
      ];
    ),
  {numE, 1, nmax},
  {SL,   AllowedNKSLTerms[numE]},
  {SpLp, AllowedNKSLTerms[numE]}
  ];
  If[And[OptionValue["Progress"],frontEndAvailable],
    NotebookDelete[progBar]
  ];
  If[OptionValue["Export"],
    (
      fname = FileNameJoin[{moduleDir, "data", "ReducedT22Table.m"}];
      Export[fname, T22Table];
    )
  ];
  Return[T22Table];
);
```

```
ReducedT22infn::usage = "ReducedT22infn[n, SL, SpLp] calculates the
    reduced matrix element of the T22 operator for the f^n
    configuration corresponding to the terms SL and SpLp. This is the
    operator corresponding to the inter-electron between spin.
It does this by using equation (4) of ''Judd, BR, HM Crosswhite, and
    Hannah Crosswhite. ''Intra-Atomic Magnetic Interactions for f
    Electrons.'' Physical Review 169, no. 1 (1968): 130.''
";
ReducedT22infn[numE_, SL_, SpLp_] := Module[
  {spin, orbital, t, idx1, idx2, S, L,
  Sp, Lp, cfpSL, cfpSpLp, parentSL,
  parentSpLp, Sb, Lb, Tnkk, phase, Sbp, Lbp},
  (
    {spin, orbital} = {1/2, 3};
    {S, L}   = FindSL[SL];
    {Sp, Lp} = FindSL[SpLp];
    t = 2;
    cfpSL    = CFP[{numE, SL}];
    cfpSpLp  = CFP[{numE, SpLp}];
    Tnkk = Sum[(
      parentSL   = cfpSL[[idx2, 1]];
      parentSpLp = cfpSpLp[[idx1, 1]];
      {Sb, Lb}   = FindSL[parentSL];
      {Sbp, Lbp} = FindSL[parentSpLp];
      phase = Phaser[Sb + Lb + spin + orbital + Sp + Lp];
      (
        phase *
        cfpSpLp[[idx1, 2]] * cfpSL[[idx2, 2]] *
        SixJay[{S, t, Sp}, {Sbp, spin, Sb}] *
        SixJay[{L, t, Lp}, {Lbp, orbital, Lb}] *
        T22Table[{numE - 1, parentSL, parentSpLp}]
      )
    ),
    {idx1, 2, Length[cfpSpLp]},
    {idx2, 2, Length[cfpSL]}
    ];
    Tnkk *= numE / (numE - 2) * Sqrt[TPO[S,Sp,L,Lp]];
    Return[Tnkk];
  )
];
```

```
ReducedT22inf2::usage = "ReducedT22inf2[SL, SpLp] returns the reduced
     matrix element of the scalar component of the double tensor T22
    for the terms SL, SpLp in f^2.
Data used here for m0, m2, m4 is from Table I of Judd, BR, HM
    Crosswhite, and Hannah Crosswhite. Intra-Atomic Magnetic
    Interactions for f Electrons. Physical Review 169, no. 1 (1968):
    130.
";
ReducedT22inf2[SL_, SpLp_] := Module[
  {statePosition, PsiPsipStates, m0, m2, m4, Tkk2m},
  (
    T22inf2 = <|
```

```
    {"3P", "3P"} -> -12 M0 - 24 M2 - 300/11 M4,
    {"3P", "3F"} -> 8/Sqrt[3] (3 M0 + M2 - 100/11 M4),
    {"3F", "3F"} -> 4/3 Sqrt[14] (-M0 + 8 M2 - 200/11 M4),
    {"3F", "3H"} -> 8/3 Sqrt[11/2] (2 M0 - 23/11 M2 - 325/121 M4),
    {"3H", "3H"} -> 4/3 Sqrt[143] (M0 - 34/11 M2 - (1325/1573) M4)
    |>;
    Which[
      MemberQ[Keys[T22inf2],{SL,SpLp}],
        Return[T22inf2[{SL,SpLp}]],
      MemberQ[Keys[T22inf2],{SpLp,SL}],
        Return[T22inf2[{SpLp,SL}]],
      True,
        Return[0]
    ]
  )
];
```

```
Reducedt11inf2::usage = "Reducedt11inf2[SL, SpLp] returns the reduced
    matrix element in f^2 of the double tensor operator t11 for the
    corresponding given terms {SL, SpLp}.
Values given here are those from Table VII of ''Judd, BR, HM
    Crosswhite, and Hannah Crosswhite. ''Intra-Atomic Magnetic
    Interactions for f Electrons.'' Physical Review 169, no. 1 (1968):
    130.''
";
Reducedt11inf2[SL_, SpLp_] := Module[
  {t11inf2},
  (
    t11inf2 = <|
      {"1S", "3P"} -> -2 P0 - 105 P2 - 231 P4 - 429 P6,
      {"3P", "3P"} -> -P0 - 45 P2 - 33 P4 + 1287 P6,
      {"3P", "1D"} -> Sqrt[15/2] (P0 + 32 P2 - 33 P4 - 286 P6),
      {"1D", "3F"} -> Sqrt[10] (-P0 - 9/2 P2 + 66 P4 - 429/2 P6),
      {"3F", "3F"} -> Sqrt[14] (-P0 + 10 P2 + 33 P4 + 286 P6),
      {"3F", "1G"} -> Sqrt[11] (P0 - 20 P2 + 32 P4 - 104 P6),
      {"1G", "3H"} -> Sqrt[10] (-P0 + 55/2 P2 - 23 P4 - 65/2 P6),
      {"3H", "3H"} -> Sqrt[55] (-P0 + 25 P2 + 51 P4 + 13 P6),
      {"3H", "1I"} -> Sqrt[13/2] (P0 - 21 P4 - 6 P6)
      |>;
    Which[
      MemberQ[Keys[t11inf2],{SL,SpLp}],
        Return[t11inf2[{SL,SpLp}]],
      MemberQ[Keys[t11inf2],{SpLp,SL}],
        Return[t11inf2[{SpLp,SL}]],
      True,
        Return[0]
    ]
  )
];
```

3.7 $\hat{\mathcal{H}}_{\text{ecs:o}}$: *electrostatically-correlated-spin-orbit*

In the same paper [JCC68] that describes the *spin-spin* and *spin-other-orbit* interactions, consideration is also given to the emergence of additional corrections due to configuration interaction as described by the following operator (which is what results from the application of perturbation theory to *second* order) (page. 134 of [JCC68])

$$\hat{\mathcal{H}}_{\text{ci}} = -\sum_{\chi}\sum_{i}\frac{1}{E_\chi}\xi(r_i)\left(\hat{\underline{s}}_i\cdot\hat{\underline{\ell}}_i\right)|\chi\rangle\langle\chi|\hat{\mathfrak{C}} - \frac{1}{E_\chi}\hat{\mathfrak{C}}|\chi\rangle\langle\chi|\xi(r_i)\left(\hat{\underline{s}}_i\cdot\hat{\underline{\ell}}_i\right) \tag{42}$$

where $\xi(r_h)(\hat{\underline{s}}_h\cdot\hat{\underline{\ell}}_h)$ is the customary spin-orbit interaction, E_χ is the energy of state $|\chi\rangle$, i is a label for the valence electrons, $\hat{\mathfrak{C}}$ stands for the Coulomb interaction, and $|\chi\rangle$ are states in the configurations with which one is "interacting". Since this term includes both the electrostatic term and the spin-orbit one, this is called the *electrostatically-correlated-spin-orbit* interaction.

This operator can be identified with the scalar component of a double tensor operator of rank 1 both for the spin and orbital parts of the wavefunction

$$\hat{\mathcal{H}}_{\text{ci}} = -\sqrt{3}\,\hat{t}_0^{(11)}. \tag{43}$$

Judd *et al.* [JCC68] then go on to list the reduced matrix elements of this operator in the $\underline{\text{f}}^2$ configuration. When this is done the Marvin integrals $\mathcal{M}^{(k)}$ appear again, but a

second set of parameters, the *pseudo-magnetic* parameters $P^{(k)}$, is also necessary

$$P^{(k)} = 6\sum_{f'} \frac{\zeta_{ff'}}{E_{ff'}} R^{(k)}(ff, ff') \text{ for } k = 0, 2, 4, 6. \tag{44}$$

Where f stands for an $\underline{\text{f}}$-electron radial eigenfunction, and f' similarly but for a configuration different from $\underline{\text{f}}^n$. And where

$$\zeta_{ff'} \coloneqq \langle f|\xi(r)|f'\rangle \tag{45}$$

$$R^{(k)}(ff, ff') \coloneqq e^2\langle f_1 f_2|\frac{r_<^k}{r_>^{k+1}}|f_1 f_2'\rangle. \tag{46}$$

In the semi-empirical approach embodied by **qlanth**, calculating these quantities *ab-initio* is not the objective, they are instead to be defined from experiments. Nonetheless, not only these expressions give theoretical justification to the model, but they also serve to justify the ratios between different orders of these quantities, their relative importance, or their sign. Consequently, both the set of three $m^{(k)}$ and the set of $P^{(k)}$ ultimately rely on a single free parameter each. Such parsimony is desirable given the large number of parameters (about 20) that the Hamiltonian ends up having.

Judd *et al.* further note that $P^{(0)}$ is proportional to the spin orbit operator, and as such its effect is absorbed by the standard spin-orbit parameter ζ. They also developed an alternative approach based on group theory arguments. They put together the *spin-other-orbit* and the *electrostatically-correlated-spin-orbit* as a sum of operators $\hat{z}_i$ with useful transformation rules

$$\langle\psi\|\hat{\mathcal{T}}^{(11)} + \hat{t}^{(11)}\|\psi'\rangle = \sum a_i\langle\psi\|\hat{z}_i\|\psi'\rangle. \tag{47}$$

At this stage a subtle point needs to be raised. As Judd points out, in the sum above, the term $\hat{z}_{13}$ that contributes with a tensorial character equal to that of the regular spin-orbit operator. As such, if the goal is obtaining a parametric Hamiltonian that can be fit with uncorrelated parameters, it is then necessary to subtract this part from $\hat{\mathcal{T}}^{(11)} + \hat{t}^{(11)}$. This point was clarified by Chen *et al.* [Che+08]. Because of this, the final form of the operator contributing both to *spin-other-orbit* and the *electrostatically-correlated-spin-orbit* is

$$\hat{\mathcal{H}}_{\text{s:oo}} + \hat{\mathcal{H}}_{\text{ecs:o}} = \hat{\mathcal{T}}^{(11)} + \hat{t}^{(11)} - \frac{1}{6}a_{13}\hat{z}_{13} \tag{48}$$

where

$$a_{13} = -33m^{(0)} + 3m^{(2)} + \frac{15}{11}m^{(4)} - 6P^{(0)} + \frac{3}{2}\left(\frac{35}{225}P^{(2)} + \frac{77}{1089}P^{(4)} + \frac{25}{1287}P^{(6)}\right). \tag{49}$$

In **qlanth** the contributions from *spin-spin*, *spin-other-orbit*, and *electrostatically-correlated-spin-orbit* are put together by the function `MagneticInteractions`. That function queries precomputed values from two associations `SpinSpinTable` and `SOOandECSOTable`. In turn these two associations are generated by the functions `GenerateSpinOrbitTable` and `GenerateSOOandECSOTable`. Note that both *spin-spin* and *spin-other-orbit* end up contributing through $m^{(k)}$, however there doesn't seem to be consensus about adding them together, as such **qlanth** allows including or excluding the *spin-spin* contribution, this is done with a control parameter σ_{SS} (1 for including, 0 for excluding).

```
1 MagneticInteractions::usage = "MagneticInteractions[{numE, SL, SLp, J
    }] returns the matrix element of the magnetic interaction between
    the terms SL and SLp in the f^numE configuration for the given
    value of J. The interaction is given by the sum of the spin-spin,
    the spin-other-orbit,  and the electrostatically-correlated-spin-
    orbit interactions.
2 The part corresponding to the spin-spin interaction is provided by
    SpinSpin[{numE, SL, SLp, J}].
3 The part corresponding to SOO and ECSO is provided by the function
    SOOandECSO[{numE, SL, SLp, J}].
4 The option ''ChenDeltas'' can be used to include or exclude the Chen
    deltas from the calculation. The default is to exclude them. If
    this option is used, then the chenDeltas association needs to be
    loaded into the session with LoadChen[].";
5 Options[MagneticInteractions] = {"ChenDeltas" -> False};
6 MagneticInteractions[{numE_, SL_, SLp_, J_}, OptionsPattern[]] :=
    Module[
7   {key, ss, sooandecso, total,
```

```
  S, L, Sp, Lp, phase, sixjay,
  M0v, M2v, M4v,
  P2v, P4v, P6v},
  (
    key         = {numE, SL, SLp, J};
    ss          = \[Sigma]SS * SpinSpinTable[key];
    sooandecso = SOOandECSOTable[key];
    total = ss + sooandecso;
    total = SimplifyFun[total];
    If[
      Not[OptionValue["ChenDeltas"]],
      Return[total]
    ];
    (* In the type A errors the wrong values are different *)
    If[MemberQ[Keys[chenDeltas["A"]], {numE, SL, SLp}],
      (
        {S, L}   = FindSL[SL];
        {Sp, Lp} = FindSL[SLp];
        phase    = Phaser[Sp + L + J];
        sixjay   = SixJay[{Sp, Lp, J}, {L, S, 1}];
        {M0v, M2v, M4v, P2v, P4v, P6v} = chenDeltas["A"][{numE, SL,
    SLp}]["wrong"];
        total    = (
                       phase * sixjay *
                       (
                         M0v*M0 + M2v*M2 + M4v*M4 +
                         P2v*P2 + P4v*P4 + P6v*P6
                       )
                   );
        total    = wChErrA * total + (1 - wChErrA) * (ss + sooandecso
    )
      )
    ];
    (* In the type B errors the wrong values are zeros all around *)
    If[MemberQ[chenDeltas["B"], {numE, SL, SLp}],
      (
        total  = (1 - wChErrB) * (ss + sooandecso)
      )
    ];
    Return[total];
  )
];
```

```
GenerateSpinOrbitTable::usage = "GenerateSpinOrbitTable[nmax]
    computes the matrix elements for the spin-orbit interaction for f^
    n configurations up to n = nmax. The function returns an
    association whose keys are lists of the form {n, SL, SpLp, J}. If
    export is set to True, then the result is exported to the data
    subfolder for the folder in which this package is in. It requires
    ReducedV1kTable to be defined.";
Options[GenerateSpinOrbitTable] = {"Export" -> True};
GenerateSpinOrbitTable[nmax_Integer:7, OptionsPattern[]] := Module[
  {numE, J, SL, SpLp, exportFname},
  (
    SpinOrbitTable =
      Table[
        {numE, SL, SpLp, J} -> SpinOrbit[numE, SL, SpLp, J],
      {numE, 1, nmax},
      {J, MinJ[numE], MaxJ[numE]},
      {SL,   Map[First, AllowedNKSLforJTerms[numE, J]]},
      {SpLp, Map[First, AllowedNKSLforJTerms[numE, J]]}
      ];
    SpinOrbitTable = Association[SpinOrbitTable];

    exportFname = FileNameJoin[{moduleDir, "data", "SpinOrbitTable.m"
    }];
    If[OptionValue["Export"],
      (
        Print["Exporting to file "<>ToString[exportFname]];
        Export[exportFname, SpinOrbitTable];
      )
    ];
    Return[SpinOrbitTable];
  )
];
```

```
GenerateSOOandECSOTable::usage = "GenerateSOOandECSOTable[nmax]
    generates the reduced matrix elements in the |LSJ> basis for the (
    spin-other-orbit + electrostatically-correlated-spin-orbit)
    operator. It returns an association where the keys are of the form
     {n, SL, SpLp, J}. If the option ''Export'' is set to True then
    the resulting object is saved to the data folder. Since this is a
    scalar operator, there is no MJ dependence. This dependence only
    comes into play when the crystal field contribution is taken into
    account.";
Options[GenerateSOOandECSOTable] = {"Export"->False};
GenerateSOOandECSOTable[nmax_, OptionsPattern[]] := (
  SOOandECSOTable = <||>;
  Do[
    SOOandECSOTable[{numE, SL, SpLp, J}] = (SOOandECSO[numE, SL, SpLp
    , J] /. Prescaling);,
  {numE, 1, nmax},
  {J, MinJ[numE], MaxJ[numE]},
  {SL,   First /@ AllowedNKSLforJTerms[numE, J]},
  {SpLp, First /@ AllowedNKSLforJTerms[numE, J]}
  ];
  If[OptionValue["Export"],
  (
    fname = FileNameJoin[{moduleDir, "data", "SOOandECSOTable.m"}];
    Export[fname, SOOandECSOTable];
  )
  ];
  Return[SOOandECSOTable];
);
```

The function GenerateSpinSpinTable calls the function SpinSpin over all possible combinations of the arguments $\{n, SL, S'L', J\}$. In turn the function SpinSpin queries the precomputed values of the double tensor $\hat{\mathcal{T}}^{(22)}$ which are stored in the association T22Table.

```
GenerateSpinSpinTable::usage = "GenerateSpinSpinTable[nmax] generates
     the reduced matrix elements in the |LSJ> basis for the (spin-
    other-orbit + electrostatically-correlated-spin-orbit) operator.
    It returns an association where the keys are of the form {numE, SL
    , SpLp, J}. If the option ''Export'' is set to True then the
    resulting object is saved to the data folder. Since this is a
    scalar operator, there is no MJ dependence. This dependence only
    comes into play when the crystal field contribution is taken into
    account.";
Options[GenerateSpinSpinTable] = {"Export"->False};
GenerateSpinSpinTable[nmax_, OptionsPattern[]] :=
  (
    SpinSpinTable = <||>;
    PrintTemporary[Dynamic[numE]];
    Do[
      SpinSpinTable[{numE, SL, SpLp, J}] = (SpinSpin[numE, SL, SpLp,
    J]);,
    {numE, 1, nmax},
    {J, MinJ[numE], MaxJ[numE]},
    {SL,   First /@ AllowedNKSLforJTerms[numE, J]},
    {SpLp, First /@ AllowedNKSLforJTerms[numE, J]}
    ];
    If[OptionValue["Export"],
    (fname = FileNameJoin[{moduleDir, "data", "SpinSpinTable.m"}];
      Export[fname, SpinSpinTable];
      )
    ];
    Return[SpinSpinTable];
  );
```

```
SpinSpin::usage = "SpinSpin[n, SL, SpLp, J] returns the matrix
    element <|SL,J|spin-spin|SpLp,J|> for the spin-spin operator
    within the configuration f^n. This matrix element is independent
    of MJ. This is obtained by querying the relevant reduced matrix
    element from the association T22Table, putting in the adequate
    phase, and 6-j symbol.
This is calculated according to equation (3) in ''Judd, BR, HM
    Crosswhite, and Hannah Crosswhite. ''Intra-Atomic Magnetic
    Interactions for f Electrons.'' Physical Review 169, no. 1 (1968):
     130.''
''.
```

```
";
SpinSpin[numE_, SL_, SpLp_, J_] := Module[
  {S, L, Sp, Lp, α, val},
  (
    α = 2;
    {S, L} = FindSL[SL];
    {Sp, Lp} = FindSL[SpLp];
    val = (
            Phaser[Sp + L + J] *
            SixJay[{Sp, Lp, J}, {L, S, α}] *
            T22Table[{numE, SL, SpLp}]
          );
    Return[val]
  )
];
```

The association `T22Table` is computed by the function `GenerateT22Table`. This function populates `T22Table` with keys of the form $\{n, SL, S'L'\}$. It does this by using the function `ReducedT22inf2` in the base case of $\underline{f}^2$, and `ReducedT22infn` for configurations above $\underline{f}^2$. When `ReducedT22infn` is called, the sum in &qn-41 is carried out using $t = 2$. When `ReducedT22inf2` is called, the reduced matrix elements from [JCC68] are used.

```
GenerateT22Table::usage = "GenerateT22Table[nmax] generates the LS
    reduced matrix elements for the double tensor operator T22 in f^n
    up to n=nmax. If the option ''Export'' is set to true then the
    resulting association is saved to the data folder. The values for
    n=1 and n=2 are taken from ''Judd, BR, HM Crosswhite, and Hannah
    Crosswhite. ''Intra-Atomic Magnetic Interactions for f Electrons
    .'' Physical Review 169, no. 1 (1968): 130.'', and the values for
    n>2 are calculated recursively using equation (4) of that same
    paper.
This is an intermediate step to the calculation of the reduced matrix
     elements of the spin-spin operator.";
Options[GenerateT22Table] = {"Export" -> True, "Progress" -> True};
GenerateT22Table[nmax_Integer, OptionsPattern[]] := (
  If[And[OptionValue["Progress"], frontEndAvailable],
    (
      numItersai = Association[Table[numE->Length[AllowedNKSLTerms[
    numE]]^2, {numE, 1, nmax}]];
      counters = Association[Table[numE->0, {numE, 1, nmax}]];
      totalIters = Total[Values[numItersai[[1;;nmax]]]];
      template1 = StringTemplate["Iteration `numiter` of `totaliter`"
    ];
      template2 = StringTemplate["`remtime` min remaining"];template3
     = StringTemplate["Iteration speed = `speed` ms/it"];
      template4 = StringTemplate["Time elapsed = `runtime` min"];
      progBar = PrintTemporary[
        Dynamic[
          Pane[
            Grid[{{Superscript["f", numE]},
                  {template1[<|"numiter"->numiter, "totaliter"->
    totalIters|>]},
                  {template4[<|"runtime"->Round[QuantityMagnitude[
    UnitConvert[(Now-startTime), "min"]], 0.1]|>]},
                  {template2[<|"remtime"->Round[QuantityMagnitude[
    UnitConvert[(Now-startTime)/(numiter)*(totalIters-numiter), "min"
    ]], 0.1]|>]},
                  {template3[<|"speed"->Round[QuantityMagnitude[Now-
    startTime, "ms"]/(numiter), 0.01]|>]},
                  {ProgressIndicator[Dynamic[numiter], {1, totalIters
    }]}},
                  Frame->All],
                  Full,
                  Alignment->Center]
              ]
            ];
    )
  ];
  T22Table = <||>;
  startTime = Now;
  numiter = 1;
  Do[
    (
      numiter+= 1;
      T22Table[{numE, SL, SpLp}] = Which[
        numE==1,
```

```
        0,
        numE==2,
        SimplifyFun[ReducedT22inf2[SL, SpLp]],
        True,
        SimplifyFun[ReducedT22infn[numE,  SL, SpLp]]
      ];
    ),
  {numE, 1, nmax},
  {SL,   AllowedNKSLTerms[numE]},
  {SpLp, AllowedNKSLTerms[numE]}
  ];
  If[And[OptionValue["Progress"],frontEndAvailable],
    NotebookDelete[progBar]
  ];
  If[OptionValue["Export"],
    (
      fname = FileNameJoin[{moduleDir, "data", "ReducedT22Table.m"}];
      Export[fname, T22Table];
    )
  ];
  Return[T22Table];
);
```

```
ReducedT22infn::usage = "ReducedT22infn[n, SL, SpLp] calculates the
    reduced matrix element of the T22 operator for the f^n
    configuration corresponding to the terms SL and SpLp. This is the
    operator corresponding to the inter-electron between spin.
It does this by using equation (4) of ''Judd, BR, HM Crosswhite, and
    Hannah Crosswhite. ''Intra-Atomic Magnetic Interactions for f
    Electrons.'' Physical Review 169, no. 1 (1968): 130.''
";
ReducedT22infn[numE_, SL_, SpLp_] := Module[
  {spin, orbital, t, idx1, idx2, S, L,
  Sp, Lp, cfpSL, cfpSpLp, parentSL,
  parentSpLp, Sb, Lb, Tnkk, phase, Sbp, Lbp},
  (
    {spin, orbital} = {1/2, 3};
    {S, L}   = FindSL[SL];
    {Sp, Lp} = FindSL[SpLp];
    t = 2;
    cfpSL    = CFP[{numE, SL}];
    cfpSpLp  = CFP[{numE, SpLp}];
    Tnkk = Sum[(
      parentSL   = cfpSL[[idx2, 1]];
      parentSpLp = cfpSpLp[[idx1, 1]];
      {Sb, Lb}   = FindSL[parentSL];
      {Sbp, Lbp} = FindSL[parentSpLp];
      phase = Phaser[Sb + Lb + spin + orbital + Sp + Lp];
      (
        phase *
        cfpSpLp[[idx1, 2]] * cfpSL[[idx2, 2]] *
        SixJay[{S, t, Sp}, {Sbp, spin, Sb}] *
        SixJay[{L, t, Lp}, {Lbp, orbital, Lb}] *
        T22Table[{numE - 1, parentSL, parentSpLp}]
      )
    ),
    {idx1, 2, Length[cfpSpLp]},
    {idx2, 2, Length[cfpSL]}
    ];
    Tnkk *= numE / (numE - 2) * Sqrt[TPO[S,Sp,L,Lp]];
    Return[Tnkk];
  )
];
```

```
ReducedT22inf2::usage = "ReducedT22inf2[SL, SpLp] returns the reduced
     matrix element of the scalar component of the double tensor T22
    for the terms SL, SpLp in f^2.
Data used here for m0, m2, m4 is from Table I of Judd, BR, HM
    Crosswhite, and Hannah Crosswhite. Intra-Atomic Magnetic
    Interactions for f Electrons. Physical Review 169, no. 1 (1968):
    130.
";
ReducedT22inf2[SL_, SpLp_] := Module[
  {statePosition, PsiPsipStates, m0, m2, m4, Tkk2m},
  (
    T22inf2 = <|
```

```
    {"3P", "3P"} -> -12 M0 - 24 M2 - 300/11 M4,
    {"3P", "3F"} -> 8/Sqrt[3] (3 M0 + M2 - 100/11 M4),
    {"3F", "3F"} -> 4/3 Sqrt[14] (-M0 + 8 M2 - 200/11 M4),
    {"3F", "3H"} -> 8/3 Sqrt[11/2] (2 M0 - 23/11 M2 - 325/121 M4),
    {"3H", "3H"} -> 4/3 Sqrt[143] (M0 - 34/11 M2 - (1325/1573) M4)
    |>;
    Which[
      MemberQ[Keys[T22inf2],{SL,SpLp}],
        Return[T22inf2[{SL,SpLp}]],
      MemberQ[Keys[T22inf2],{SpLp,SL}],
        Return[T22inf2[{SpLp,SL}]],
      True,
        Return[0]
    ]
  )
];
```

The function GenerateSOOandECSOTable calls the function SOOandECSO over all possible combinations of the arguments $\{n, SL, S'L', J\}$ and uses their values to populate the association SOOandECSOTable. In turn the function SOOandECSO queries the precomputed values of &qn-48 as stored in the association SOOandECSOLSTable.

```
GenerateSOOandECSOTable::usage = "GenerateSOOandECSOTable[nmax]
    generates the reduced matrix elements in the |LSJ> basis for the (
    spin-other-orbit + electrostatically-correlated-spin-orbit)
    operator. It returns an association where the keys are of the form
     {n, SL, SpLp, J}. If the option ''Export'' is set to True then
    the resulting object is saved to the data folder. Since this is a
    scalar operator, there is no MJ dependence. This dependence only
    comes into play when the crystal field contribution is taken into
    account.";
Options[GenerateSOOandECSOTable] = {"Export"->False};
GenerateSOOandECSOTable[nmax_, OptionsPattern[]] := (
  SOOandECSOTable = <||>;
  Do[
    SOOandECSOTable[{numE, SL, SpLp, J}] = (SOOandECSO[numE, SL, SpLp
    , J] /. Prescaling);,
  {numE, 1, nmax},
  {J, MinJ[numE], MaxJ[numE]},
  {SL,   First /@ AllowedNKSLforJTerms[numE, J]},
  {SpLp, First /@ AllowedNKSLforJTerms[numE, J]}
  ];
  If[OptionValue["Export"],
  (
    fname = FileNameJoin[{moduleDir, "data", "SOOandECSOTable.m"}];
    Export[fname, SOOandECSOTable];
  )
  ];
  Return[SOOandECSOTable];
);
```

```
SOOandECSO::usage = "SOOandECSO[n, SL, SpLp, J] returns the matrix
    element <|SL,J|spin-spin|SpLp,J|> for the combined effects of the
    spin-other-orbit interaction and the electrostatically-correlated-
    spin-orbit (which originates from configuration interaction
    effects) within the configuration f^n. This matrix element is
    independent of MJ. This is obtained by querying the relevant
    reduced matrix element by querying the association
    SOOandECSOLSTable and putting in the adequate phase and 6-j symbol
    . The SOOandECSOLSTable puts together the reduced matrix elements
    from three operators.
This is calculated according to equation (3) in ''Judd, BR, HM
    Crosswhite, and Hannah Crosswhite. ''Intra-Atomic Magnetic
    Interactions for f Electrons.'' Physical Review 169, no. 1 (1968):
     130.''.
";
SOOandECSO[numE_, SL_, SpLp_, J_] := Module[
  {S, Sp, L, Lp, α, val},
  (
    α = 1;
    {S, L}   = FindSL[SL];
    {Sp, Lp} = FindSL[SpLp];
    val = (
            Phaser[Sp + L + J] *
            SixJay[{Sp, Lp, J}, {L, S, α}] *
            SOOandECSOLSTable[{numE, SL, SpLp}]
```

```
          );
    Return[val];
  )
];
```

```
SOOandECSO::usage = "SOOandECSO[n, SL, SpLp, J] returns the matrix
    element <|SL,J|spin-spin|SpLp,J|> for the combined effects of the
    spin-other-orbit interaction and the electrostatically-correlated-
    spin-orbit (which originates from configuration interaction
    effects) within the configuration f^n. This matrix element is
    independent of MJ. This is obtained by querying the relevant
    reduced matrix element by querying the association
    SOOandECSOLSTable and putting in the adequate phase and 6-j symbol
    . The SOOandECSOLSTable puts together the reduced matrix elements
    from three operators.
This is calculated according to equation (3) in ''Judd, BR, HM
    Crosswhite, and Hannah Crosswhite. ''Intra-Atomic Magnetic
    Interactions for f Electrons.'' Physical Review 169, no. 1 (1968):
     130.''.
";
SOOandECSO[numE_, SL_, SpLp_, J_] := Module[
  {S, Sp, L, Lp, α, val},
  (
    α = 1;
    {S, L}   = FindSL[SL];
    {Sp, Lp} = FindSL[SpLp];
    val = (
            Phaser[Sp + L + J] *
            SixJay[{Sp, Lp, J}, {L, S, α}] *
            SOOandECSOLSTable[{numE, SL, SpLp}]
          );
    Return[val];
  )
];
```

The association `SOOandECSOLSTable` is computed by the function `GenerateSOOandECSOLSTable`. This function populates `SOOandECSOLSTable` with keys of the form $\{n, SL, S'L'\}$. It does this by using the function `ReducedSOOandECSOinf2` in the base case of $\underline{f}^2$, and `ReducedSOOandECSOinfn` for configurations above $\underline{f}^2$. When `ReducedSOOandECSOinfn` is called the sum in &qn-41 is carried out using $t = 1$. When `ReducedSOOandECSOinf2` is called the reduced matrix elements from [JCC68] are used.

```
ReducedSOOandECSOinfn::usage = "ReducedSOOandECSOinfn[numE, SL, SpLp]
    calculates the reduced matrix elements of the (spin-other-orbit +
    ECSO) operator for the f^numE configuration corresponding to the
    terms SL and SpLp. This is done recursively, starting from
    tabulated values for f^2 from ''Judd, BR, HM Crosswhite, and
    Hannah Crosswhite. ''Intra-Atomic Magnetic Interactions for f
    Electrons.'' Physical Review 169, no. 1 (1968): 130.'', and by
    using equation (4) of that same paper.
";
ReducedSOOandECSOinfn[numE_, SL_, SpLp_] := Module[
  {spin, orbital, t, S, L, Sp, Lp,
  idx1, idx2, cfpSL, cfpSpLp, parentSL,
  Sb, Lb, Sbp, Lbp, parentSpLp, funval},
  (
    {spin, orbital} = {1/2, 3};
    {S, L}   = FindSL[SL];
    {Sp, Lp} = FindSL[SpLp];
    t = 1;
    cfpSL    = CFP[{numE, SL}];
    cfpSpLp  = CFP[{numE, SpLp}];
    funval   = Sum[
      (
        parentSL = cfpSL[[idx2, 1]];
        parentSpLp = cfpSpLp[[idx1, 1]];
        {Sb, Lb}   = FindSL[parentSL];
        {Sbp, Lbp} = FindSL[parentSpLp];
        phase = Phaser[Sb + Lb + spin + orbital + Sp + Lp];
        (
          phase *
          cfpSpLp[[idx1, 2]] * cfpSL[[idx2, 2]] *
          SixJay[{S, t, Sp}, {Sbp, spin, Sb}] *
          SixJay[{L, t, Lp}, {Lbp, orbital, Lb}] *
          SOOandECSOLSTable[{numE - 1, parentSL, parentSpLp}]
```

```
        )
      ),
    {idx1, 2, Length[cfpSpLp]},
    {idx2, 2, Length[cfpSL]}
    ];
    funval *= numE / (numE - 2) * Sqrt[TPO[S, Sp, L, Lp]];
    Return[funval];
  )
];
```

```
ReducedSOOandECSOinf2::usage = "ReducedSOOandECSOinf2[SL, SpLp]
    returns the reduced matrix element corresponding to the operator (
    T11 + t11 - a13 * z13 / 6) for the terms {SL, SpLp}. This
    combination of operators corresponds to the spin-other-orbit plus
    ECSO interaction.
The T11 operator corresponds to the spin-other-orbit interaction, and
     the t11 operator (associated with electrostatically-correlated
    spin-orbit) originates from configuration interaction analysis. To
     their sum a factor proportional to the operator z13 is subtracted
     since its effect is redundant to the spin-orbit interaction. The
    factor of 1/6 is not on Judd's 1968 paper, but it is on ''Chen,
    Xueyuan, Guokui Liu, Jean Margerie, and Michael F Reid. ''A Few
    Mistakes in Widely Used Data Files for Fn Configurations
    Calculations.'' Journal of Luminescence 128, no. 3 (2008):
    421-27''.
The values for the reduced matrix elements of z13 are obtained from
    Table IX of the same paper. The value for a13 is from table VIII.
Rigurously speaking the Pk parameters here are subscripted. The
    conversion to superscripted parameters is performed elsewhere with
     the Prescaling replacement rules.
";
ReducedSOOandECSOinf2[SL_, SpLp_]  := Module[
  {a13, z13, z13inf2, matElement, redSOOandECSOinf2},
  (
    a13 = (-33 M0 + 3 M2 + 15/11 M4 -
          6 P0 + 3/2 (35 P2 + 77 P4 + 143 P6));
    z13inf2 = <|
        {"1S","3P"} -> 2,
        {"3P","3P"} -> 1,
        {"3P","1D"} -> -Sqrt[(15/2)],
        {"1D","3F"} -> Sqrt[10],
        {"3F","3F"} -> Sqrt[14],
        {"3F","1G"} -> -Sqrt[11],
        {"1G","3H"} -> Sqrt[10],
        {"3H","3H"} -> Sqrt[55],
        {"3H","1I"} -> -Sqrt[(13/2)]
        |>;
    matElement = Which[
      MemberQ[Keys[z13inf2],{SL,SpLp}],
        z13inf2[{SL,SpLp}],
      MemberQ[Keys[z13inf2],{SpLp,SL}],
        z13inf2[{SpLp,SL}],
      True,
        0
    ];
    redSOOandECSOinf2 = (
        ReducedT11inf2[SL, SpLp] +
        Reducedt11inf2[SL, SpLp] -
        a13 / 6 * matElement
    );
    redSOOandECSOinf2 = SimplifyFun[redSOOandECSOinf2];
    Return[redSOOandECSOinf2];
  )
];
```

3.8 $\hat{\mathcal{H}}_{\curlywedge}$: three-body effective operators

The three-body operators in the semi-empirical Hamiltonian are due to the *configuration-interaction* effects of the Coulomb repulsion. More specifically, they originate from configuration interaction between the ground configuration $(4\underline{f})^n$ and single electron excitations to the $(4\underline{f})^{n\pm1}(n'\underline{\ell}')^{\mp1}$ configurations.

The operators that can be used to span the resulting effects were initially studied by Wybourne and Rajnak in 1963 [RW63], their analysis was complemented soon after by

Judd [Jud66], and revisited again by Judd in 1984 [JS84].

This model interaction is spanned by a set of 14 $\hat{t}_i$ of operators ($\hat{t}$ from *t*hree)

$$\hat{\mathcal{H}}_{\curlywedge} = T'^{(2)}\hat{t}_2{}' + T'^{(11)}\hat{t}_{11}{}' \sum_{\substack{k=2,3,4,6,7,8,\\ 11,12,14,15,\\ 16,17,18,19}} T^{(k)}\hat{t}_k, \tag{50}$$

where $\hat{t}_2$ and $\hat{t}_{11}$ are operators that have orthogonal alternatives represented by $\hat{t}_2{}'$ and $\hat{t}_{11}{}'$ (see [JS84]). **qlanth** includes the legacy operator $\hat{t}_2$ since it was used for important work during and before the 1980s.

The omission of some indices in this sum has to do with the fact that the way in which these are defined in terms of their index (see [Jud66]) gives rise to two-body operators which can be absorbed by the two-body terms in the Hamiltonian. As such, it is not so much that they are not included, but rather that their effects are considered to be accounted for elsewhere. This is representative of a common feature of configuration interaction: it gives rise to new intra-configuration operators, but it also contributes to already present operators; this makes it harder to approximate the model parameters *ab-initio*, but is not a practical obstacle for the semi-empirical approach (although it certainly complicates the physical interpretation that each parameter has). Furthermore, it is often the case that the operator set is limited to the subset {2,3,4,6,7,8}; a practice that is justified *post-facto* after seeing that these are sufficient to describe the data.

The calculation of a three body operator matrix elements across the $\underline{\mathrm{f}}^n$ configurations is analogous to how a two-body operator is calculated. Except that in this case what is needed are the reduced matrix elements in $\underline{\mathrm{f}}^3$ and the equation that is used to propagate these across the other configurations is equation 4 of [Jud66] (here adding the explicit dependence on J and M_J):

$$\langle \underline{\mathrm{f}}^n\psi|\hat{t}_i|\underline{\mathrm{f}}^n\psi'\rangle = \delta(J,J')\delta(M_J,M_J')\frac{n}{n-3}\sum_{\bar{\psi}\bar{\psi}'}\left(\psi\left\{\left|\bar{\psi}\right.\right)\left(\psi'\left\{\left|\bar{\psi}'\right.\right)\langle \underline{\mathrm{f}}^{n-1}\bar{\psi}|\hat{t}_i|\underline{\mathrm{f}}^{n-1}\bar{\psi}'\rangle. \right. \tag{51}$$

The sum in this expression runs over the parents in $\underline{\mathrm{f}}^{n-1}$ that are common to both the daughter terms ψ and ψ' in $\underline{\mathrm{f}}^n$. The equation above yielding LSJMJ matrix elements, and being diagonal in J, M_J as is due to a scalar operator.

In **qlanth** this is all implemented in the function `GenerateThreeBodyTables`. Where the matrix elements in $\underline{\mathrm{f}}^3$ are from [JS84], where the data has been digitized in the files `Judd1984-1.csv` and `Judd1984-2.csv`, which are parsed through the function `ParseJudd1984`.

In `GenerateThreeBodyTables` a special case is made for $\hat{t}_2$ and $\hat{t}_{11}$ which are calculated differently beyond the half-filled shell. In the case of the other $\hat{t}_k$ operators, beyond $\underline{\mathrm{f}}^7$ the matrix elements simply see a global sign flip, whereas in the case of $\hat{t}_2$ and $\hat{t}_{11}$ the coefficients of fractional parentage beyond $\underline{\mathrm{f}}^7$ are used. This yields the unexpected result that in the $\underline{\mathrm{f}}^{12}$ configuration, which corresponds to two holes, there is a non-zero three body operator $\hat{t}_2$. This is an arcane result that was corrected by Judd in 1984 [JS84], but which lingered long enough that important work in the 1980s was calculated with it. When calculations are carried out, if $\hat{t}_2{}'/\hat{t}_{11}{}'$ is used then $\hat{t}_2/\hat{t}_{11}$ should not be used and vice versa.

One additional feature of $\hat{t}_2$ that needs to be accounted for, is that it doesn't have the simple relationship for conjugate configurations that all the other $\hat{t}_i$ operators have. For the sake of simplicity, and to avoid having to explicitly store matrix elements beyond $\underline{\mathrm{f}}^7$ **qlanth** takes the approach of adding a control parameter `t2Switch` which needs to be set to 1 if below or at $\underline{\mathrm{f}}^7$ and set to 0 if above $\underline{\mathrm{f}}^7$.

```
GenerateThreeBodyTables::usage = "This function generates the reduced
    matrix elements for the three body operators using the
   coefficients of fractional parentage, including those beyond f^7."
   ;
Options[GenerateThreeBodyTables] = {"Export" -> False};
GenerateThreeBodyTables[OptionsPattern[]] := (
  tiKeys         = (StringReplace[ToString[#], {"T" -> "t_{", "p" -> "
   }^{'"}] <> "}") & /@ TSymbols;
  TSymbolsAssoc = AssociationThread[tiKeys -> TSymbols];
  juddOperators = ParseJudd1984[];
  (* op3MatrixElement[SL, SpLp, opSymbol] returns the value for the
   reduced matrix element of the operator opSymbol for the terms {SL,
    SpLp} in the f^3 configuration. *)
  op3MatrixElement[SL_, SpLp_, opSymbol_] := (
    jOP = juddOperators[{3, opSymbol}];
```

```
    key = {SL, SpLp};
    val = If[MemberQ[Keys[jOP], key],
      jOP[key],
      0];
    Return[val];
  );
  (* ti: This is the implementation of formula (2) in Judd & Suskin
    1984. It computes the reduced matrix elements of ti in f^n by
    using the reduced matrix elements in f^3 and the coefficients of
    fractional parentage. If the option ''Fast'' is set to True then
    the values for n>7 are simply computed as the negatives of the
    values in the complementary configuration; this except for t2 and
    t11 which are treated as special cases. *)
  Options[ti] = {"Fast" -> True};
  ti[nE_, SL_, SpLp_, tiKey_, opOrder_ : 3, OptionsPattern[]] :=
    Module[
    {nn, S, L, Sp, Lp,
    cfpSL, cfpSpLp,
    parentSL, parentSpLp,
    tnk, tnks},
    (
      {S, L}   = FindSL[SL];
      {Sp, Lp} = FindSL[SpLp];
      fast     = OptionValue["Fast"];
      numH     = 14 - nE;
      If[fast && Not[MemberQ[{"t_{2}","t_{11}"},tiKey]] && nE > 7,
        Return[-tktable[{numH, SL, SpLp, tiKey}]]
      ];
      If[(S == Sp && L == Lp),
      (
        cfpSL   = CFP[{nE, SL}];
        cfpSpLp = CFP[{nE, SpLp}];
        tnks = Table[(
            parentSL   = cfpSL[[nn, 1]];
            parentSpLp = cfpSpLp[[mm, 1]];
            cfpSL[[nn, 2]] * cfpSpLp[[mm, 2]] *
            tktable[{nE - 1, parentSL, parentSpLp, tiKey}]
            ),
            {nn, 2, Length[cfpSL]},
            {mm, 2, Length[cfpSpLp]}
            ];
        tnk = Total[Flatten[tnks]];
        ),
      tnk = 0;
      ];
      Return[ nE / (nE - opOrder) * tnk];
    )
  ];
  (* Calculate the reduced matrix elements of t^i for n up to 14 *)
  tktable = <||>;
  Do[(
    Do[(
      tkValue = Which[numE <= 2,
        (*Initialize n=1,2 with zeros*)
        0,
        numE == 3,
        (* Grab matrix elem in f^3 from Judd 1984 *)
        SimplifyFun[op3MatrixElement[SL, SpLp, opKey]],
        True,
        SimplifyFun[ti[numE, SL, SpLp, opKey, If[opKey == "e_{3}", 2,
   3]]]
        ];
      tktable[{numE, SL, SpLp, opKey}] = tkValue;
      ),
    {SL, AllowedNKSLTerms[numE]},
    {SpLp, AllowedNKSLTerms[numE]},
    {opKey, Append[tiKeys, "e_{3}"]}
    ];
    PrintTemporary[StringJoin["\[ScriptF]", ToString[numE], "
   configuration complete"]];
    ),
    {numE, 1, 14}
  ];

  (* Now use those reduced matrix elements to determine their sum as
    weighted by their corresponding strengths Ti *)
```

```
  ThreeBodyTable = <||>;
  Do[
    Do[
    (
      ThreeBodyTable[{numE, SL, SpLp}] = (
        Sum[(
          If[tiKey == "t_{2}", t2Switch, 1] *
          tktable[{numE, SL, SpLp, tiKey}] *
          TSymbolsAssoc[tiKey] +
          If[tiKey == "t_{2}", 1 - t2Switch, 0] *
          (-tktable[{14 - numE, SL, SpLp, tiKey}]) *
          TSymbolsAssoc[tiKey]
          ),
        {tiKey, tiKeys}
        ]
      );
    ),
    {SL, AllowedNKSLTerms[numE]},
    {SpLp, AllowedNKSLTerms[numE]}
    ];
  PrintTemporary[StringJoin["\[ScriptF]", ToString[numE], " matrix
    complete"]];,
  {numE, 1, 7}
  ];

  ThreeBodyTables = Table[(
    terms = AllowedNKSLTerms[numE];
    singleThreeBodyTable =
      Table[
        {SL, SLp} -> ThreeBodyTable[{numE, SL, SLp}],
        {SL, terms},
        {SLp, terms}
      ];
    singleThreeBodyTable  = Flatten[singleThreeBodyTable];
    singleThreeBodyTables = Table[(
        notNullPosition = Position[TSymbols, notNullSymbol][[1, 1]];
        reps = ConstantArray[0, Length[TSymbols]];
        reps[[notNullPosition]] = 1;
        rep = AssociationThread[TSymbols -> reps];
        notNullSymbol -> Association[(singleThreeBodyTable /. rep)]
        ),
      {notNullSymbol, TSymbols}
      ];
    singleThreeBodyTables = Association[singleThreeBodyTables];
    numE -> singleThreeBodyTables),
    {numE, 1, 7}
];

ThreeBodyTables = Association[ThreeBodyTables];
If[OptionValue["Export"],
  (
    threeBodyTablefname = FileNameJoin[{moduleDir, "data", "
    ThreeBodyTable.m"}];
    Export[threeBodyTablefname, ThreeBodyTable];
    threeBodyTablesfname = FileNameJoin[{moduleDir, "data", "
    ThreeBodyTables.m"}];
    Export[threeBodyTablesfname, ThreeBodyTables];
  )
 ];
Return[{ThreeBodyTable, ThreeBodyTables}];
);
```

```
ParseJudd1984::usage = "This function parses the data from tables 1
    and 2 of Judd from Judd, BR, and MA Suskin. ''Complete Set of
    Orthogonal Scalar Operators for the Configuration f^3''. JOSA B 1,
     no. 2 (1984): 261-65.";
Options[ParseJudd1984] = {"Export" -> False};
ParseJudd1984[OptionsPattern[]] := (
  ParseJuddTab1[str_] := (
    strR = ToString[str];
    strR = StringReplace[strR, ".5" -> "^(1/2)"];
    num = ToExpression[strR];
    sign = Sign[num];
    num = sign*Simplify[Sqrt[num^2]];
    If[Round[num] == num, num = Round[num]];
    Return[num]);
```

```
12
13  (* Parse table 1 from Judd 1984 *)
14  judd1984Fname1 = FileNameJoin[{moduleDir, "data", "Judd1984-1.csv"
      }];
15  data = Import[judd1984Fname1, "CSV", "Numeric" -> False];
16  headers = data[[1]];
17  data = data[[2 ;;]];
18  data = Transpose[data];
19  \[Psi] = Select[data[[1]], # != "" &];
20  \[Psi]p = Select[data[[2]], # != "" &];
21  matrixKeys = Transpose[{\[Psi], \[Psi]p}];
22  data = data[[3 ;;]];
23  cols = Table[ParseJuddTab1 /@ Select[col, # != "" &], {col, data}];
24  cols = Select[cols, Length[#] == 21 &];
25  tab1 = Prepend[Prepend[cols, \[Psi]p], \[Psi]];
26  tab1 = Transpose[Prepend[Transpose[tab1], headers]];
27
28  (* Parse table 2 from Judd 1984 *)
29  judd1984Fname2 = FileNameJoin[{moduleDir, "data", "Judd1984-2.csv"
      }];
30  data = Import[judd1984Fname2, "CSV", "Numeric" -> False];
31  headers = data[[1]];
32  data = data[[2 ;;]];
33  data = Transpose[data];
34  {operatorLabels, WUlabels, multiFactorSymbols, multiFactorValues} =
       data[[;; 4]];
35  multiFactorValues = ParseJuddTab1 /@ multiFactorValues;
36  multiFactorValues = AssociationThread[multiFactorSymbols ->
      multiFactorValues];
37
38  (*scale values of table 1 given the values in table 2*)
39  oppyS = {};
40  normalTable =
41    Table[header = col[[1]];
42      If[StringContainsQ[header, " "],
43        (
44          multiplierSymbol = StringSplit[header, " "][[1]];
45          multiplierValue = multiFactorValues[multiplierSymbol];
46          operatorSymbol = StringSplit[header, " "][[2]];
47          oppyS = Append[oppyS, operatorSymbol];
48        ),
49        (
50          multiplierValue = 1;
51          operatorSymbol = header;
52        )
53      ];
54      normalValues = 1/multiplierValue*col[[2 ;;]];
55      Join[{operatorSymbol}, normalValues], {col, tab1[[3 ;;]]}
56    ];
57
58  (*Create an association for the reduced matrix elements in the f^3
      config*)
59  juddOperators = Association[];
60  Do[(
61    col      = normalTable[[colIndex]];
62    opLabel  = col[[1]];
63    opValues = col[[2 ;;]];
64    opMatrix = AssociationThread[matrixKeys -> opValues];
65    Do[(
66      opMatrix[Reverse[mKey]] = opMatrix[mKey]
67      ),
68    {mKey, matrixKeys}
69    ];
70    juddOperators[{3, opLabel}] = opMatrix),
71    {colIndex, 1, Length[normalTable]}
72  ];
73
74  (* special case of t2 in f3 *)
75  (* this is the same as getting the reduced matrix elements from
      Judd 1966 *)
76  numE = 3;
77  e3Op      = juddOperators[{3, "e_{3}"}];
78  t2prime   = juddOperators[{3, "t_{2}^{'}"}];
79  prefactor = 1/(70 Sqrt[2]);
80  t2Op = (# -> (t2prime[#] + prefactor*e3Op[#])) & /@ Keys[t2prime];
81  t2Op = Association[t2Op];
```

```
  juddOperators[{3, "t_{2}"}] = t2Op;

  (*Special case of t11 in f3*)
  t11 = juddOperators[{3, "t_{11}"}];
  eβprimeOp = juddOperators[{3, "e_{\\beta}^{'}"}];
  t11primeOp = (# -> (t11[#] + Sqrt[3/385] eβprimeOp[#])) & /@ Keys[
    t11];
  t11primeOp = Association[t11primeOp];
  juddOperators[{3, "t_{11}^{'}"}] = t11primeOp;
  If[OptionValue["Export"],
    (
      (*export them*)
      PrintTemporary["Exporting ..."];
      exportFname = FileNameJoin[{moduleDir, "data", "juddOperators.m
    "}];
      Export[exportFname, juddOperators];
    )
  ];
  Return[juddOperators];
);
```

3.9 $\hat{\mathcal{H}}_{\mathbf{cf}}$: crystal-field

The crystal-field partially accounts for the influence of the surrounding lattice on the ion. The simplest picture of this influence imagines the lattice as responsible for an electric field felt at the position of the ion. This electric field corresponding to an electrostatic potential described as a multipolar sum of the form:

$$V(r_i, \theta_i, \phi_i) = \sum_{k=1}^{\infty} \sum_{q=-k}^{k} \mathcal{A}_q^{(k)} r_i^k \mathcal{C}_q^{(k)}(\theta_i, \phi_i) \tag{52}$$

where the $\mathcal{C}_q^{(k)}$ are spherical harmonics normalized with the Racah convention

$$\mathcal{C}_q^{(k)} = \sqrt{\frac{4\pi}{2k+1}} Y_q^{(k)}. \tag{53}$$

Here we have chosen a coordinate system with its origin at the position of the nucleus, and in which we only have positive powers of the distance r_i because we have expanded the contributions from all the surrounding ions as a sum over spherical harmonics centered at the position of the nucleus, without r ever large enough to reach any of the positions of the lattice ions.

Furthermore, since we have n valence electrons, then the total crystal field potential is

$$\hat{\mathcal{H}}_{\mathrm{cf}}(\vec{r}) = \sum_{i=1}^{n} \sum_{k=0}^{\infty} \sum_{q=-k}^{k} \mathcal{A}_q^{(k)} r_i^k \mathcal{C}_q^{(k)}(\theta_i, \phi_i). \tag{54}$$

And if we average the radial coordinate,

$$\hat{\mathcal{H}}_{\mathrm{cf}} = \sum_{i=1}^{n} \sum_{k=1}^{\infty} \sum_{q=-k}^{k} \mathcal{B}_q^{(k)} \mathcal{C}_q^{(k)}(i) \tag{55}$$

where the radial average is included as

$$\mathcal{B}_q^{(k)} := \mathcal{A}_q^{(k)} \langle 4f | r^k | 4f \rangle. \tag{56}$$

$\mathcal{B}_q^{(k)}$ may be complex in general. However, since the sum in Eqn-54 needs to result in a real and Hermitian operator, there are restrictions on $\mathcal{B}_q^{(k)}$ that need to be accounted for. Once the behavior of $\mathcal{C}_q^{(k)}$ under complex conjugation is considered, $\mathcal{C}_q^{(k)*} = (-1)^q \mathcal{C}_{-q}^{(k)}$, it is necessary that

$$\mathcal{B}_q^{(k)} = (-1)^q \mathcal{B}_{-q}^{(k)*}. \tag{57}$$

Presently the sum over q spans both its negative and positive values. This can be limited to only the non-negative values of q. Separating the real and imaginary parts of $\mathcal{B}_q^{(k)}$ such that $\mathcal{B}_q^{(k)} = B_q^{(k)} + iS_q^{(k)}$ for $q \neq 0$ and $\mathcal{B}_0^{(k)} = 2B_0^{(k)}$ the sum for the crystal field can then be written as

$$\hat{\mathcal{H}}_{\mathrm{cf}}(\vec{r}) = \sum_{i=1}^{n} \sum_{k=0}^{\infty} \sum_{q=0}^{k} B_q^{(k)} \left(\mathcal{C}_q^{(k)} + (-1)^q \mathcal{C}_{-q}^{(k)} \right) + iS_q^{(k)} \left(\mathcal{C}_q^{(k)} - (-1)^q \mathcal{C}_{-q}^{(k)} \right). \tag{58}$$

A staple of the Wigner-Racah algebra is writing up operators of interest in terms of standard ones for which the matrix elements are straightforward. One such operator is the unit tensor operator $\hat{u}^{(k)}$ for a single electron. The Wigner-Eckart theorem – on which all of this algebra is an elaboration – effectively separates the dynamical and geometrical parts of a given interaction; the unit tensor operators isolate the geometric contributions. This irreducible tensor operator $\hat{u}^{(k)}$ is defined as the tensor operator having the following reduced matrix elements (written in terms of the triangular delta, see section on notation):

$$\langle \ell \| \hat{u}^{(k)} \| \ell' \rangle = 1. \tag{59}$$

In terms of this tensor one may then define the symmetric (in the sense that the resulting operator is equitable among all electrons) unit tensor operator for n particles as

$$\hat{U}^{(k)} = \sum_{i}^{n} \hat{u}_i^{(k)}. \tag{60}$$

This tensor is relevant to the calculation of the above matrix elements since

$$C_q^{(k)} = \langle \underline{\ell} \| C^{(k)} \| \underline{\ell}' \rangle \hat{u}_q^{(k)} = (-1)^{\underline{\ell}} \sqrt{[\![\underline{\ell}]\!][\![\underline{\ell}']\!]} \begin{pmatrix} \underline{\ell} & k & \underline{\ell}' \\ 0 & 0 & 0 \end{pmatrix} \hat{u}_q^{(k)}. \tag{61}$$

With this, the matrix elements of $\hat{\mathcal{H}}_{\rm cf}$ in the $|LSJM_J\rangle$ basis are:

$$\overbrace{\langle \underline{\ell}^n \alpha SLJM_J | \hat{\mathcal{H}}_{\rm cf} | \underline{\ell}^n \alpha' SL'J'M_{J'} \rangle}^{\text{Wybourne eqn. 6-3}} = \sum_{k=1}^{\infty} \sum_{q=-k}^{k} \mathcal{B}_q^{(k)} \langle \underline{\ell}^n \alpha SLJM_J | \hat{U}_q^{(k)} | \underline{\ell}^n \alpha' SL'J'M_{J'} \rangle \langle \underline{\ell} \| \hat{C}^{(k)} \| \underline{\ell} \rangle \tag{62}$$

where the matrix elements of $\hat{U}_q^{(k)}$ can be resolved with a 3j symbol as

$$\overbrace{\langle \underline{\ell}^n \alpha SLJM_J | \hat{U}_q^{(k)} | \underline{\ell}^n \alpha' S'L'J'M_{J'} \rangle}^{\text{Wybourne eqn. 6-4}} = (-1)^{J-M_J} \begin{pmatrix} J & k & J' \\ -M_J & q & M_{J'} \end{pmatrix} \langle \underline{\ell}^n \alpha SLJ \| \hat{U}^{(k)} \| \underline{\ell}^n \alpha' S'L' \rangle \tag{63}$$

and reduced a second time with the inclusion of a 6j symbol resulting in

$$\begin{aligned} \overbrace{\langle \underline{\ell}^n \alpha SLJ \| \hat{U}^{(k)} \| \underline{\ell}^n \alpha' S'L' \rangle}^{\text{Wybourne eqn. 6-5}} = (-1)^{S+L+J'+k} \sqrt{[\![J]\!][\![J']\!]} \times \\ \begin{Bmatrix} J & J' & k \\ L' & L & S \end{Bmatrix} \langle \underline{\ell}^n \alpha SL \| \hat{U}^{(k)} \| \underline{\ell}^n \alpha' S'L' \rangle. \end{aligned} \tag{64}$$

This last reduced matrix element is finally computed by summing over $\bar{\alpha}\bar{L}\bar{S}$ which are the $\underline{\mathrm{f}}^{n-1}$ parents which are common to $|\alpha LS\rangle$ and $|\alpha' L'S'\rangle$ from the $\underline{\mathrm{f}}^n$ configuration:

$$\begin{aligned} \overbrace{\langle \underline{\ell}^n \alpha SL \| \hat{U}^{(k)} \| \underline{\ell}^n \alpha' S'L' \rangle}^{\text{Cowan eqn. 11.53}} = \delta(S,S') n (-1)^{\underline{\ell}+L+k} \sqrt{[\![L]\!][\![L']\!]} \times \\ \sum_{\bar{\alpha}\bar{L}\bar{S}} (-1)^{\bar{L}} \begin{Bmatrix} \underline{\ell} & k & \underline{\ell} \\ L & \bar{L} & L' \end{Bmatrix} \left(\underline{\ell}^n \alpha LS \middle\{ \underline{\ell}^{n-1} \bar{\alpha}\bar{L}\bar{S} \right) \left(\underline{\ell}^{n-1} \bar{\alpha}\bar{L}\bar{S} \middle\} \underline{\ell}^n \alpha' L'S' \right). \end{aligned} \tag{65}$$

From the $\langle \underline{\ell} \| \hat{C}^{(k)} \| \underline{\ell} \rangle$, and given that we are using $\underline{\ell} = \underline{\mathrm{f}} = 3$, we can see that by the triangular condition $\triangle(3,k,3)$ the non-zero contributions only come from $k = 0,1,2,3,4,5,6$. An additional selection rule on k comes from considerations of parity. Since both the bra and the ket in $\langle \underline{\ell}^n \alpha SLJM_J | \hat{\mathcal{H}}_{\rm cf} | \underline{\ell}^n \alpha' SL'J'M_{J'} \rangle$ have the same parity, then the overall parity of the braket is determined by the parity of $C_q^{(k)}$, and since the parity of $C_q^{(k)}$ is $(-1)^k$ then for the braket to be non-zero we require that k should also be even. In view of this, in all the above equations for the crystal field the values for k should be limited to $2,4,6$. The value of $k = 0$ having been omitted from the start since this only contributes a common energy shift. Putting everything together:

$$\hat{\mathcal{H}}_{\rm cf}(\vec{r}) = \sum_{i=1}^{n} \sum_{k=2,4,6} \sum_{q=0}^{k} B_q^{(k)} \left(C_q^{(k)} + (-1)^q C_{-q}^{(k)} \right) + i S_q^{(k)} \left(C_q^{(k)} - (-1)^q C_{-q}^{(k)} \right). \tag{66}$$

The above equations are implemented in **qlanth** by the function CrystalField. This function puts together the symbolic sum in Eqn-62 by using the function Cqk. Cqk then uses the diagonal reduced matrix elements of $\mathcal{C}_q^{(k)}$ and the precomputed values for Uk (stored in ReducedUkTable).

The required reduced matrix elements of $\hat{U}^{(k)}$ are calculated by the function ReducedUk, which is used by GenerateReducedUkTable to precompute its values.

```
Bqk::usage = "Real part of the Bqk coefficients.";
Bqk[q_, 2] := {B02/2, B12, B22}[[q + 1]];
Bqk[q_, 4] := {B04/2, B14, B24, B34, B44}[[q + 1]];
Bqk[q_, 6] := {B06/2, B16, B26, B36, B46, B56, B66}[[q + 1]];
```

```
Sqk::usage = "Imaginary part of the Bqk coefficients.";
Sqk[q_, 2] := {0, S12, S22}[[q + 1]];
Sqk[q_, 4] := {0, S14, S24, S34, S44}[[q + 1]];
Sqk[q_, 6] := {0, S16, S26, S36, S46, S56, S66}[[q + 1]];
```

```
Cqk::usage = "Cqk[numE, q, k, NKSL, J, M, NKSLp, Jp, Mp]. In Wybourne
    (1965) see equations 6-3, 6-4, and 6-5. Also in TASS see equation
    11.53.";
Cqk[numE_, q_, k_, NKSL_, J_, M_, NKSLp_, Jp_, Mp_] := Module[
  {S, Sp, L, Lp, orbital, val},
  (
    orbital = 3;
    {S, L}   = FindSL[NKSL];
    {Sp, Lp} = FindSL[NKSLp];
    f1  = ThreeJay[{J, -M}, {k, q}, {Jp, Mp}];
    val =
      If[f1==0,
        0,
        (
          f2 = SixJay[{L, J, S}, {Jp, Lp, k}] ;
          If[f2==0,
            0,
            (
              f3 = ReducedUkTable[{numE, orbital, NKSL, NKSLp, k}];
              If[f3==0,
                0,
                (
                  (
                    Phaser[J - M + S + Lp + J + k] *
                    Sqrt[TPO[J, Jp]] *
                    f1 *
                    f2 *
                    f3 *
                    Ck[orbital, k]
                  )
                )
              ]
            )
          ]
        )
      ];
    Return[val];
  )
];
```

```
CrystalField::usage = "CrystalField[n, NKSL, J, M, NKSLp, Jp, Mp]
    gives the general expression for the matrix element of the crystal
     field Hamiltonian parametrized with Bqk and Sqk coefficients as a
     sum over spherical harmonics Cqk.
Sometimes this expression only includes Bqk coefficients, see for
    example eqn 6-2 in Wybourne (1965), but one may also split the
    coefficient into real and imaginary parts as is done here, in an
    expression that is patently Hermitian.";
CrystalField[numE_, NKSL_, J_, M_, NKSLp_, Jp_, Mp_] := (
  Sum[
    (
      cqk  = Cqk[numE,  q, k, NKSL, J, M, NKSLp, Jp, Mp];
      cmqk = Cqk[numE, -q, k, NKSL, J, M, NKSLp, Jp, Mp];
      Bqk[q, k]   * (cqk + (-1)^q * cmqk) +
      I*Sqk[q, k] * (cqk - (-1)^q * cmqk)
      ),
  {k, {2, 4, 6}},
```

```
  {q, 0, k}
  ]
)
```

```
ReducedUk::usage = "ReducedUk[n, l, SL, SpLp, k] gives the reduced
    matrix element of the symmetric unit tensor operator U^(k). See
    equation 11.53 in TASS.";
ReducedUk[numE_, l_, SL_, SpLp_, k_] := Module[
  {spin, orbital, Uk, S, L,
  Sp, Lp, Sb, Lb, parentSL,
  cfpSL, cfpSpLp, Ukval,
  SLparents, SLpparents,
  commonParents, phase},
    {spin, orbital} = {1/2, 3};
    {S, L}          = FindSL[SL];
    {Sp, Lp}        = FindSL[SpLp];
    If[Not[S == Sp],
      Return[0]
    ];
    cfpSL       = CFP[{numE, SL}];
    cfpSpLp     = CFP[{numE, SpLp}];
    SLparents   = First /@ Rest[cfpSL];
    SLpparents  = First /@ Rest[cfpSpLp];
    commonParents = Intersection[SLparents, SLpparents];
    Uk = Sum[(
      {Sb, Lb} = FindSL[\[Psi]b];
      Phaser[Lb] *
        CFPAssoc[{numE, SL, \[Psi]b}] *
        CFPAssoc[{numE, SpLp, \[Psi]b}] *
        SixJay[{orbital, k, orbital}, {L, Lb, Lp}]
    ),
    {\[Psi]b, commonParents}
    ];
    phase     = Phaser[orbital + L + k];
    prefactor = numE * phase * Sqrt[TPO[L,Lp]];
    Ukval     = prefactor*Uk;
    Return[Ukval];
]
```

Each of the 32 crystallographic point groups requires only a limited number of non-zero crystal field parameters. In **qlanth** these can be queried programatically with the use of the function `CrystalFieldForm`. These were taken from a table in Benelli and Gatteschi [BG15] and their corresponding expressions (for a single electron) are in the equations below with a table linking to the corresponding equations. Note that these expressions bring with them an implicit choice for the orientation of the coordinate system (see Section 4).

$\mathcal{S}_2$: &eqn-67	$\mathcal{C}_s$: &eqn-68	$\mathcal{C}_{1h}$: &eqn-69	$\mathcal{C}_2$: &eqn-70	$\mathcal{C}_{2h}$: &eqn-71
$\mathcal{C}_{2v}$: &eqn-72	$\mathcal{D}_2$: &eqn-73	$\mathcal{D}_{2h}$: &eqn-74	$\mathcal{S}_4$: &eqn-75	$\mathcal{C}_4$: &eqn-76
$\mathcal{C}_{4h}$: &eqn-77	$\mathcal{D}_{2d}$: &eqn-78	$\mathcal{C}_{4v}$: &eqn-79	$\mathcal{D}_4$: &eqn-80	$\mathcal{D}_{4h}$: &eqn-81
$\mathcal{C}_3$: &eqn-82	$\mathcal{S}_6$: &eqn-83	$\mathcal{C}_{3h}$: &eqn-84	$\mathcal{C}_{3v}$: &eqn-85	$\mathcal{D}_3$: &eqn-86
$\mathcal{D}_{3d}$: &eqn-87	$\mathcal{D}_{3h}$: &eqn-88	$\mathcal{C}_6$: &eqn-89	$\mathcal{C}_{6h}$: &eqn-90	$\mathcal{C}_{6v}$: &eqn-91
$\mathcal{D}_6$: &eqn-92	$\mathcal{D}_{6h}$: &eqn-93	$\mathcal{T}$: &eqn-94	$\mathcal{T}_h$: &eqn-95	$\mathcal{T}_d$: &eqn-96
$\mathcal{O}$: &eqn-97	$\mathcal{O}_h$: &eqn-98			

Table 1: Expressions for the crystal field in the 32 crystallographic point groups

Crystal field expressions

$$\begin{aligned}\hat{\mathcal{H}}_{\mathrm{cf}}(\mathcal{S}_2) = {} & B_0^{(2)}\mathcal{C}_0^{(2)} + B_1^{(2)}\mathcal{C}_1^{(2)} + \left(B_2^{(2)} + iS_2^{(2)}\right)\mathcal{C}_2^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} \\ & + \left(B_1^{(4)} + iS_1^{(4)}\right)\mathcal{C}_1^{(4)} + \left(B_2^{(4)} + iS_2^{(4)}\right)\mathcal{C}_2^{(4)} + \left(B_3^{(4)} + iS_3^{(4)}\right)\mathcal{C}_3^{(4)} + \left(B_4^{(4)} + iS_4^{(4)}\right)\mathcal{C}_4^{(4)} \\ & + B_0^{(6)}\mathcal{C}_0^{(6)} + \left(B_1^{(6)} + iS_1^{(6)}\right)\mathcal{C}_1^{(6)} + \left(B_2^{(6)} + iS_2^{(6)}\right)\mathcal{C}_2^{(6)} + \left(B_3^{(6)} + iS_3^{(6)}\right)\mathcal{C}_3^{(6)} \\ & + \left(B_4^{(6)} + iS_4^{(6)}\right)\mathcal{C}_4^{(6)} + \left(B_5^{(6)} + iS_5^{(6)}\right)\mathcal{C}_5^{(6)} + \left(B_6^{(6)} + iS_6^{(6)}\right)\mathcal{C}_6^{(6)} \end{aligned} \quad (67)$$

$$\hat{\mathscr{H}}_{\rm cf}\left(\mathcal{C}_s\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_2^{(2)}\mathcal{C}_2^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + \left(B_2^{(4)} + iS_2^{(4)}\right)\mathcal{C}_2^{(4)} + \left(B_4^{(4)} + iS_4^{(4)}\right)\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + \left(B_2^{(6)} + iS_2^{(6)}\right)\mathcal{C}_2^{(6)} + \left(B_4^{(6)} + iS_4^{(6)}\right)\mathcal{C}_4^{(6)} + \left(B_6^{(6)} + iS_6^{(6)}\right)\mathcal{C}_6^{(6)} \quad (68)$$

$$\hat{\mathscr{H}}_{\rm cf}\left(\mathcal{C}_{1h}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_2^{(2)}\mathcal{C}_2^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + \left(B_2^{(4)} + iS_2^{(4)}\right)\mathcal{C}_2^{(4)} + \left(B_4^{(4)} + iS_4^{(4)}\right)\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + \left(B_2^{(6)} + iS_2^{(6)}\right)\mathcal{C}_2^{(6)} + \left(B_4^{(6)} + iS_4^{(6)}\right)\mathcal{C}_4^{(6)} + \left(B_6^{(6)} + iS_6^{(6)}\right)\mathcal{C}_6^{(6)} \quad (69)$$

$$\hat{\mathscr{H}}_{\rm cf}\left(\mathcal{C}_2\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_2^{(2)}\mathcal{C}_2^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + \left(B_2^{(4)} + iS_2^{(4)}\right)\mathcal{C}_2^{(4)} + \left(B_4^{(4)} + iS_4^{(4)}\right)\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + \left(B_2^{(6)} + iS_2^{(6)}\right)\mathcal{C}_2^{(6)} + \left(B_4^{(6)} + iS_4^{(6)}\right)\mathcal{C}_4^{(6)} + \left(B_6^{(6)} + iS_6^{(6)}\right)\mathcal{C}_6^{(6)} \quad (70)$$

$$\hat{\mathscr{H}}_{\rm cf}\left(\mathcal{C}_{2h}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_2^{(2)}\mathcal{C}_2^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + \left(B_2^{(4)} + iS_2^{(4)}\right)\mathcal{C}_2^{(4)} + \left(B_4^{(4)} + iS_4^{(4)}\right)\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + \left(B_2^{(6)} + iS_2^{(6)}\right)\mathcal{C}_2^{(6)} + \left(B_4^{(6)} + iS_4^{(6)}\right)\mathcal{C}_4^{(6)} + \left(B_6^{(6)} + iS_6^{(6)}\right)\mathcal{C}_6^{(6)} \quad (71)$$

$$\hat{\mathscr{H}}_{\rm cf}\left(\mathcal{C}_{2v}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_2^{(2)}\mathcal{C}_2^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_2^{(4)}\mathcal{C}_2^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_2^{(6)}\mathcal{C}_2^{(6)} + B_4^{(6)}\mathcal{C}_4^{(6)} + B_6^{(6)}\mathcal{C}_6^{(6)} \quad (72)$$

$$\hat{\mathscr{H}}_{\rm cf}\left(\mathcal{D}_2\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_2^{(2)}\mathcal{C}_2^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_2^{(4)}\mathcal{C}_2^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_2^{(6)}\mathcal{C}_2^{(6)} + B_4^{(6)}\mathcal{C}_4^{(6)} + B_6^{(6)}\mathcal{C}_6^{(6)} \quad (73)$$

$$\hat{\mathscr{H}}_{\rm cf}\left(\mathcal{D}_{2h}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_2^{(2)}\mathcal{C}_2^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_2^{(4)}\mathcal{C}_2^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_2^{(6)}\mathcal{C}_2^{(6)} + B_4^{(6)}\mathcal{C}_4^{(6)} + B_6^{(6)}\mathcal{C}_6^{(6)} \quad (74)$$

$$\hat{\mathscr{H}}_{\rm cf}\left(\mathcal{S}_4\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + \left(B_4^{(6)} + iS_4^{(6)}\right)\mathcal{C}_4^{(6)} \quad (75)$$

$$\hat{\mathscr{H}}_{\rm cf}\left(\mathcal{C}_4\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + \left(B_4^{(6)} + iS_4^{(6)}\right)\mathcal{C}_4^{(6)} \quad (76)$$

$$\hat{\mathscr{H}}_{\rm cf}\left(\mathcal{C}_{4h}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + \left(B_4^{(6)} + iS_4^{(6)}\right)\mathcal{C}_4^{(6)} \quad (77)$$

$$\hat{\mathscr{H}}_{\rm cf}\left(\mathcal{D}_{2d}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_4^{(6)}\mathcal{C}_4^{(6)} \quad (78)$$

$$\hat{\mathscr{H}}_{\rm cf}\left(\mathcal{C}_{4v}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_4^{(6)}\mathcal{C}_4^{(6)} \quad (79)$$

$$\hat{\mathscr{H}}_{\rm cf}\left(\mathcal{D}_4\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_4^{(6)}\mathcal{C}_4^{(6)} \quad (80)$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{D}_{4h}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_4^{(6)}\mathcal{C}_4^{(6)} \tag{81}$$

$$\begin{aligned}\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{C}_3\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_3^{(4)}\mathcal{C}_3^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} \\ + \left(B_3^{(6)} + iS_3^{(6)}\right)\mathcal{C}_3^{(6)} + \left(B_6^{(6)} + iS_6^{(6)}\right)\mathcal{C}_6^{(6)}\end{aligned} \tag{82}$$

$$\begin{aligned}\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{S}_6\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_3^{(4)}\mathcal{C}_3^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} \\ + \left(B_3^{(6)} + iS_3^{(6)}\right)\mathcal{C}_3^{(6)} + \left(B_6^{(6)} + iS_6^{(6)}\right)\mathcal{C}_6^{(6)}\end{aligned} \tag{83}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{C}_{3h}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_6^{(6)}\mathcal{C}_6^{(6)} \tag{84}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{C}_{3v}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_3^{(4)}\mathcal{C}_3^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_3^{(6)}\mathcal{C}_3^{(6)} + B_6^{(6)}\mathcal{C}_6^{(6)} \tag{85}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{D}_3\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_3^{(4)}\mathcal{C}_3^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_3^{(6)}\mathcal{C}_3^{(6)} + B_6^{(6)}\mathcal{C}_6^{(6)} \tag{86}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{D}_{3d}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_3^{(4)}\mathcal{C}_3^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_3^{(6)}\mathcal{C}_3^{(6)} + B_6^{(6)}\mathcal{C}_6^{(6)} \tag{87}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{D}_{3h}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_6^{(6)}\mathcal{C}_6^{(6)} \tag{88}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{C}_6\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_6^{(6)}\mathcal{C}_6^{(6)} \tag{89}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{C}_{6h}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_6^{(6)}\mathcal{C}_6^{(6)} \tag{90}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{C}_{6v}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_6^{(6)}\mathcal{C}_6^{(6)} \tag{91}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{D}_6\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_6^{(6)}\mathcal{C}_6^{(6)} \tag{92}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{D}_{6h}\right) = B_0^{(2)}\mathcal{C}_0^{(2)} + B_0^{(4)}\mathcal{C}_0^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_6^{(6)}\mathcal{C}_6^{(6)} \tag{93}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{T}\right) = B_0^{(4)}\mathcal{C}_0^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_4^{(6)}\mathcal{C}_4^{(6)} \tag{94}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{T}_h\right) = B_0^{(4)}\mathcal{C}_0^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_4^{(6)}\mathcal{C}_4^{(6)} \tag{95}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{T}_d\right) = B_0^{(4)}\mathcal{C}_0^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_4^{(6)}\mathcal{C}_4^{(6)} \tag{96}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{O}\right) = B_0^{(4)}\mathcal{C}_0^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_4^{(6)}\mathcal{C}_4^{(6)} \tag{97}$$

$$\hat{\mathcal{H}}_{\rm cf}\left(\mathcal{O}_h\right) = B_0^{(4)}\mathcal{C}_0^{(4)} + B_4^{(4)}\mathcal{C}_4^{(4)} + B_0^{(6)}\mathcal{C}_0^{(6)} + B_4^{(6)}\mathcal{C}_4^{(6)} \tag{98}$$

(END) Crystal field expressions (END)

```
CrystalFieldForm::usage = "CrystalFieldForm[symmetryGroup] returns an
     association that describes the crystal field parameters that are
    necessary to desribe a crystal field for the given symmetry group.

The symmetry group must be given as a string in Schoenflies notation
    and must be one of C1, Ci, S2, Cs, C1h, C2, C2h, C2v, D2, D2h, S4,
     C4, C4h, D2d, C4v, D4, D4h, C3, S6, C3h, C3v, D3, D3d, D3h, C6,
    C6h, C6v, D6, D6h, T, Th, Td, O, Oh.

The returned association has three keys:
  ''BqkSqk'' whose values is a list with the nonzero Bqk and Sqk
    parameters;
  ''constraints'' whose value is either an empty list, or a lists of
    replacements rules that are constraints on the Bqk and Sqk
    parameters;
  ''simplifier'' whose value is an association that can be used to
    set to zero the crystal field parameters that are zero for the
    given symmetry group;
  ''aliases'' whose value is a list with the integer by which the
    point group is also known for and an alternate Schoenflies symbol
    if it exists.

This uses data from table 3.3 in Benelli and Gatteschi, 2015.";
CrystalFieldForm[symmetryGroupString_] := (
  If[Not@ValueQ[crystalFieldFunctionalForms],
    crystalFieldFunctionalForms = Import[FileNameJoin[{moduleDir, "
    data", "crystalFieldFunctionalForms.m"}]];
  ];
  cfForm = crystalFieldFunctionalForms[symmetryGroupString];
  simplifier = Association[(# -> 0) &/@ Complement[cfSymbols, cfForm[
    "BqkSqk"]]];
  Return[Join[cfForm, <|"simplifier"->simplifier|>]];
)
```

3.10 $\hat{\mu}$ and $\hat{\mathcal{H}}_{\mathbf{Z}}$: the magnetic dipole operator and the Zeeman term

In Hartree atomic units, the operator associated with the magnetic dipole operator for an electron is

$$\hat{\mu} = -\mu_{\mathrm{B}} \left(\hat{L} + g_s \hat{S}\right)^{(1)}, \text{with } \mu_{\mathrm{B}} = 1/2. \tag{99}$$

Here we have emphasized the fact that the magnetic dipole operator corresponds to a rank-1 spherical tensor operator.

In the $|LSJM_J\rangle$ basis that we use in **qlanth** the LSJ reduced-matrix elements are computed using equation 15.7 in [Cow81]

$$\langle \alpha LSJ \| \left(\hat{L} + g_s \hat{S}\right)^{(1)} \| \alpha' L'S'J' \rangle = \delta(\alpha LSJ, \alpha' L'S'J') \sqrt{J(J+1)(2J+1)} + \delta(\alpha LS, \alpha' L'S') (-1)^{L+S+J+1} \sqrt{[\![J]\!][\![J]\!]} \begin{Bmatrix} L & S & J \\ 1 & J' & S \end{Bmatrix}. \tag{100}$$

Then these reduced matrix elements are used to resolve the M_J components for $q = -1, 0, 1$ through Wigner-Eckart:

$$\langle \alpha LSJM_J | \left(\hat{L} + g_s \hat{S}\right)^{(1)}_q | \alpha' L'S'J'M_{J'} \rangle = (-1)^{J-M_J} \begin{pmatrix} J & 1 & J' \\ -M_J & q & M'_J \end{pmatrix} \langle \alpha LSJ \| \left(\hat{L} + g_s \hat{S}\right)^{(1)} \| \alpha' L'S'J' \rangle. \tag{101}$$

These two above are put together in `JJBlockMagDip` for given $\{n, J, J'\}$ returning a rank-3 array representing the quantities $\{M_J, M'_J, q\}$.

```
JJBlockMagDip::usage = "JJBlockMagDip[numE, J, Jp] returns an array
    for the LSJM matrix elements of the magnetic dipole operator
    between states with given J and Jp. The option ''Sparse'' can be
    used to return a sparse matrix. The default is to return a sparse
    matrix.
See eqn 15.7 in TASS.
Here it is provided in atomic units in which the Bohr magneton is
    1/2.
\[Mu] = -(1/2) (L + gs S)
```

```
We are using the Racah convention for the reduced matrix elements in
    the Wigner-Eckart theorem. See TASS eqn 11.15.
";
Options[JJBlockMagDip]={"Sparse"->True};
JJBlockMagDip[numE_, braJ_, ketJ_, OptionsPattern[]] := Module[
  {braSLJs, ketSLJs,
  braSLJ,   ketSLJ,
  braSL,    ketSL,
  braS,     braL,
  ketS,     ketL,
  braMJ,    ketMJ,
  matValue, magMatrix,
  summand1, summand2,
  threejays},
  (
    braSLJs  = AllowedNKSLJMforJTerms[numE,braJ];
    ketSLJs  = AllowedNKSLJMforJTerms[numE,ketJ];
    magMatrix  = Table[
      braSL     = braSLJ[[1]];
      ketSL     = ketSLJ[[1]];
      {braS, braL}   = FindSL[braSL];
      {ketS, ketL}   = FindSL[ketSL];
      braMJ       = braSLJ[[3]];
      ketMJ       = ketSLJ[[3]];
      summand1    = If[Or[braJ  != ketJ,
                          braSL != ketSL],
        0,
        Sqrt[braJ*(braJ+1)*TPO[braJ]]
      ];
      (* looking at the string includes checking L=L', S=S', and \
    alpha=\alpha'*)
      summand2 = If[braSL!= ketSL,
        0,
        (gs-1) *
          Phaser[braS+braL+ketJ+1] *
            Sqrt[TPO[braJ]*TPO[ketJ]] *
              SixJay[{braJ,1,ketJ},{braS,braL,braS}] *
                Sqrt[braS(braS+1)TPO[braS]]
      ];
      matValue = summand1 + summand2;
      (* We are using the Racah convention for red matrix elements in
     Wigner-Eckart *)
      threejays = (ThreeJay[{braJ, -braMJ}, {1, #}, {ketJ, ketMJ}] &)
     /@ {-1,0,1};
      threejays *= Phaser[braJ-braMJ];
      matValue  = - 1/2 * threejays * matValue;
      matValue,
    {braSLJ, braSLJs},
    {ketSLJ, ketSLJs}
    ];
    If[OptionValue["Sparse"],
      magMatrix = SparseArray[magMatrix]
    ];
    Return[magMatrix];
  )
];
```

The JJ' blocks that are generated with this function are then put together by MagDipoleMatrixAssembly into the final matrix form and the cartesian components calculated according to

$$\hat{\mu}_x = \frac{\hat{\mu}^{(1)}_{-1} - \hat{\mu}^{(1)}_{+1}}{\sqrt{2}}, \tag{102}$$

$$\hat{\mu}_y = i\frac{\hat{\mu}^{(1)}_{-1} + \hat{\mu}^{(1)}_{+1}}{\sqrt{2}}, \tag{103}$$

$$\hat{\mu}_z = \hat{\mu}^{(1)}_0. \tag{104}$$

```
MagDipoleMatrixAssembly::usage = "MagDipoleMatrixAssembly[numE]
    returns the matrix representation of the operator - 1/2 (L + gs S)
     in the f^numE configuration. The function returns a list with
    three elements corresponding to the x,y,z components of this
    operator. The option ''FilenameAppendix'' can be used to append a
    string to the filename from which the function imports from in
```

```
    order to patch together the array. For numE beyond 7 the function
    returns the same as for the complementary configuration. The
    option ''ReturnInBlocks'' can be used to return the matrices in
    blocks. The default is to return the matrices in flattened form
    and as sparse array.";
Options[MagDipoleMatrixAssembly]={
  "FilenameAppendix"->"",
  "ReturnInBlocks"->False};
MagDipoleMatrixAssembly[nf_Integer, OptionsPattern[]] := Module[
  {ImportFun, numE, appendTo,
  emFname, JJBlockMagDipTable,
  Js, howManyJs, blockOp,
  rowIdx, colIdx},
  (
    ImportFun = ImportMZip;
    numE      = nf;
    numH      = 14 - numE;
    numE      = Min[numE,numH];

    appendTo  = (OptionValue["FilenameAppendix"]<>"-magDip");
    emFname   = JJBlockMatrixFileName[numE,"FilenameAppendix"->
    appendTo];
    JJBlockMagDipTable = ImportFun[emFname];

    Js        = AllowedJ[numE];
    howManyJs = Length[Js];
    blockOp   = ConstantArray[0,{howManyJs,howManyJs}];
    Do[
      blockOp[[rowIdx,colIdx]] = JJBlockMagDipTable[{numE,Js[[rowIdx
    ]],Js[[colIdx]]}],
      {rowIdx,1,howManyJs},
      {colIdx,1,howManyJs}
    ];
    If[OptionValue["ReturnInBlocks"],
      (
        opMinus = Map[#[[1]]&, blockOp, {4}];
        opZero  = Map[#[[2]]&, blockOp, {4}];
        opPlus  = Map[#[[3]]&, blockOp, {4}];
        opX =   (opMinus - opPlus)/Sqrt[2];
        opY = I (opPlus + opMinus)/Sqrt[2];
        opZ = opZero;
      ),
        blockOp = ArrayFlatten[blockOp];
        opMinus = blockOp[[;; , ;; , 1]];
        opZero  = blockOp[[;; , ;; , 2]];
        opPlus  = blockOp[[;; , ;; , 3]];
        opX =   (opMinus - opPlus)/Sqrt[2];
        opY = I (opPlus + opMinus)/Sqrt[2];
        opZ = opZero;
    ];
    Return[{opX, opY, opZ}];
  )
];
```

Using the cartesian components of the magnetic dipole operator, the matrix elements of the Zeeman term can then be evaluated. This term can be included in the Hamiltonian through an option in `HamMatrixAssembly`. Since the magnetic dipole operator is calculated in atomic units, and it seeming desirable that the input units of the magnetic field be Tesla, a conversion factor is included so that the final terms be congruent with the energy units assumed in the other terms in the Hamiltonian, namely the energy pseudo-unit Kayser (cm^{-1}). The conversion factor is called `teslaToKayser` in the file `qonstants.m`.

3.11 Alternative operator bases

One feature from the operators used in &qn-1 is that when data is fit to this model, the parameters are correlated. This has the consequence that using a partial set of operators (say those describing the free-ion part) results in parameter values that change noticeably (by perhaps 10%) when additional interactions are brought into the analysis. The semi-empirical Hamiltonian as written in &qn-1 can be described as a linear combination of a basis set of operators. Correlations in fitted parameters may be removed by using a different operator basis, with this having the added benefit or reducing parameters uncertainties [New82]. This removal of correlations is achieved by making the basis operators

pair-wise orthogonal between themselves.[18]

The operators $\hat{f}_k$, $\hat{L}^2$, $\hat{C}(\mathcal{G}_2)$, $\hat{C}(\mathcal{SO}(7))$, and $\hat{t}_2$ can be made orthogonal among themselves as prescribed by Judd, Crosswhite, and Suskin [JC84; JS84]. In there the change in the operator basis has an accompanying relationship between the coefficients in the old and the new bases, as given below. (Note the n dependence on the equation for $E_\perp^{(3)}$.)

$$E_\perp^{(1)} = \frac{4\alpha}{5} + \frac{\beta}{30} + \frac{\gamma}{25} + \frac{14F^{(2)}}{405} + \frac{7F^{(4)}}{297} + \frac{350F^{(6)}}{11583} \tag{105}$$

$$E_\perp^{(2)} = \frac{F^{(2)}}{2025} - \frac{F^{(4)}}{3267} + \frac{175F^{(6)}}{1656369} \tag{106}$$

$$E_\perp^{(3)} = -\frac{2\alpha}{5} + \frac{F^{(2)}}{135} + \frac{2F^{(4)}}{1089} - \frac{175F^{(6)}}{42471} + \frac{nT^{(2)}}{70\sqrt{2}} - \frac{T^{(2)}}{35\sqrt{2}} \tag{107}$$

$$\alpha_\perp = \frac{4\alpha}{5} \tag{108}$$

$$\beta_\perp = -4\alpha - \frac{\beta}{6} \tag{109}$$

$$\gamma_\perp = \frac{8\alpha}{5} + \frac{\beta}{15} + \frac{2\gamma}{25} \tag{110}$$

$$T_\perp^{(2)} = T^{(2)} \tag{111}$$

(In general, if a new operator basis $\hat{O}'$ is defined by a linear transformation $\mathcal{M}$ of the original basis $\hat{O}$ such that $\hat{O}' = \mathcal{M}\hat{O}$, then for a given linear combination of the original operator basis, $\mathcal{H} = \vec{\eta} \cdot \hat{O}$, a new linear combination $\vec{\eta}'$ of the new operator basis $\hat{O}'$, can be defined such that $\vec{\eta} \cdot \hat{O} = \vec{\eta}' \cdot \hat{O}'$ by taking $\vec{\eta}' = (\mathcal{M}^{-1})^T \vec{\eta}$.)

Using these coefficients and their accompanying operators, the semi-empirical Hamiltonian is now

$$\begin{aligned}\hat{\mathcal{H}}_{\text{eff}}^\perp = \hat{\mathcal{H}}_0 &+ \sum_{k=0,2,4,6} E_\perp^{(k)} \hat{e}_k^\perp + \zeta \sum_{i=1}^{N} \left(\hat{s}_i \cdot \hat{l}_i\right) \\ &+ \alpha_\perp \hat{\alpha}^\perp + \beta_\perp \hat{\beta}^\perp + \gamma_\perp \hat{\gamma}^\perp \\ &+ T_\perp^{(2)} \hat{t}_2^\perp + \sum_{\substack{k=2,3,4,6,7,8,\\ 11,12,14,15,\\ 16,17,18,19}} T^{(k)} \hat{t}_k \\ &+ \sum_{k=2,4,6} P^{(k)} \hat{p}_k + \sum_{k=0,2,4} \mathcal{M}^{(k)} \hat{m}_k \\ &+ \sum_{i=1}^{N} \sum_{k=2,4,6} \sum_{q=-k}^{k} \mathcal{B}_q^{(k)} \mathcal{C}_q^{(k)}(i).\end{aligned} \tag{112}$$

Some operators remain unchanged in this parametrization, specifically $\hat{C}_q^k$, $\hat{t}_k$ ($k \neq 2$), and the spin-orbit term. However some operators are still non-orthogonal. Operators from different *families* are mutually orthogonal, and all pairs within each family are orthogonal as well. Nevertheless, any two operators from either $\hat{m}_k$ or $\hat{p}_k$ are non-orthogonal. This residual non-orthogonality in the Hamiltonian can be resolved using the $\hat{z}_i$ operators, originally introduced by Judd in his discussion of intra-atomic magnetic interactions [JCC68]. This parametrization, which leaves $\hat{m}_k$ or $\hat{p}_k$ as they are, is therefore not entirely orthogonal, and we refer to it as the *mostly*-orthogonal Hamiltonian.

In **qlanth** the symbolic array that represents the *mostly*-orthogonal Hamiltonian can be obtained with the adequate setting of the option `OperatorBasis` in `HamMatrixAssembly`. In that option, the standard operator basis (i.e. the one use by Carnall *et al.* [Car+89]) is termed the *legacy* basis.

```
1 HamMatrixAssembly::usage = "HamMatrixAssembly[numE] returns the
    Hamiltonian matrix for the f^n_i configuration. The matrix is
    returned as a SparseArray.
2 The function admits an optional parameter ''FilenameAppendix'' which
    can be used to modify the filename to which the resulting array is
     exported to.
```

[18] Two operators $\hat{\mathcal{O}}_1$ and $\hat{\mathcal{O}}_2$ are orthogonal if $\text{tr}(\hat{\mathcal{O}}_1^\dagger \hat{\mathcal{O}}_2) = 0$.

```
It also admits an optional parameter ''IncludeZeeman'' which can be used to include the Zeeman interaction. The default is False.
The option ''Set t2Switch'' can be used to toggle on or off setting the t2 selector automatically or not, the default is True, which replaces the parameter according to numE.
The option ''ReturnInBlocks'' can be used to return the matrix in block or flattened form. The default is to return it in flattened form.";
Options[HamMatrixAssembly] = {
      "FilenameAppendix" -> "",
      "IncludeZeeman"    -> False,
      "Set t2Switch"     -> True,
      "ReturnInBlocks"   -> False,
      "OperatorBasis"    -> "Legacy"};
HamMatrixAssembly[nf_, OptionsPattern[]] := Module[
  {numE, ii, jj, howManyJs, Js, blockHam, opBasis},
  (
    (*######################################*)
    ImportFun = ImportMZip;
    opBasis = OptionValue["OperatorBasis"];
    If[Not[MemberQ[{"Legacy", "MostlyOrthogonal", "Orthogonal"}, opBasis]],
      Print["Operator basis ", opBasis, " not recognized, using ''Legacy'' basis."];
      opBasis = "Legacy";
    ];
    If[opBasis == "Orthogonal",
      Print["Operator basis ''Orthogonal'' not implemented yet, aborting ..."];
      Return[Null];
    ];
    (*######################################*)
    If[opBasis == "MostlyOrthogonal",
      (
        blockHam = HamMatrixAssembly[nf,
        "OperatorBasis" -> "Legacy",
        "FilenameAppendix" -> OptionValue["FilenameAppendix"],
        "IncludeZeeman" -> OptionValue["IncludeZeeman"],
        "Set t2Switch" -> OptionValue["Set t2Switch"],
        "ReturnInBlocks" -> OptionValue["ReturnInBlocks"]];
        paramChanger = Which[
          nf < 7,
          <|
            F0 -> 1/91 (54 E1p+91 E0p+78 γp),
            F2 -> (15/392 *
              (
                140   E1p  +
                20020 E2p +
                1540  E3p  +
                770 αp -
                70 γp  +
                22 Sqrt[2] T2p -
                11 Sqrt[2] nf T2p t2Switch -
                11 Sqrt[2] (14-nf) T2p (1 - t2Switch)
              )
            ),
            F4 -> (99/490 *
              (
                70   E1p -
                9100 E2p +
                280  E3p +
                140 αp -
                35 γp  +
                4 Sqrt[2] T2p -
                2 Sqrt[2] nf T2p t2Switch -
                2 Sqrt[2] (14-nf) T2p (1-t2Switch)
              )
              ),
            F6 -> (5577/7000 *
              (
                20  E1p +
                700 E2p -
                140 E3p -
                70 αp -
                10 γp -
                2 Sqrt[2] T2p +
```

```
71                Sqrt[2] nf T2p t2Switch +
72                Sqrt[2] (14-nf) T2p (1-t2Switch)
73              )
74              ),
75            ζ  -> ζ,
76            α -> (5 αp)/4,
77            β  -> -6  (5 αp + βp),
78            γ -> 5/2 (2 βp + 5 γp),
79            T2 -> 0
80          |>,
81        nf >= 7,
82          <|
83            F0 -> 1/91 (54 E1p+91 E0p+78 γp),
84            F2 -> (15/392 *
85              (
86                140   E1p  +
87                20020 E2p +
88                1540  E3p  +
89                770 αp -
90                70 γp  +
91                22 Sqrt[2] T2p -
92                11 Sqrt[2] nf T2p
93              )
94            ),
95            F4 -> (99/490 *
96              (
97                70   E1p -
98                9100 E2p +
99                280  E3p +
100               140 αp -
101               35 γp  +
102               4 Sqrt[2] T2p -
103               2 Sqrt[2] nf T2p
104             )
105             ),
106           F6 -> (5577/7000 *
107             (
108               20  E1p +
109               700 E2p -
110               140 E3p -
111               70 αp -
112               10 γp -
113               2 Sqrt[2] T2p +
114               Sqrt[2] nf T2p
115             )
116             ),
117           ζ  -> ζ,
118           α -> (5 αp)/4,
119           β  -> -6  (5 αp + βp),
120           γ -> 5/2 (2 βp + 5 γp),
121           T2 -> 0
122         |>
123       ];
124       blockHamM0 = Which[
125         OptionValue["ReturnInBlocks"] == False,
126         ReplaceInSparseArray[blockHam, paramChanger],
127         OptionValue["ReturnInBlocks"] == True,
128         Map[ReplaceInSparseArray[#, paramChanger]&, blockHam, {2}]
129       ];
130       Return[blockHamM0];
131     )
132   ];
133   (*########################################*)
134
135   (*hole-particle equivalence enforcement*)
136   numE = nf;
137   allVars = {E0, E1, E2, E3, ζ, F0, F2, F4, F6, M0, M2, M4, T2, T2p
      ,
138     T3, T4, T6, T7, T8, P0, P2, P4, P6, gs,
139     α, β, γ, B02, B04, B06, B12, B14, B16,
140     B22, B24, B26, B34, B36, B44, B46, B56, B66, S12, S14, S16, S22
      ,
141     S24, S26, S34, S36, S44, S46, S56, S66, T11p, T12, T14, T15,
      T16,
142     T17, T18, T19, Bx, By, Bz};
143   params0 = AssociationThread[allVars, allVars];
```

```
    If[nf > 7,
      (
        numE = 14 - nf;
        params = HoleElectronConjugation[params0];
        If[OptionValue["Set t2Switch"], params[t2Switch] = 0];
      ),
      params = params0;
      If[OptionValue["Set t2Switch"], params[t2Switch] = 1];
    ];
    (* Load symbolic expressions for LS,J,J' energy sub-matrices. *)
    emFname = JJBlockMatrixFileName[numE, "FilenameAppendix" ->
    OptionValue["FilenameAppendix"]];
    JJBlockMatrixTable = ImportFun[emFname];
    (*Patch together the entire matrix representation using J,J'
    blocks.*)
    PrintTemporary["Patching JJ blocks ..."];
    Js         = AllowedJ[numE];
    howManyJs  = Length[Js];
    blockHam   = ConstantArray[0, {howManyJs, howManyJs}];
    Do[
      blockHam[[jj, ii]] = JJBlockMatrixTable[{numE, Js[[ii]], Js[[jj
    ]]}];,
    {ii, 1, howManyJs},
    {jj, 1, howManyJs}
    ];

    (* Once the block form is created flatten it *)
    If[Not[OptionValue["ReturnInBlocks"]],
      (blockHam = ArrayFlatten[blockHam];
      blockHam  = ReplaceInSparseArray[blockHam, params];
      ),
      (blockHam  = Map[ReplaceInSparseArray[#, params]&,blockHam
    ,{2}];)
    ];

    If[OptionValue["IncludeZeeman"],
      (
        PrintTemporary["Including Zeeman terms ..."];
        {magx, magy, magz} = MagDipoleMatrixAssembly[numE, "
    ReturnInBlocks" -> OptionValue["ReturnInBlocks"]];
        blockHam += - teslaToKayser * (Bx * magx + By * magy + Bz *
    magz);
      )
    ];
    Return[blockHam];
  )
];
```

4 Coordinate system

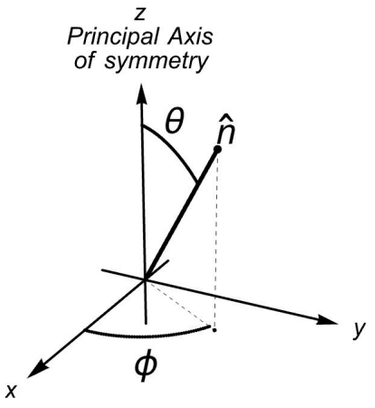
Before adding interactions lacking spherical symmetry, the orientation of the coordinate system is irrelevant. At the point when the crystal field is added, this orientation becomes relevant in the sense that only certain orientations of the coordinate system yield an expression for the crystal field potential in its simplest form[19]. To accomplish this the z-axis needs to be taken as one of the principal axis of symmetry[20]. To complete the orientation of the coordinate system, the x and y axes are chosen as symmetry axes perpendicular to the z-axis. Furthermore, certain choices for the orientation of the coordinate system also allow one to make certain crystal field parameters real, or to fix their sign.

5 Spectroscopic measurements and uncertainty

We may categorize the uncertainty in the parameters fitted to experimental data in three categories: experimental, model, and others.

Before listing the sources that contribute to experimental error, let's briefly recount the types of experiments that are used in order to determine level energies and state labels. The first type is absorption spectroscopy, in which a crystal, adequately doped, is

[19] Of course, the crystal field potential can be expressed in any rotated coordinate system, but in these the potential would include additional $C_q^{(k)}$ with linear combinations of the $B_q^{(k)}$ [20] A principal axis is a symmetry axis having the most rotational symmetry in the relevant symmetry group. For example, in cubic groups, the principal axis is the 111 diagonal.



illuminated with a broad spectrum light source, and the wavelength dependent absorption by the crystal thus determined. The crystals absorb radiation depending on the availability of a transition energy between the thermally populated low-lying states and excited levels. This data therefore provides transition energies between the ground multiplet and excited states. Furthermore, from this data one can also estimate the probability that a photon of a given wavelength is absorbed by the ions in the crystal, and from this one estimates the oscillator strength of a given transition.

In order to inform to what two multiplets the transitions belong to, here one may already count how many lines arrange themselves in groups. Alas, this type of absorption spectroscopy is lacking in that it only provides information about transitions between the ground and excited states, and may even elide such transitions that are too weak to be observed. In view of this, some variety of emission spectroscopy becomes relevant. In these, the ions inside of the crystal are excited (thermally, electrically, or radiatively) and the light produced by the relaxing transitions are then registered. This has the benefit that one has now populated other states than the ground state (perhaps with aid of non-radiative transitions inside of the crystal or energy transfer) so that now one can also have information about transitions that depart from a state different than ground. From this type of spectroscopy, given a transition, one may also determine its transition rate, given the availability of time-resolved emission; given this one may then give an upper bound on the spontaneous rate of identified transitions.

In these analyses a few things may not go according to plan:

1. **Several non-equivalent symmetry sites**. Ions may not be located in sites with the same crystal symmetry. As such it will be problematic to interpret their crystal splittings based on the assumption of a single symmetry.

2. **Non-homogeneous crystal field**. Even if they are located in sites with the same point symmetry, it may also be that the crystal field they experience has variations across the bulk of the crystal. As such, the observations would then rather be about an ensemble of crystal fields, instead of a single one.

3. **Crystal impurities**. The doped crystals may contain impurities that will lead to the false identification of transitions to the ion of study. This may be disambiguated from pooling together several experiments.

4. **Non-radiative transitions**, mediated by the crystal, will lead to shifts in transition energies, both in emission, and in absorption. This yields a confounding factor for the *radiative* transition energies that are in principle required to be valid inputs to the model Hamiltonian. Comparison of emission and absorption lines is key to determine the relevance of this.

5. **Spectrometer resolution**. The spectrometers used have a finite resolution. In the setups typically used for this, the nominal resolution might be of the order of 0.1 nm.

6. **Crystal transparency**. Observation of transitions within the ions requires that the crystal be mostly transparent at the relevant wavelengths.

In the works of Carnall and others, the nominal uncertainty in the state energies is of $1\mathcal{K}$, this being the precision to which the used experimental energies are quoted.

With regards to model uncertainties, the following factors may be considered to contribute to it:

1. **Intra-configuration transitions**. When energy levels reach a certain threshold, observations may no longer be intra-configuration transitions, but rather inter-configuration transitions. These transitions should not be included, so care must be taken to exclude them from the analysis.

2. **Unaccounted configuration interaction**. The model makes an attempt at describing configuration interaction effects, but this is only carried to second order in the types of considered interactions, and not all interactions are considered.

Finally, in the "others" category we have the two following:

1. **Numerical precision.** No longer relevant with modern computers, however, at the time at which some of these calculations were done, numerical precision might account for some of the discrepancies one finds when comparing current calculations to old ones.

2. **Errors in tables with reduced matrix elements**. The Crosswhite group at Argonne National Lab produced a set of tables with the reduced matrix elements of operators. However, at some point, these tables became slightly corrupted, and subsequent codes that used them carried those errors with them. In **qlanth** this problem is avoided since all reduced matrix elements are calculated from scratch.

When the model parameters are fitted to experimental data and their uncertainties are being estimated, **qlanth** offers two approaches. In the first approach a given constant uncertainty in the energy levels is assumed, this in turns determines the relevant contour of χ^2, and from this the uncertainties in the model parameters are calculated.

In an alternative approach, the uncertainties are determined *a posteriori*. The model parameters are fit to minimize the square differences between calculated energies and the experimental ones. Then, a single experimental uncertainty is assumed in all the energy levels, and taken equal to the minimum root mean square error, as taken over the available degrees of freedom. This uncertainty σ together with a chosen confidence interval p is then used to determine the contours of χ^2, which in turn determine the corresponding confidence interval in the model parameters. In a sense, the model is assumed to be valid, and the resulting uncertainties in the model parameters are adjusted to allow for this possibility.

In this dissertation the uncertainty in the experimental data was assumed to be constant and equal to $1\,\mathrm{cm}^{-1}$. And when the data for magnetic dipole transitions was calculated, an uncertainty equal to the σ of the related parametric fit was assumed.

6 Transitions

qlanth can also compute magnetic dipole transition rates within states and levels, as well as forced electric dipole transition rates between levels.

6.1 State description

6.1.1 Magnetic dipole transitions

qlanth can also calculate a few quantities related to magnetic dipole transitions. With $\hat{\mu} = \{\hat{\mu}_x, \hat{\mu}_y, \hat{\mu}_z\}$ the magnetic dipole operator, the line strength between two eigenstates $|\nu\rangle$ and $|\nu'\rangle$ is defined as (see for example equation 14.31 in [Cow81])

$$\hat{S}(\psi, \psi') \coloneqq \left|\langle\psi|\hat{\mu}|\psi'\rangle\right|^2 = \left|\langle\psi|\hat{\mu}_x|\psi'\rangle\right|^2 + \left|\langle\psi|\hat{\mu}_y|\psi'\rangle\right|^2 + \left|\langle\psi|\hat{\mu}_z|\psi'\rangle\right|^2 \tag{113}$$

In **qlanth** this is computed with the function `MagDipLineStrength`, which given a set of eigenvectors computes the sum above, and returns an array that contains all possible pairings of $|\psi\rangle$ and $|\psi'\rangle$ in $\hat{S}(\psi, \psi')$.

```
MagDipLineStrength::usage = "MagDipLineStrength[theEigensys, numE]
    takes the eigensystem of an ion and the number numE of f-electrons
     that correspond to it and calculates the line strength array Stot
    .
The option ''Units'' can be set to either ''SI'' (so that the units
    of the returned array are (A m^2)^2) or to ''Hartree''.
The option ''States'' can be used to limit the states for which the
    line strength is calculated. The default, All, calculates the line
     strength for all states. A second option for this is to provide
    an index labelling a specific state, in which case only the line
    strengths between that state and all the others are computed.
```

```
The returned array should be interpreted in the eigenbasis of the
    Hamiltonian. As such the element Stot[[i,i]] corresponds to the
    line strength states between states |i> and |j>.";
Options[MagDipLineStrength]={"Reload MagOp" -> False, "Units"->"SI",
    "States" -> All};
MagDipLineStrength[theEigensys_List, numE0_Integer, OptionsPattern[]]
     := Module[
  {numE, allEigenvecs, Sx, Sy, Sz, Stot, factor},
  (
    numE = Min[14-numE0, numE0];
    (*If not loaded then load it, *)
    If[Or[
        Not[MemberQ[Keys[magOp], numE]],
        OptionValue["Reload MagOp"]],
    (
      magOp[numE] = ReplaceInSparseArray[#, {gs->2}]& /@
    MagDipoleMatrixAssembly[numE];
    )
    ];
    allEigenvecs = Transpose[Last /@ theEigensys];
    Which[OptionValue["States"] === All,
      (
        {Sx, Sy, Sz} = (ConjugateTranspose[allEigenvecs].#.
    allEigenvecs) & /@ magOp[numE];
        Stot         = Abs[Sx]^2+Abs[Sy]^2+Abs[Sz]^2;
      ),
      IntegerQ[OptionValue["States"]],
      (
        singleState  = theEigensys[[OptionValue["States"],2]];
        {Sx, Sy, Sz} = (ConjugateTranspose[allEigenvecs].#.
    singleState) & /@ magOp[numE];
        Stot         = Abs[Sx]^2+Abs[Sy]^2+Abs[Sz]^2;
      )
    ];
    Which[
      OptionValue["Units"] == "SI",
        Return[4 \[Mu]B^2 * Stot],
      OptionValue["Units"] == "Hartree",
        Return[Stot],
      True,
      (
        Print["Invalid option for ''Units''. Options are ''SI'' and
    ''Hartree''."];
        Abort[];
      )
    ];
  )
];
```

Using the line strength $\hat{\mathcal{S}}$, the transition rate $\mathrm{A_{MD}}$ for the spontaneous transition $|\psi_i\rangle \rightsquigarrow |\psi_f\rangle$ is then given by (from table 7.3 of [TLJ99])

$$\mathrm{A_{MD}}\left(|\psi_i\rangle \rightsquigarrow |\psi_f\rangle\right) = \frac{16\pi^3\mu_0}{3h}\frac{n^3}{\lambda_{if}^3}\frac{\hat{\mathcal{S}}(\psi_i,\psi_f)}{g_i}, \tag{114}$$

where λ is the vacuum-equivalent wavelength of the transition between $|\nu\rangle$ and $|\nu'\rangle$, n the refractive index of the medium containing the ion, and g_i the degeneracy of the initial state $|\psi_i\rangle$. At the state level of description, J is no longer a good quantum number so the degeneracy $g_i = 1$.

```
MagDipoleRates::usage = "MagDipoleRates[eigenSys, numE] calculates
    the magnetic dipole transition rate array for the provided
    eigensystem. The option ''Units'' can be set to ''SI'' or to ''
    Hartree''. If the option ''Natural Radiative Lifetimes'' is set to
     true then the reciprocal of the rate is returned instead.
    eigenSys is a list of lists with two elements, in each list the
    first element is the energy and the second one the corresponding
    eigenvector.
Based on table 7.3 of Thorne 1999, using g2=1.
The energy unit assumed in eigenSys is kayser.
The returned array should be interpreted in the eigenbasis of the
    Hamiltonian. As such the element AMD[[i,i]] corresponds to the
    transition rate (or the radiative lifetime, depending on options)
    between eigenstates |i> and |j>.
```

```
By default this assumes that the refractive index is unity, this may be changed by setting the option ''RefractiveIndex'' to the desired value.
The option ''Lifetime'' can be used to return the reciprocal of the transition rates. The default is to return the transition rates.";
Options[MagDipoleRates]={"Units"->"SI", "Lifetime"->False, "RefractiveIndex"->1};
MagDipoleRates[eigenSys_List, numE0_Integer,OptionsPattern[]] := Module[
  {AMD, Stot, eigenEnergies,
  transitionWaveLengthsInMeters, nRefractive},
  (
    nRefractive    = OptionValue["RefractiveIndex"];
    numE           = Min[14-numE0, numE0];
    Stot           = MagDipLineStrength[eigenSys, numE, "Units" -> OptionValue["Units"]];
    eigenEnergies = Chop[First/@eigenSys];
    energyDiffs    = Outer[Subtract, eigenEnergies, eigenEnergies];
    energyDiffs    = ReplaceDiagonal[energyDiffs, Indeterminate];
    (* Energies assumed in kayser.*)
    transitionWaveLengthsInMeters = 0.01/energyDiffs;

    unitFactor     = Which[
    OptionValue["Units"]=="Hartree",
    (
      (* The bohrRadius factor in SI needed to convert the wavelengths which are assumed in m*)
      16 \[Pi]^3 (\[Mu]0Hartree /(3 hPlanckFine)) * bohrRadius^3
    ),
    OptionValue["Units"]=="SI",
    (
      16 \[Pi]^3 \[Mu]0/(3 hPlanck)
    ),
    True,
    (
      Print["Invalid option for ''Units''. Options are ''SI'' and ''Hartree''."];
      Abort[];
    )
    ];
    AMD = unitFactor / transitionWaveLengthsInMeters^3 * Stot * nRefractive^3;
    Which[OptionValue["Lifetime"],
      Return[1/AMD],
      True,
      Return[AMD]
    ]
  )
];
```

A final quantity of interest is the oscillator strength for the transition between the ground state $|\psi_g\rangle$ and an excited state $|\psi_e\rangle$. The oscillator strength is a dimensionless quantity which is indicative of how strong absorption is. The oscillator strength may be defined for other initial states than the ground state, but since this is the state most likely to be populated in ordinary experimental conditions, this is the initial state that is of most frequent interest. The oscillator strength is given by [CFW65]

$$\mathrm{f_{MD}}\left(|\psi_g\rangle \rightsquigarrow |\psi_e\rangle\right) = \frac{8\pi^2 m_e}{3\,hc\,e^2}\frac{n}{\lambda_{ge}}\frac{\hat{\mathcal{S}}(\psi_g,\psi_e)}{g_g} \tag{115}$$

where g_g is the degeneracy of the ground state. At the level of detail that the eigenstates are described in **qlanth** where J is no longer a good quantum number, $g_g = 1$.

In **qlanth** the function GroundMagDipoleOscillatorStrength implements the calculation of the oscillator strengths from the ground state to all the excited ones.

```
GroundMagDipoleOscillatorStrength::usage = "
    GroundMagDipoleOscillatorStrength[eigenSys, numE] calculates the magnetic diople oscillator strengths between the ground state and the excited states as given by eigenSys.
Based on equation 8 of Carnall 1965, removing the 2J+1 factor since this degeneracy has been removed by the crystal field.
eigenSys is a list of lists with two elements, in each list the first element is the energy and the second one the corresponding eigenvector.
The energy unit assumed in eigenSys is Kayser.
```

```
The oscillator strengths are dimensionless.
The returned array should be interpreted in the eigenbasis of the
    Hamiltonian. As such the element fMDGS[[i]] corresponds to the
    oscillator strength between ground state and eigenstate |i>.
By default this assumes that the refractive index is unity, this may
    be changed by setting the option ''RefractiveIndex'' to the
    desired value.";
Options[GroundMagDipoleOscillatorStrength]={"RefractiveIndex"->1};
GroundMagDipoleOscillatorStrength[eigenSys_List, numE_Integer,
    OptionsPattern[]] := Module[
  {eigenEnergies, SMDGS, GSEnergy, energyDiffs,
  transitionWaveLengthsInMeters, unitFactor, nRefractive},
  (
    eigenEnergies    = First/@eigenSys;
    nRefractive      = OptionValue["RefractiveIndex"];
    SMDGS            = MagDipLineStrength[eigenSys,numE, "Units"->"SI
    ", "States"->1];
    GSEnergy         = eigenSys[[1,1]];
    energyDiffs      = eigenEnergies-GSEnergy;
    energyDiffs[[1]] = Indeterminate;
    transitionWaveLengthsInMeters = 0.01/energyDiffs;
    unitFactor       = (8\[Pi]^2 me)/(3 hPlanck eCharge^2 cLight);
    fMDGS            = unitFactor / transitionWaveLengthsInMeters *
    SMDGS * nRefractive;
    Return[fMDGS];
  )
];
```

6.2 Level description

6.2.1 Forced electric dipole transitions

Any two eigenfunctions that are approximated within the limits of a single configuration cannot help but have the same parity as they are spanned by basis vectors with definite and shared parity. Analysis of the amplitudes for different transition operators can then inform as to what transitions are forbidden, which are those in which the product of the parity of the two participating wavefunctions and that of the transition operator results in odd parity. As such, within the single configuration approximation, since the product of the two participating wavefunctions is always even, then any transition described by an operator of odd parity is forbidden. This is the content of Laporte's parity selection rule. Since the parity of the magnetic dipole operator is even [21], then this operator allows for intra-configuration transitions, and since the parity of the electric dipole operator is odd, then these types of intra-configuration transitions are forbidden.

However, much as configuration interaction is an essential component in the description of the electronic structure, it has a bearing on the energy spectrum and the intra-configuration wavefunctions themselves. Configuration interaction may also be used to bring back into the analysis the fact that the *actual* wavefunctions will also have at least a small part of them in other configurations, even if most of them may be within the ground configuration. It is therefore the case that the *actual* parity of the wavefunctions is mixed, and therefore intra-configuration [22] electric dipole transitions are actually allowed. These electric dipole transitions are called *forced* electric dipole transitions.

Judd [Jud62] and Ofelt [Ofe62] came separately to similar versions of this analysis, and showed after a series of approximations that the forced electric dipole transitions could be described by the intra-configuration matrix elements of the multi-electron unit operators $\hat{U}^{(k)}$ (for k=2,4,6) together with a set of three accompanying coefficients $\{\Omega_{(2)}, \Omega_{(4)}, \Omega_{(6)}\}$. These coefficients have a definite form related to the overlap between the mixed parity parts of the corrected wavefunctions, but they can also be considered as additional phenomenological parameters.

Judd-Ofelt theory is based on the level description, and its mathematical expression is the following. Given two intermediate coupling levels $|\alpha SLJ\rangle$ and $|\alpha' S'L'J'\rangle$, the oscillator strength between them is approximated as [Jud62]

$$\mathrm{f}_{\text{f-ED}}\left(|\alpha LSJ\rangle \rightsquigarrow |\alpha' S'L'J'\rangle\right) = \mathcal{R}\frac{8\pi^2 m_e}{3h}\frac{\nu}{2J+1}\frac{\chi}{n}\sum_{k=2,4,6}\Omega_{(k)}\left|\langle \underline{\mathrm{f}}^n \alpha SLJ \| \hat{U}^{(k)} \| \underline{\mathrm{f}}^n \alpha' S'L'J'\rangle\right|^2, \tag{116}$$

[21] The parity of the electric quadrupole operator is also even, but we haven't included it in **qlanth**

[22] Calling these *intra*-configuration transitions is somewhat of a misnomer since their nature is tied to the fact that the single-configuration description is wanting.

where ν is the frequency of the transition, χ the local field correction, n the refractive index of the crystal host, and $\mathcal{R} = 1$ in the case of absorption and $\mathcal{R} = n^2$ in the case of emission.

The local field correction χ accounts for the difference between the macroscopic and microscopic electric fields, in the case of ions embedded for crystals the most common choice is

$$\chi = \frac{n^2 + 2}{3} \tag{117}$$

and for other environments (or emitters other than ions such as molecules) different alternatives are relevant (see [DR06]).

In **qlanth** Judd-Ofelt theory is implemented with help of the functions JuddOfeltUkSquared and LevelElecDipoleOscillatorStrength.

```
JuddOfeltUkSquared::usage = "JuddOfeltUkSquared[numE, params]
    calculates the matrix elements of the Uk operator in the level
    basis. These are calculated according to equation (7) in Carnall
    1965.
The function returns a list with the following elements:
  - basis : A list with the allowed {SL, J} terms in the f^numE
    configuration. Equal to BasisLSJ[numE].
  - eigenSys : A list with the eigensystem of the Hamiltonian for the
     f^n configuration.
  - levelLabels : A list with the labels of the major components of
    the level eigenstates.
  - LevelUkSquared : An association with the squared matrix elements
    of the Uk operators in the level eigenbasis. The keys being {2, 4,
     6} corresponding to the rank of the Uk operator. The basis in
    which the matrix elements are given is the one corresponding to
    the level eigenstates given in eigenSys and whose major SLJ
    components are given in levelLabels. The matrix is symmetric and
    given as a SymmetrizedArray.
  The function admits the following options:
  ''PrintFun'' : A function that will be used to print the progress
    of the calculations. The default is PrintTemporary.";
Options[JuddOfeltUkSquared] = {"PrintFun" -> PrintTemporary};
JuddOfeltUkSquared[numE_, params_, OptionsPattern[]] := Module[
  {eigenChanger, numEH, basis, eigenSys,
  Js, Ukmat, LevelUkSquared, kRank,
  S, L, Sp, Lp, J, Jp, phase,
  braTerm, ketTerm, levelLabels,
  eigenVecs, majorComponentIndices},
  (
    If[Not[ValueQ[ReducedUkTable]],
      LoadUk[]
    ];
    numEH = Min[numE, 14-numE];
    PrintFun = OptionValue["PrintFun"];
    PrintFun["> Calculating the levels for the given parameters ..."
    ];
    {basis, majorComponents, eigenSys} = LevelSolver[numE, params];
    (* The change of basis matrix to the eigenstate basis *)
    eigenChanger = Transpose[Last /@ eigenSys];
    PrintFun["Calculating the matrix elements of Uk in the physical
    coupling basis ..."];
    LevelUkSquared = <||>;
    Do[(
      Ukmat = Table[(
          {S, L}   = FindSL[braTerm[[1]]];
          J        = braTerm[[2]];
          Jp       = ketTerm[[2]];
          {Sp, Lp} = FindSL[ketTerm[[1]]];
          phase    = Phaser[S + Lp + J + kRank];
          Simplify @ (
            phase *
            Sqrt[TPO[J]*TPO[Jp]] *
            SixJay[{J, Jp, kRank}, {Lp, L, S}] *
            ReducedUkTable[{numEH, 3, braTerm[[1]], ketTerm[[1]],
    kRank}]
          )
        ),
        {braTerm, basis},
        {ketTerm, basis}
      ];
      Ukmat = (Transpose[eigenChanger] . Ukmat . eigenChanger)^2;
```

```
      Ukmat = Chop@Ukmat;
      LevelUkSquared[kRank] = SymmetrizedArray[Ukmat, Dimensions[
    eigenChanger], Symmetric[{1, 2}]];
      ),
      {kRank, {2, 4, 6}}
    ];
    LSJmultiplets = (RemoveTrailingDigits[#[[1]]] <> ToString[
    InputForm[#[[2]]]]) & /@ basis;
    eigenVecs = Last /@ eigenSys;
    majorComponentIndices = Ordering[Abs[#]][[-1]] & /@ eigenVecs;
    levelLabels = LSJmultiplets[[majorComponentIndices]];
    Return[{basis, eigenSys, levelLabels, LevelUkSquared}];
  )
];
```

```
LevelElecDipoleOscillatorStrength::usage = "
    LevelElecDipoleOscillatorStrength[numE, levelParams,
    juddOfeltParams] uses Judd-Ofelt theory to estimate the forced
    electric dipole oscillator strengths ions whose level description
    is determined by levelParams.
The third parameter juddOfeltParams is an asssociation with keys
    equal to the three Judd-Ofelt intensity parameters {\[CapitalOmega
    ]2, \[CapitalOmega]4, \[CapitalOmega]6} and corresponding values
    in cm^2.
The local field correction implemented here corresponds to the one
    given by the virtual cavity model of Lorentz.
The function returns a list with the following elements:
  - basis : A list with the allowed {SL, J} terms in the f^numE
    configuration. Equal to BasisLSJ[numE].
  - eigenSys : A list with the eigensystem of the Hamiltonian for the
     f^n configuration in the level description.
  - levelLabels : A list with the labels of the major components of
    the calculated levels.
  - oStrengthArray : A square array whose elements represent the
    oscillator strengths between levels such that the element
    oStrengthArray[[i,j]] is the oscillator strength between the
    levels |Subscript[\[Psi], i]> and |Subscript[\[Psi], j]>. In this
    array, the elements below the diagonal represent emission
    oscillator strengths, and elements above the diagonal represent
    absorption oscillator strengths.
The function admits the following three options:
  ''PrintFun'' : A function that will be used to print the progress
    of the calculations. The default is PrintTemporary.
  ''RefractiveIndex'' : The refractive index of the medium where the
    transitions are taking place. This may be a number or a function.
    If a number then the oscillator strengths are calculated for
    assuming a wavelength-independent refractive index. If a function
    then the refractive indices are calculated accordingly to the
    wavelength of each transition (the function must admit a single
    argument equal to the wavelength in nm). The default is 1.
  ''LocalFieldCorrection'' : The local field correction to be used.
    The default is ''VirtualCavity''. The options are: ''VirtualCavity
    '' and ''EmptyCavity''.
The equation implemented here is the one given in eqn. 29 from the
    review article of Hehlen (2013). See that same article for a
    discussion on the local field correction.
";
Options[LevelElecDipoleOscillatorStrength]={
  "PrintFun"        -> PrintTemporary,
  "RefractiveIndex" -> 1,
  "LocalFieldCorrection" -> "VirtualCavity"
};
LevelElecDipoleOscillatorStrength[numE_, levelParams_Association,
    juddOfeltParams_Association, OptionsPattern[]] := Module[
  {PrintFun, basis, eigenSys, levelLabels,
  LevelUkSquared, eigenEnergies, energyDiffs,
  oStrengthArray, nRef, \[Chi], nRefs,
  \[Chi]OverN, groundLevel, const,
  transitionFrequencies, wavelengthsInNM,
  fieldCorrectionType},
  (
    PrintFun = OptionValue["PrintFun"];
    nRef     = OptionValue["RefractiveIndex"];
    PrintFun["Calculating the Uk^2 matrix elements for the given
    parameters ..."];
    {basis, eigenSys, levelLabels, LevelUkSquared} =
```

```
    JuddOfeltUkSquared[numE, levelParams, "PrintFun"->PrintFun];
    eigenEnergies = First/@eigenSys;
    (* converted to cm^-2 *)
    const         = (8\[Pi]^2)/3 me/hPlanck * 10^(-4);
    energyDiffs   = Transpose@Outer[Subtract,eigenEnergies,
    eigenEnergies];
    (* since energies are assumed in Kayser, speed of light needs to
    be in cm/s, so that the frequencies are in 1/s *)
    transitionFrequencies = energyDiffs*cLight*100;
    (* grab the J for each level *)
    levelJs        = #[[2]] & /@ eigenSys;
    oStrengthArray = (
      juddOfeltParams[\[CapitalOmega]2]*LevelUkSquared[2]+
      juddOfeltParams[\[CapitalOmega]4]*LevelUkSquared[4]+
      juddOfeltParams[\[CapitalOmega]6]*LevelUkSquared[6]
    );
    oStrengthArray = Abs@(const * transitionFrequencies *
    oStrengthArray);
    (* it is necessary to divide each oscillator strength by the
    degeneracy of the initial level *)
    oStrengthArray = MapIndexed[1/(2 levelJs[[#2[[1]]]]+1) #1 &,
    oStrengthArray,{2}];
    (* including the effects of the refractive index *)
    fieldCorrectionType = OptionValue["LocalFieldCorrection"];
    Which[
      nRef === 1,
        True,
      NumberQ[nRef],
      (
        \[Chi] = Which[
          fieldCorrectionType == "VirtualCavity",
          (
            ( (nRef^2 + 2) / 3 )^2
          ),
          fieldCorrectionType == "EmptyCavity",
          (
            ( 3 * nRef^2 / ( 2 * nRef^2 + 1 ) )^2
          )
        ];
        \[Chi]OverN    = \[Chi] / nRef;
        oStrengthArray = \[Chi]OverN * oStrengthArray;
        (* the refractive index participates differently in
    absorption and in emission *)
        aFunction      = If[#2[[1]]>#2[[2]], #1 * nRef^2, #1]&;
        oStrengthArray = MapIndexed[aFunction, oStrengthArray, {2}];
      ),
      True,
      (
        wavelengthsInNM = Abs[1 / energyDiffs] * 10^7;
        nRefs           = Map[nRef, wavelengthsInNM];
        Print["Calculating the oscillator strengths for the given
    refractive index ..."];
        \[Chi] = Which[
          fieldCorrectionType == "VirtualCavity",
          (
            ( (nRefs^2 + 2) / 3 )^2
          ),
          fieldCorrectionType == "EmptyCavity",
          (
            ( 3 * nRefs^2 / ( 2*nRefs^2 + 1 ) )^2
          )
        ];
        \[Chi]OverN     = \[Chi] / nRefs;
        oStrengthArray  = \[Chi]OverN * oStrengthArray
      )
    ];
    Return[{basis, eigenSys, levelLabels, oStrengthArray}];
  )
];
```

6.2.2 Magnetic dipole transitions

In Hartree atomic units, the magnetic dipole line strength between levels $|\alpha LSJ\rangle$ and $|\alpha' S' L' J'\rangle$ is given by

$$\hat{\mathcal{S}}(|\alpha LSJ\rangle, |\alpha' S' L' J'\rangle) = \left| \langle \alpha LSJ \| \frac{1}{2}(\hat{L} + g\hat{S}) \| \alpha' S' L' J' \rangle \right|^2 \tag{118}$$

In **qlanth** the line strength can be calculated using the function LevelMagDipoleLineStrength.

```
LevelMagDipoleLineStrength::usage = "LevelMagDipoleLineStrength[
    eigenSys, numE] calculates the magnetic dipole line strengths for
    an ion whose level description is determined by levelParams. The
    function returns a square array whose elements represent the
    magnetic dipole line strengths between the levels given in
    eigenSys such that the element magDipoleLineStrength[[i,j]] is the
     line strength between the levels |Subscript[\[Psi], i]> and |
    Subscript[\[Psi], j]>. Eigensys must be such that it consists of a
     lists of lists where in each list the last element corresponds to
     the eigenvector of a level (given as a row) in the standard basis
     for levels of the f^numE configuration.
The function admits the following options:
  ''Units'' : The units in which the line strengths are given. The
    default is ''SI''. The options are ''SI'' and ''Hartree''. If ''SI
    '' then the unit of the line strength is (A m^2)^2 = (J/T)^2. If
    ''Hartree'' then the line strength is given in units of 2 \[Mu]B."
    ;
Options[LevelMagDipoleLineStrength] = {
  "Units"->"SI"
  };
LevelMagDipoleLineStrength[theEigensys_List, numE0_Integer,
    OptionsPattern[]] := Module[
  {numE, levelMagOp, allEigenvecs, magDipoleLineStrength, units},
  (
    numE          = Min[14-numE0,numE0];
    levelMagOp    = LevelMagDipoleMatrixAssembly[numE];
    allEigenvecs  = Transpose[Last/@theEigensys];
    units         = OptionValue["Units"];
    magDipoleLineStrength          = Transpose[allEigenvecs].
    levelMagOp.allEigenvecs;
    magDipoleLineStrength          = Abs[magDipoleLineStrength]^2;
    Which[
      units=="SI",
        Return[4 \[Mu]B^2 * magDipoleLineStrength],
      units=="Hartree",
        Return[magDipoleLineStrength]
    ];
  )
];
```

In atomic units, the magnetic dipole oscillator strength for a transition between level $|\alpha LSJ\rangle$ and an excited level $|\alpha' S' L' J'\rangle$ is given by [Rud07]

$$\mathrm{f_{MD}}\left(|\alpha LSJ\rangle \rightsquigarrow |\alpha' S' L' J'\rangle\right) = \frac{2n}{3} \frac{\mathcal{E}(|\alpha' S' L' J'\rangle) - \mathcal{E}(|\alpha LSJ\rangle)}{2J+1} \alpha^2 \hat{\mathcal{S}}\left(|\alpha LSJ\rangle, |\alpha' S' L' J'\rangle\right) \tag{119}$$

where $\mathcal{E}(|\alpha LSJ\rangle)$ is the energy of level $|\alpha LSJ\rangle$, n is the refractive index of the medium, and α is the fine structure constant. In obtaining this expression one considers the transition from one state of the initial level into another single state of the final level. Furthermore, here it is assumed that all the states of the initial level are equally populated.

In **qlanth** the function LevelMagDipoleOscillatorStrength can be used to calculate these.

```
LevelMagDipoleOscillatorStrength::usage = "
    LevelMagDipoleOscillatorStrength[eigenSys, numE] calculates the
    magnetic dipole oscillator strengths for an ion whose level
    description is determined by levelParams. The refractive index of
    the medium is relevant, but here it is assumed to be 1, this can
    be changed through the option ''RefractiveIndex''. eigenSys must
    consist of a lists of lists with three elements: the first element
     being the energy of the level, the second element being the J of
    the level, and the third element being the eigenvector of the
    level.
  The function returns a list with the following elements:
```

```
    - basis : A list with the allowed {SL, J} terms in the f^numE
    configuration. Equal to BasisLSJ[numE].
    - eigenSys : A list with the eigensystem of the Hamiltonian for
    the f^n configuration in the level description.
    - levelLabels : A list with the labels of the major components of
     the calculated levels.
    - magDipoleOstrength : A square array whose elements represent
    the magnetic dipole oscillator strengths between the levels given
    in eigenSys such that the element magDipoleOstrength[[i,j]] is the
     oscillator strength between the levels |Subscript[\[Psi], i]> and
     |Subscript[\[Psi], j]>. In this array the elements below the
    diagonal represent emission oscillator strengths, and elements
    above the diagonal represent absorption oscillator strengths. The
    emission oscillator strengths are negative. The oscillator
    strength is a dimensionless quantity.
  The function admits the following option:
    ''RefractiveIndex'' : The refractive index of the medium where
    the transitions are taking place. This may be a number or a
    function. If a number then the oscillator strengths are calculated
     assuming a wavelength-independent refractive index as given. If a
     function then the refractive indices are calculated accordingly
    to the vaccum wavelength of each transition (the function must
    admit a single argument equal to the wavelength in nm). The
    default is 1.
  For reference see equation (27.8) in Rudzikas (2007). The
    expression for the line strenght is the simplest when using atomic
     units, (27.8) is missing a factor of \[Alpha]^2.";
Options[LevelMagDipoleOscillatorStrength]={
  "RefractiveIndex" -> 1
};
LevelMagDipoleOscillatorStrength[eigenSys_, numE_, OptionsPattern[]]
    := Module[
  {eigenEnergies, eigenVecs, levelJs,
  energyDiffs, magDipoleOstrength, nRef,
  wavelengthsInNM, nRefs, degenDivisor},
  (
    basis          = BasisLSJ[numE];
    eigenEnergies  = First/@eigenSys;
    nRef           = OptionValue["RefractiveIndex"];
    eigenVecs      = Last/@eigenSys;
    levelJs        = #[[2]]&/@eigenSys;
    energyDiffs    = -Outer[Subtract,eigenEnergies,eigenEnergies];
    energyDiffs   *= kayserToHartree;
    magDipoleOstrength = LevelMagDipoleLineStrength[eigenSys, numE, "
    Units"->"Hartree"];
    magDipoleOstrength = 2/3 * \[Alpha]Fine^2 * energyDiffs *
    magDipoleOstrength;
    degenDivisor       = #1 / ( 2 * levelJs[[#2[[1]]]] + 1 ) &;
    magDipoleOstrength = MapIndexed[degenDivisor, magDipoleOstrength,
     {2}];
    Which[nRef===1,
      True,
      NumberQ[nRef],
        (
          magDipoleOstrength = nRef * magDipoleOstrength;
        ),
      True,
        (
          wavelengthsInNM    = Abs[kayserToHartree / energyDiffs] *
    10^7;
          nRefs              = Map[nRef, wavelengthsInNM];
          magDipoleOstrength = nRefs * magDipoleOstrength;
        )
    ];
    Return[{basis, eigenSys, magDipoleOstrength}];
  )
];
```

A final quantity of interest is the spontaneous magnetic dipole decay rate from one level to a lower lying one. In atomic units this rate is determined by

$$\Gamma_{\mathrm{MD}}\left(|\alpha LSJ\rangle \rightsquigarrow |\alpha' S' L' J'\rangle\right) = \frac{4n^3}{3}\frac{\left(\mathcal{E}(|\alpha LSJ\rangle) - \mathcal{E}(|\alpha' S' L' J'\rangle)\right)^3}{2J+1}\alpha^5 \hat{\mathcal{S}}\left(|\alpha LSJ\rangle, |\alpha' S' L' J'\rangle\right). \tag{120}$$

In **qlanth** the spontaneous decay rates may be calculated through the function LevelMagDipoleSpotaneousDecayRates.

```
LevelMagDipoleSpotaneousDecayRates::usage = "
    LevelMagDipoleSpotaneousDecayRates[eigenSys, numE] calculates the
    spontaneous emission rates for the magnetic dipole transitions
    between the levels given in eigenSys. The function returns a
    square array whose elements represent the spontaneous emission
    rates between the levels given in eigenSys such that the element
    [[i,j]] of the returned array is the rate of spontaneous emission
    from the level |Subscript[\[Psi], i]> to the level |Subscript[\[
    Psi], j]>. In this array the elements below the diagonal represent
     emission rates, and elements above the diagonal are identically
    zero.
The function admits two optional arguments:
  + \"Units\" : The units in which the rates are given. The default
    is \"SI\". The options are \"SI\" and \"Hartree\". If \"SI\" then
    the rates are given in s^-1. If \"Hartree\" then the rates are
    given in the atomic unit of frequency.
  + \"RefractiveIndex\" : The refractive index of the medium where
    the transitions are taking place. This may be a number or a
    function. If a number then the rates are calculated assuming a
    wavelength-independent refractive index as given. If a function
    then the refractive indices are calculated accordingly to the
    vaccum wavelength of each transition (the function must admit a
    single argument equal to the wavelength in nm). The default is 1."
    ;
Options[LevelMagDipoleSpotaneousDecayRates] = {
  "Units" -> "SI",
  "RefractiveIndex" -> 1};
LevelMagDipoleSpotaneousDecayRates[eigenSys_List, numE_Integer,
    OptionsPattern[]] := Module[
  {
    levMDlineStrength, eigenEnergies, energyDiffs, levelJs,
    spontaneousRatesInHartree, spontaneousRatesInSI, degenDivisor,
    units,
    nRef, nRefs, wavelengthsInNM
  },
  (
    nRef               = OptionValue["RefractiveIndex"];
    units              = OptionValue["Units"];
    levMDlineStrength = LowerTriangularize@LevelMagDipoleLineStrength
    [eigenSys,numE,"Units"->"Hartree"];
    levMDlineStrength = SparseArray[levMDlineStrength];
    eigenEnergies      = First /@ eigenSys;
    energyDiffs        = Outer[Subtract, eigenEnergies, eigenEnergies
    ];
    energyDiffs        = kayserToHartree * energyDiffs;
    energyDiffs        = SparseArray[LowerTriangularize[energyDiffs]];
    levelJs            = #[[2]]& /@ eigenSys;
    spontaneousRatesInHartree = 4/3 αFine^5 * energyDiffs^3 *
    levMDlineStrength;
    degenDivisor               = #1 / (2*levelJs[[#2[[1]]]] + 1)&;
    spontaneousRatesInHartree = MapIndexed[degenDivisor,
    spontaneousRatesInHartree, {2}];
    Which[nRef===1,
      True,
      NumberQ[nRef],
        (
          spontaneousRatesInHartree = nRef^3 *
    spontaneousRatesInHartree;
        ),
      True,
        (
          wavelengthsInNM    = Abs[kayserToHartree / energyDiffs] *
    10^7;
          nRefs              = Map[nRef, wavelengthsInNM];
          spontaneousRatesInHartree = nRefs^3 *
    spontaneousRatesInHartree;
        )
    ];
    If[units == "SI",
      (
        spontaneousRatesInSI = 1/hartreeTime *
    spontaneousRatesInHartree;
        Return[SparseArray@spontaneousRatesInSI];
      ),
      Return[SparseArray@spontaneousRatesInHartree];
    ];
```

```
  )
];
```

7 Parameter constraints

When there is a scarcity of experimental data, one useful strategy to reduce the number of free parameters is to enforce some constraints between ratios of Slater integrals $F^{(k)}$, Marvin integrals $\mathcal{M}^{(k)}$, and pseudo-magnetic parameters $P^{(k)}$.

For the Slater integrals one may leave only $F^{(2)}$ as free parameter, and fix the ratios $F^{(4)}/F^{(2)}$ and $F^{(6)}/F^{(2)}$.

For the Marvin integrals one often leaves only a single free parameter $\mathcal{M}^{(0)}$, and give values to $\mathcal{M}^{(2)}$ and $\mathcal{M}^{(4)}$, by fixing the ratios $\mathcal{M}^{(2)}/\mathcal{M}^{(0)}$ and $\mathcal{M}^{(4)}/\mathcal{M}^{(0)}$.

For the pseudo-magnetic parameters again the common practice is to only leave a single free parameter $P^{(2)}$, and give values to $P^{(4)}$ and $P^{(6)}$, by fixing the ratios $P^{(4)}/P^{(2)}$ and $P^{(6)}/P^{(2)}$.

The values for all these ratios were historically obtained by using the integral expressions for the corresponding parameters, and calculating them using Hartree-Fock solutions to the radial parts of the wavefunctions. Examples of these ratios can be seen in the sections with data for LaF_3 and $LiYF_4$.

8 Fitting experimental data

qlanth also has the capacity to fit the semi-empirical Hamiltonian to experimental data. This is included in the sub-module `fittings.m` (see Appendix 17.2). This sub-module includes the function `ClassicalFit` which uses a truncated Hamiltonian (based on free-ion energies) to fit a given subset of the model parameters to given experimental data. It yields an extensive set of results, including fitted parameters and uncertainties. If the truncation energy parameter is set to infinity, then the fitting is performed with no truncation.

This function, however, is specifically used for fitting data for a single ion in a specific host. In the case of fitting data for several ions, it may be necessary to use parameter trends $\mathcal{P}(n)$. This is necessary since not only there might be some ions where there is no data (and where one would then propose a "synthetic" solution), but also since there are cases where there are too few data points to justify varying all of the model parameters.[23]

In these cases where one is fitting data for all or most of the lanthanide ions in a given host, it is useful to first fit the model in cases where there are the most data points, and to build up a parameter model $\mathcal{P}(n)$ for each parameter as the fitting of all the ions progresses. One feature often used in fitting for several ions (see Carnall *et al.* [Car+89] and Cheng *et al.* [Che+16]) is that when there is scarcity of data (as mentioned above), one can then use the trends in the $\mathcal{P}(n)$ in order to fix some parameter values at a given column (and proceed to vary others to fit the data to the model).[24]

Here below is a detailed explanation of the parameters required by `ClassicalFit`. The code for this function `ClassicalFit` may be found in Appendix 17.2.

- `numE`: number of electrons in the system, specifying the electronic configuration.
- `expData`: experimental data, a list of lists where each sublist represents an energy level and associated parameters. The first element of the sublists must represent energies, the other elements in the sublists are ignored but can be given to be kept together with the fitted data. The data must be ordered in increasing order of energy. **IMPORTANT:** if there are known unknown levels, these should be made explicit, anything other than a number will be interpreted as a level of undetermined energy in the corresponding gap. **ALSO IMPORTANT:** in the case of odd electron cases, `expData` needs to explicitly include the duplicate energies corresponding to Kramers' degeneracy; the gaps also need to be adequately duplicated in these cases.

[23] The extreme case of this scarcity being Yb and Ce, where there are only 7 non-degenerate energies, but where the crystal-field alone might require more parameters than these (for instance in C_{2v} symmetry one needs 9 $B_q^{(k)}$ parameters). [24] For example, in the Table **??** for LaF_3, in the case of Yb, only two parameters are varied (ζ and ϵ) and the values for the crystal field obtained from linear fits to the previously fitted $B_q^{(k)}$ in Pr, Nd, Dy, Sm*, Ho*, Er, and Tm. (* not all $B_q^{(k)}$)

- `excludeDataIndices`: indices in `expData` to be excluded from the fitting process. This can be used to exclude experimental data which is present, but which is considered dubious. In the case of odd electron configurations, these indices need to explicitly include the double degeneracy of Kramers doublets.

- `problemVars`: symbols representing the parameters to be fitted, some of which may be constrained (set fixed or proportional to others). **IMPORTANT:** if `problemVars` is a proper subset of all the parameters needed to evaluate the simplified Hamiltonian, the values for the other necessary parameters are taken from the Carnall *et al.* [Car+89] systematic study of LaF_3.

- `startValues`: an association with the initial values for the independent parameters given in `problemVars`. Independent parameters are those that remain once the constraints have been accounted for.

- σ`exp`: estimated uncertainty in the energy level differences between experimental and calculated values.

- `constraints`: a list of replacement rules defining constraints on the parameters. These constraints can either pin down a value, or apply proportionality ratios between them. If constrained by proportionally factors, these ratios are usually taken from Hartree-Fock calculations.

Here is a description of the different steps that this algorithm implements.

1. **Initialization**: sets initial conditions, processes options, and prepares data structures. Manages settings like the truncation energy, logging preferences, and computational accuracy goals.

2. **Data Preparation**: determines valid data points, excluding specified indices, and establishes truncation energy for the model.

3. **Hamiltonian Assembly and Simplification**: constructs the Hamiltonian while preserving its block structure, applies simplification rules, and processes the diagonal blocks to retain only free-ion parameters.

4. **Level Calculation**: determines the level description using free-ion parameters.

5. **Compilation and Truncation of Hamiltonian**: compiles the Hamiltonian and truncates it based on the set truncation energy, optimizing for computational efficiency.

6. **Fitting Process Initialization**: prepares variables and functions for optimization, including eigenvalue calculations and difference evaluations.

7. **Optimization**: employs the Levenberg-Marquardt method to optimize parameters, minimizing the discrepancy between calculated and experimental energy levels.

8. **Post-Processing**: calculates the Hamiltonian's eigensystem at the solution, deriving statistics like RMS deviation, parameter uncertainties, and covariance matrix.

9. **Output Compilation**: aggregates all relevant data and results into the output association `solCompendium`, documenting the fitting process and outcomes.

10. **Logging and Return**: saves the comprehensive fitting results to a log file and returns the detailed output data.

This function admits several options. Importantly here one may permit the model to have a constant shift to all the levels and the truncation energy can be set. Here one can also provide simplification rules that are applied to the compiled version of the Hamiltonian.

- `Energy Uncertainty in K`: used for error estimation, it can be either `Automatic` in which case the (σ of the fit is used as the uncertainty of the energies) or a numeric value in which case that is the value used for error propagation.

- `TruncationEnergy`: determines the energy level at which the Hamiltonian is truncated. If set to `Automatic`, the truncation energy is derived from the maximum energy present in the experimental data (`expData`). Otherwise, it can be manually set to a specific value.
- `MagneticSimplifier`: provides a list of replacement rules to simplify the magnetic parameters in the Hamiltonian, aiding in the reduction of computational complexity.
- `MagFieldSimplifier`: offers a list of replacement rules to specify a magnetic field, enhancing the flexibility in modeling magnetic effects within the system.
- `SymmetrySimplifier`: A list of replacement rules used to simplify the crystal field components of the Hamiltonian, facilitating a more efficient fitting process.
- `OtherSimplifier`: an additional list of replacement rules applied to the Hamiltonian before computation, allowing for further customization and simplification of the model, such as disabling specific interactions or effects. **IMPORTANT**: here the default is that the spin-spin contribution (as controlled by the σSS parameter) for the Marvin integrals is *not* included.
- `MaxHistory`: this option controls the length of the logs for the solver, enabling users to adjust the amount of log data retained during the fitting process.
- `MaxIterations`: sets the maximum number of iterations that the fitting algorithm (`NMinimize`) will execute, allowing control over the computational effort spent on the fitting.
- `FilePrefix`: specifies the prefix for the filenames under which the fitting results are saved. By default, the prefix is set to "calcs", and the files are saved in the "log/calcs" directory.
- `AddConstantShift`: if set to `True`, this option allows for a constant shift in the energy levels during the fitting process. This is particularly useful for fine-tuning the model to better match experimental data.
- `AccuracyGoal`: defines the accuracy goal for the `NMinimize` function used in the fitting process, allowing users to set the desired level of precision for the fit.
- `PrintFun`: specifies the function used to print progress messages during the fitting process. The default is `PrintTemporary`, which displays temporary output that can be useful for monitoring the fitting's progress.
- `SlackChannel`: names the Slack channel to which progress messages will be sent. If set to `None`, this feature is disabled, and no messages are sent to Slack.
- `ProgressView`: controls whether a progress window is displayed during the fitting process. When set to `True`, it provides an auxiliary notebook is created automatically with plots showing the progress of `NMinimize`.
- `SignatureCheck`: if `True`, the function ends prematurely and prints the list of the symbols that define the Hamiltonian after all basic simplifications have been applied without considering the given constraints.
- `SaveEigenvectors`: determines whether both the eigenvectors and eigenvalues of the fitted model are saved. If set to `False`, only the energies are saved.
- `AppendToLogFile`: what is provided here is appended to the log file under the "Appendix" key, enabling additional data to be stored alongside the fitting results.

The function returns an association with the following keys.

- `bestRMS`: the best root mean square deviation found during the fitting process.
- `bestParams`: the optimal set of parameters found through the fitting process.
- `paramSols`: a list of the parameter solutions at each step of the fitting algorithm.
- `timeTaken/s`: the total time taken to complete the fitting process, measured in seconds.

- `simplifier`: the replacement rules used to reduce the define the free-ion Hamiltonian.
- `excludeDataIndices`: the indices that were excluded from the fitting process as specified in the input.
- `startValues`: the initial values for the problem variables as given in the input.
- `freeIonSymbols`: symbols used in the intermediate coupling level calculation.
- `truncationEnergy`: the energy level at which the Hamiltonian was truncated.
- `numE`: the number of electrons in the f^{numE} configuration.
- `expData`: the experimental data used for the fitting process.
- `problemVars`: the variables considered during the fitting process.
- `maxIterations`: the maximum number of iterations used in the fitting process.
- `hamDim`: the dimension of the full Hamiltonian before simplifications or truncations.
- `allVars`: all the symbols defining the Hamiltonian under the applied simplifications.
- `freeBies`: the free-ion parameters used to calculate the intermediate coupling levels.
- `truncatedDim`: the dimension of the truncated Hamiltonian.
- `compiledIntermediateFname`: the file name of the compiled function used for the truncated Hamiltonian.
- `fittedLevels`: the number of levels that were fitted.
- `actualSteps`: the actual number of steps taken by the fitting algorithm.
- `solWithUncertainty`: a list of replacement rules showing the best fit value and its uncertainty for each parameter.
- `rmsHistory`: as list of the RMS values found during the fitting process.
- `Appendix`: an association appended to the log file under the "Appendix" key.
- `presentDataIndices`: the indices in `expData` that were used for fitting.
- `states`: a list of eigenvalues and eigenvectors for the fitted model, available if eigenvectors were saved.
- `energies`: a list of the energies of the fitted levels, adjusted if an energy shift was included in the fitting.

9 Accompanying notebooks

The code for this dissertation is accompanied by the following auxiliary *Mathematica* notebooks, which either document the functions included in the code, or serve as aids in the calculation of matrix elements.

- 🖫 `/notebooks/qlanth.nb`: gives an overview of functions included in **`qlanth`**.
- 🖫 `/notebooks/qlanth - Table Generator.nb`: generates the basic tables on which every calculation is based. This means that LS-reduced matrix elements are used to calculate matrix elements in the $|LSJM_J\rangle$ basis.
- 🖫 `/notebooks/qlanth - JJBlock Calculator.nb`: can be used to generate the J-J' blocks for the different interactions. The data files produced here are necessary for `HamMatrixAssembly` to work. These blocks are creating by putting together matrix elements from different interactions.
- 🖫 `/notebooks/The Lanthanides in LaF3.nb`: runs **`qlanth`** over the lanthanide ions in LaF_3 and compares the results against the published values from Carnall *et al.* [Car+89]. It also calculates magnetic dipole transition rates and oscillator strengths.

10 Compiled data for LaF_3:Ln^{3+} and $LiYF_4$:Ln^{3+}

The study of Carnall *et al.* [Car+89] on lanthanum fluoride was a systematic review of trivalent lanthanide ions in LaF_3. In this work they fitted the experimental data for all of the lanthanide ions using the single-configuration effective Hamiltonian. In their appendices one can find their calculated values, together with the experimental values that they used for their least squares fittings. In **qlanth** this data can be accessed by invoking the command `LoadCarnall` which brings into the session an association that has keys that have as values the tables and appendices from this article. Fig-6 shows the results of a calculation done with **qlanth** for the energy levels in LaF_3. Additionally the function `LoadLaF3Parameters` can be used to query the data for the fitted parameters, which may serve as a useful starting point for the description of the lanthanides ions in hosts other than LaF_3.

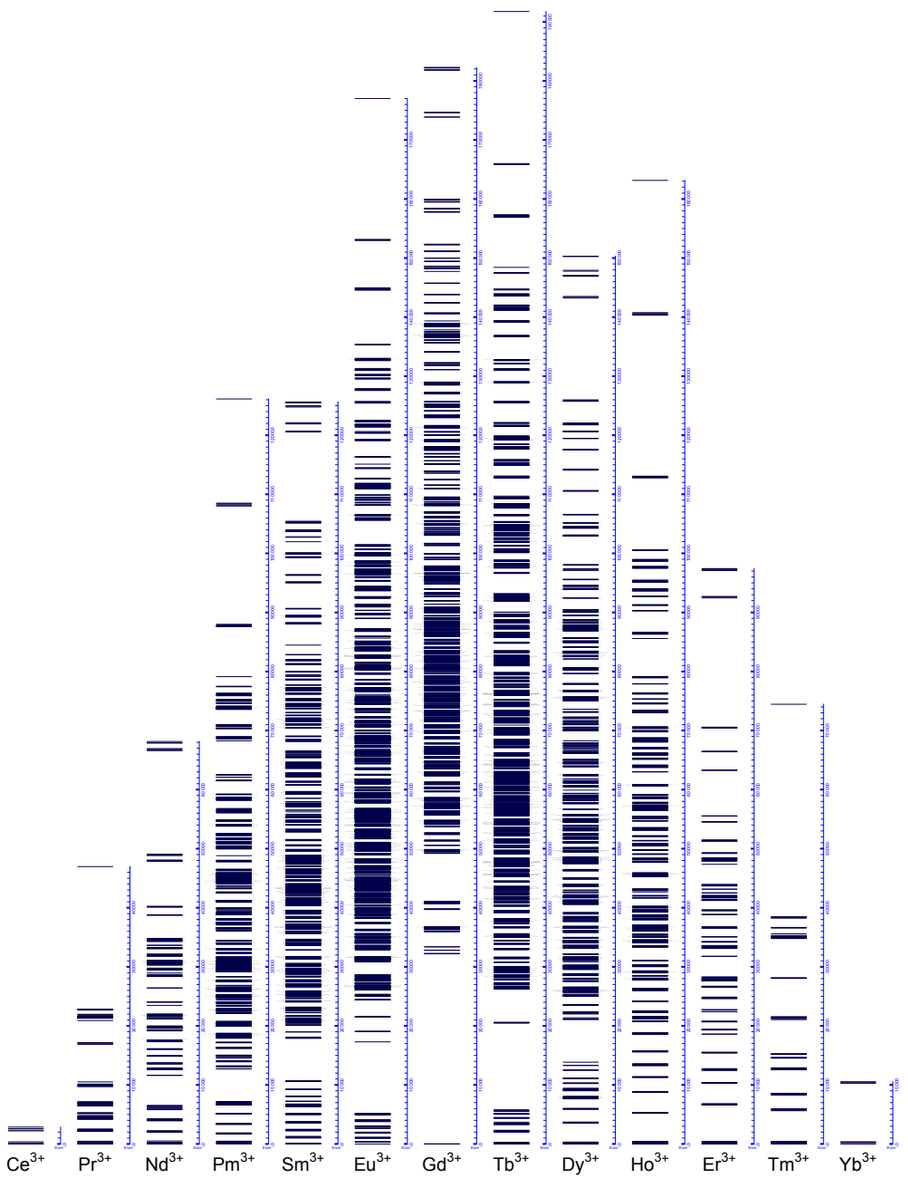


Figure 6: Energy levels in LaF_3.

Similarly, Cheng *et al.* [Che+16] compiled data for $LiYF_4$. In **qlanth** model parameters for $LiYF_4$ can be obtained by calling the function `LoadLiYF4Parameters`. Fig-7 shows the results of a calculation done with **qlanth** for the energy levels in $LiYF_4$.

```
Carnall::usage = "Association of data from Carnall et al (1989) with
    the following keys: {data, annotations, paramSymbols, elementNames
    , rawData, rawAnnotations, annnotatedData, appendix:Pr:Association
    , appendix:Pr:Calculated, appendix:Pr:RawTable, appendix:Headings}
    ";
```

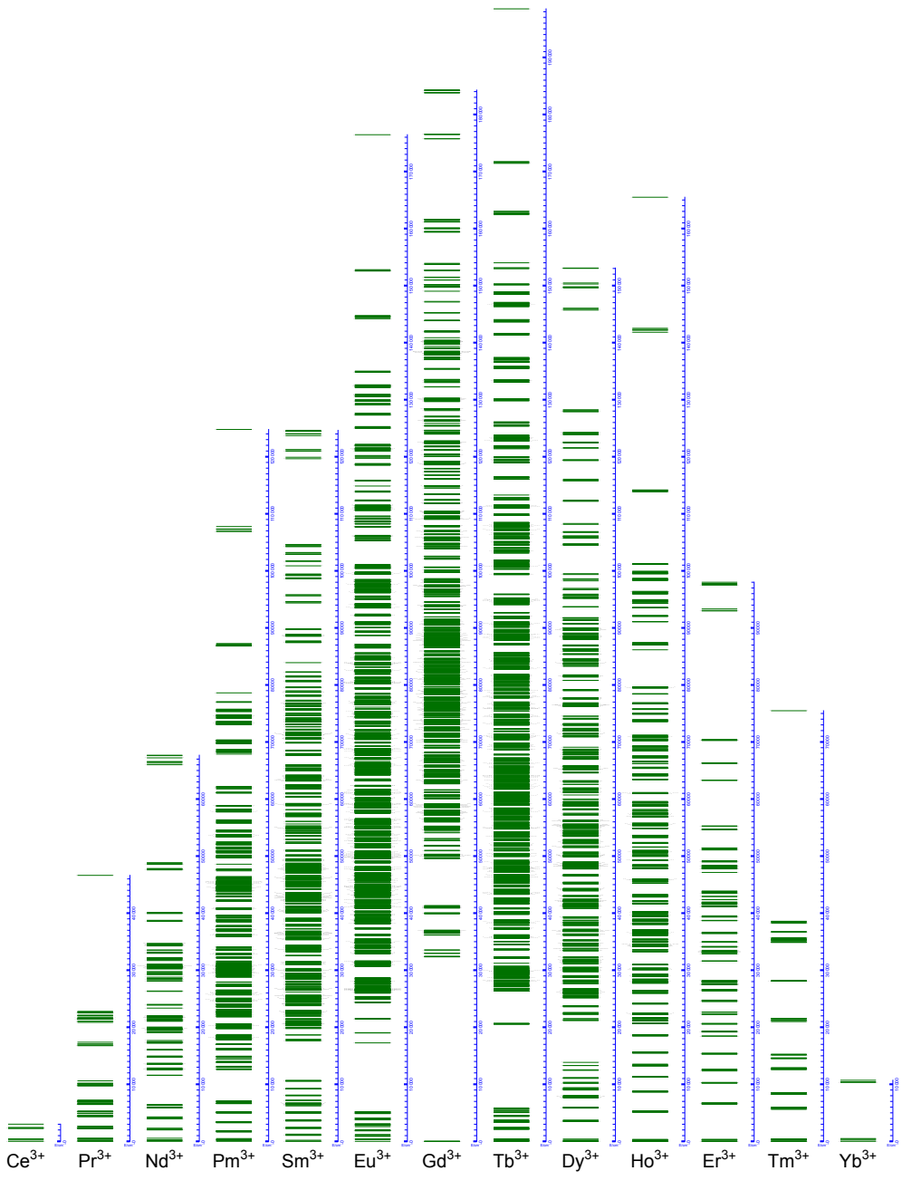


Figure 7: Energy levels in $LiYF_4$.

```
LoadCarnall::usage = "LoadCarnall[] loads data for trivalent
    lanthanides in LaF3 using the data from Bill Carnall's 1989 paper.
    ";
LoadCarnall[] := (
  If[ValueQ[Carnall], Return[]];
  carnallFname = FileNameJoin[{moduleDir, "data", "Carnall.m"}];
  If[!FileExistsQ[carnallFname],
    (PrintTemporary[">> Carnall.m not found, generating ..."];
      Carnall = ParseCarnall[];
    ),
    Carnall = Import[carnallFname];
  ];
);
```

```
LoadLaF3Parameters::usage = "LoadLaF3Parameters[ln] takes a string
    with the symbol the element of a trivalent lanthanide ion and
    returns model parameters for it. It is based on the data for LaF3.
     If the option ''Free Ion'' is set to True then the function sets
    all crystal field parameters to zero. Through the option ''gs'' it
     allows modyfing the electronic gyromagnetic ratio. For
    completeness this function also computes the E parameters using
    the F parameters quoted on Carnall.";
Options[LoadLaF3Parameters] = {
    "Free Ion"->False,
    "gs"->2.002319304386,
    "With Uncertainties"->False
    };
```

```
LoadLaF3Parameters[Ln_String, OptionsPattern[]] := Module[
  {params, uncertain,
  uncertainKeys, uncertainRules},
  (
    If[Not[ValueQ[Carnall]],
      LoadCarnall[];
    ];
    params = Association[Carnall["data"][Ln]];
    (*If a free ion then all the parameters from the crystal field
    are set to zero*)
    If[OptionValue["Free Ion"],
      Do[params[cfSymbol] = 0, {cfSymbol, cfSymbols}]
    ];
    params[F0] = 0;
    params[M2] = 0.56 * params[M0]; (*See Carnall 1989,Table I,
    caption,probably fixed based on HF values*)
    params[M4] = 0.31 * params[M0]; (*See Carnall 1989,Table I,
    caption,probably fixed based on HF values*)
    params[P0] = 0;
    params[P4] = 0.5 * params[P2];  (*See Carnall 1989,Table I,
    caption,probably fixed based on HF values*)
    params[P6] = 0.1 * params[P2];  (*See Carnall 1989,Table I,
    caption,probably fixed based on HF values*)
    params[gs] = OptionValue["gs"];
    {params[E0], params[E1], params[E2], params[E3]} = FtoE[params[F0
    ], params[F2], params[F4], params[F6]];
    params[E0] = 0;
    If[
      Not[OptionValue["With Uncertainties"]],
      Return[params],
      (
        uncertain     = Association[Carnall["annotations"][Ln]];
        uncertainKeys = Keys[uncertain];
        uncertain     = If[# == "Not allowed to vary in fitting." ||
    # == "Interpolated",
            0., #] & /@ uncertain;
        paramKeys = Keys[params];
        uncertainVals = Sort[Intersection[paramKeys, uncertainKeys]]
    /. Association[uncertain];
        uncertainRules = MapThread[Rule, {Sort[uncertainKeys],
    uncertainVals}];
        Which[
          MemberQ[{"Ce", "Yb"}, Ln],
          (
            subsetL = {F0};
            subsetR = {0};
          ),
          True,
          (
          subsetL = {F0, M2, M4, P0, P4, P6, E0, E1, E2, E3};
          subsetR = {0, M0*0.65, M0*0.31, 0, P2*0.5, P2*0.1,
            0,
            Sqrt[(196 F2^2)/164025 + (49 F4^2)/88209 + (122500 F6^2)
    /134165889],
            Sqrt[F2^2/4100625 + F4^2/10673289 + (30625 F6^2)
    /2743558264161],
            Sqrt[F2^2/18225 + (4 F4^2)/1185921 + (30625 F6^2)
    /1803785841]};
          )
        ];
        uncertainRules = Join[uncertainRules, MapThread[Rule, {
    subsetL,subsetR /. uncertainRules}]];
        uncertainRules = Association[uncertainRules];
        Which[
          Ln == "Eu",
          (
            uncertainRules[F4] = 12.121;
            uncertainRules[F6] = 15.872;
          ),
          Ln == "Gd",
          (
            uncertainRules[F4] = 12.07;
          ),
          Ln == "Tb",
          (
            uncertainRules[F4] = 41.006;
```

```
          )
        ];
        If[MemberQ[{"Eu","Gd","Tb"},Ln],
          (
            uncertainRules[E1] = Sqrt[(196 F2^2)/164025 + (49 F4^2)
    /88209 + (122500 F6^2)/134165889] /. uncertainRules;
            uncertainRules[E2] = Sqrt[F2^2/4100625 + F4^2/10673289 +
    (30625 F6^2)/2743558264161] /. uncertainRules;
            uncertainRules[E3] = Sqrt[F2^2/18225 + (4 F4^2)/1185921 +
     (30625 F6^2)/1803785841] /. uncertainRules;
          )
        ];
        uncertainKeys = First /@ Normal[uncertainRules];
        fullParams = Association[MapThread[Rule, {uncertainKeys,
    MapThread[Around, {uncertainKeys /. params, uncertainKeys /.
    uncertainRules}]}]];
        Return[Join[params, fullParams]]
      )
      ];
  )
];
```

```
LoadLiYF4Parameters::usage="LoadLiYF4Parameters[ln] takes a string
    with the symbol the element of a trivalent lanthanide ion and
    returns model parameters for it. It return the data for LiYF4 from
     Cheng et al.";
LoadLiYF4Parameters[ln_, OptionsPattern[]]:=(
  If[!ValueQ[paramsLiYF4],
    paramsChengLiYF4 = Import[FileNameJoin[{moduleDir,"data","
    chengLiYF4.m"}]];
  ];
  Return[paramsChengLiYF4[ln]];
)
```

11 sparsefn.py

qlanth is also accompanied by seven Python scripts `sparsef[1-7].py`. Each of these contains a function `effective_hamiltonian_f[1-7]` which returns a sparse array (using this data structure as provided by `scipy`) for given values for the model parameters.

There is an eight Python script called `basisLSJMJ.py` which contains a dictionary whose keys are f1, f2, f3, f4, f5, f6, and f7, and whose values are lists that contain the ordered basis in which the array produced by the `sparsefn.py` should be understood to be in. Each basis vector is a list with three elements {LS string in NK notation, J, M_J}.

In those it is left up to the user to make the adequate change of signs in the parameters for configurations above $\underline{f}^7$. These include changing the signs of all in &qn-18 and setting `t2Switch` to 0. For configurations at or below $\underline{f}^7$ it is necessary to set `t2Switch` to 1.

12 Data sources

The data (and their provenance) upon which **qlanth** bases its calculations is the following:

- Coefficients of fractional parentage and seniority numbers from Velkov [Vel00].
- Terms labels from $\underline{f}^1$ to $\underline{f}^7$ from Nielson and Koster [NK63].
- 3j-symbol [Wol24b] and 6j-symbol [Wol24a] values from *Mathematica* (v 13.2),
- Reduced matrix elements for the three body operators from Judd [JS84].
- Reduced matrix elements for the magnetic interactions from Judd [JCC68].

13 Other details

- Fitting the experimental data for the entire row might take about 45 minutes, if run for the first time, but takes much less time once compiled functions from the truncated (or not truncated) Hamiltonian have been saved to disk.
- The code was run in *Mathematica* version 13.2 on MacOS Sonoma 14.5.

14 Units

Following the tradition of the spectroscopic community, all the matrix elements of the Hamiltonian are calculated using the Kayser ($\mathcal{K} \equiv \text{cm}^{-1}$) as the (pseudo) energy unit. All the parameters (except the magnetic field which is in Tesla) in the effective Hamiltonian are assumed to be in this unit. As is customary, the angular momentum operators assume atomic units in which $\hbar = 1$.

Some constants and conversion values are included in the file `qonstants.m`.

```
BeginPackage["qonstants`"];

(* Physical Constants*)
bohrRadius = 5.29177210903 * 10^-9;
ee         = 1.602176634 * 10^-19;

(* Spectroscopic niceties*)
theLanthanides = {"Ce", "Pr", "Nd", "Pm", "Sm", "Eu", "Gd", "Tb", "Dy
    ", "Ho", "Er", "Tm", "Yb"};
theActinides   = {"Ac", "Th", "Pa", "U", "Np", "Pu", "Am", "Cm", "Bk"
    , "Cf", "Es", "Fm", "Md", "No", "Lr"};
theTrivalents  = {"Pr", "Nd", "Pm", "Sm", "Eu", "Gd", "Tb", "Dy", "Ho
    ", "Er", "Tm"};
specAlphabet   = "SPDFGHIKLMNOQRTUV";
complementaryNumE = {1,13,2,12,3,11,4,10,5,9,6,8,7};

(* SI *)
hPlanck = 6.62607015 * 10^-34;          (* Planck's constant in J s
    *)
hBar    = hPlanck    / (2 \[Pi]);       (* reduced Planck's constant
    in J s *)
\[Mu]B  = 9.2740100783 * 10^-24;        (* Bohr magneton in SI *)
me      = 9.1093837015 * 10^-31;        (* electron mass in kg *)
cLight  = 2.99792458   * 10^8;          (* speed of light in m/s *)
eCharge = 1.602176634  * 10^-19;        (* elementary charge in SI *)
\[Epsilon]0  = 8.8541878128 * 10^-12;   (* electric permittivity in
    vacuum in SI *)
\[Mu]0  = 4 \[Pi] * 10^-7;              (* magnetic permeability in
    vacuum in SI *)
αFine = 1/137.036;               (* fine structure constant *)

bohrRadius    = 5.29177*10^-11;         (* Bohr radius in m *)
hartreeEnergy = hBar^2 / (me * bohrRadius^2); (* Hartree energy in J
    *)
hartreeTime   = hBar / hartreeEnergy; (* Hartree time in s *)

(* Hartree atomic units *)
hPlanckHartree = 2 \[Pi]; (* Planck's constant in Hartree *)
meHartree      = 1;       (* electron mass in Hartree *)
cLightHartree  = 137.036; (* speed of light in Hartree *)
eChargeHartree = 1;       (* elementary charge in Hartree *)
\[Mu]0Hartree  = αFine^2; (* magnetic permeability in vacuum in
    Hartree *)

(* some conversion factors *)
eVToJoule       = eCharge;
jouleToeV       = 1 / eVToJoule;
jouleToHartree  = 1 / hartreeEnergy;
eVToKayser      = eCharge /( hPlanck * cLight * 100 ); (* 1 eV =
    8065.54429 cm^-1 *)
kayserToeV      = 1 / eVToKayser;
teslaToKayser   = 2 * \[Mu]B / hPlanck / cLight / 100;
kayserToHartree = kayserToeV * eVToJoule * jouleToHartree;
hartreeToKayser = 1 / kayserToHartree;

EndPackage[];
```

15 Notation

orbital angular momentum operator of a single electron

$$\overbrace{\hat{l}} \tag{121}$$

total orbital angular momentum operator

$$\overbrace{\hat{L}} \tag{122}$$

spin angular momentum operator of a single electron

$$\overbrace{\hat{s}} \tag{123}$$

total spin angular momentum operator

$$\overbrace{\hat{S}} \tag{124}$$

Shorthand for all other quantum numbers

$$\overbrace{\Lambda} \tag{125}$$

orbital angular momentum number

$$\overbrace{\underline{\ell}} \tag{126}$$

spinning angular momentum number

$$\overbrace{\underline{s}} \tag{127}$$

Coulomb non-central potential

$$\overbrace{\hat{\mathfrak{C}}} \tag{128}$$

LS-reduced matrix element of operator $\hat{O}$ between ΛLS and $\Lambda' L' S'$

$$\overbrace{\langle \Lambda LS \| \hat{O} \| \Lambda' L' S' \rangle} \tag{129}$$

LSJ-reduced matrix element of operator $\hat{O}$ between ΛLSJ and $\Lambda' L' S' J'$

$$\overbrace{\langle \Lambda LSJ \| \hat{O} \| \Lambda' L' S' J' \rangle} \tag{130}$$

Spectroscopic term αLS in Russel-Saunders notation

$$\overbrace{{}^{2S+1}\alpha \mathrm{L} \equiv |\alpha LS\rangle} \tag{131}$$

spherical tensor operator of rank k

$$\overbrace{\hat{X}^{(k)}} \tag{132}$$

q-component of the spherical tensor operator $\hat{X}^{(k)}$

$$\overbrace{\hat{X}^{(k)}_q} \tag{133}$$

The coefficient of fractional parentage from the parent term $|\underline{\ell}^{n-1} \alpha' L' S'\rangle$ for the daughter term $|\underline{\ell}^n \alpha LS\rangle$

$$\overbrace{\left(\underline{\ell}^{n-1} \alpha' L' S' \middle\} \underline{\ell}^n \alpha LS\right)} \tag{134}$$

16 Definitions

two plus one

$$\overbrace{[\![x]\!] \coloneqq 2x+1} \tag{135}$$

irreducible unit tensor operator of rank k

$$\overbrace{\hat{u}^{(k)}} \tag{136}$$

symmetric unit tensor operator for n equivalent electrons

$$\overbrace{\hat{U}^{(k)} \coloneqq \sum_{i=1}^{n} \hat{u}^{(k)}} \tag{137}$$

Renormalized spherical harmonics

$$\overbrace{C^{(k)}_q \coloneqq \sqrt{\frac{4\pi}{2k+1}} \mathrm{Y}^{(k)}_q} \tag{138}$$

Triangle "delta" between j_1, j_2, j_3

$$\overbrace{\triangle(j_1, j_2, j_3) \coloneqq \begin{cases} 1 & \text{if } j_1 = (j_2 + j_3), (j_2 + j_3 - 1), \ldots, |j_2 - j_3| \\ 0 & \text{otherwise} \end{cases}} \tag{139}$$

17 code

17.1 qlanth.m

This file encapsulates the main functions in **qlanth** and contains all the physics related functions.

```
(* ------------------------------------------------------------------------
+--------------------------------------------------------------------------+
|                                                                          |
|                                                                          |
|                      __                              __      __          |
|              ____ _   / /  ____ _   ____      / /_   / /_          |
|           / __ `/  / /  / __ `/  / __ \    / __/  / __ \         |
|          / /_/ /  / /  / /_/ /  / / / /  / /_    / / / /         |
|          \__, /  /_/   \__, _/  /_/ /_/   \__/  /_/ /_/          |
|            /_/                                                           |
|                                                                          |
|                                                                          |
+--------------------------------------------------------------------------+
This code was initially authored by Christopher M. Dodson and Rashid
Zia, and then rewritten and expanded by Juan David Lizarazo Ferro in
the years 2022-2024 under the advisory of Dr. Rashid Zia. It has
also benefited from the discussions with Tharnier Puel at the
University of Iowa.

It grew out of a collaboration sponsored by the NSF (NSF
DMR-1922025) between the groups of Dr. Rashid Zia at Brown
University, the Quantum Engineering Laboratory at the University of
Pennsylvania led by Dr. Lee Bassett, and the group of Dr. Michael
Flatté at the University of Iowa.

It uses an effective Hamiltonian to describe the electronic
structure of lanthanide ions in crystals. This effective Hamiltonian
includes terms representing the following interactions/relativistic
corrections: spin-orbit, electrostatic repulsion, spin-spin, crystal
field, and spin-other-orbit.

The Hilbert space used in this effective Hamiltonian is limited to
single f^n configurations. The inaccuracy of this single
configuration description is partially compensated by the inclusion
of configuration interaction terms as parametrized by the Casimir
operators of SO(3), G(2), and SO(7), and by three-body effective
operators ti.

The parameters included in this model are listed in the string
paramAtlas.

The notebook "qlanth.nb" contains a gallery with many of the
functions included in this module with some simple use cases.

The notebook "The Lanthanides in LaF3.nb" is an example in which the
results from this code are compared against the published results by
Carnall et. al for the energy levels of lanthinde ions in crystals
of lanthanum trifluoride.

VERSION: SEPTEMBER 2024

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```

```
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------------------------------------------------------------------- *)

BeginPackage["qlanth`"];
Needs["qonstants`"];
Needs["qplotter`"];
Needs["misc`"];

paramAtlas = "
E0: linear combination of F_k, see eqn. (2-80) in Wybourne 1965
E1: linear combination of F_k, see eqn. (2-80) in Wybourne 1965
E2: linear combination of F_k, see eqn. (2-80) in Wybourne 1965
E3: linear combination of F_k, see eqn. (2-80) in Wybourne 1965
```

```
146
147 ζ: spin-orbit strength parameter.
148
149 F0: Direct Slater integral F^0, produces an overall shift of all
    energy levels.
150 F2: Direct Slater integral F^2
151 F4: Direct Slater integral F^4, possibly constrained by ratio to F^2
152 F6: Direct Slater integral F^6, possibly constrained by ratio to F^2
153
154 M0: 0th Marvin integral
155 M2: 2nd Marvin integral
156 M4: 4th Marvin integral
157 \[Sigma]SS: spin-spin override, if 0 spin-spin is omitted, if 1 then
    spin-spin is included
158
159 T2:  three-body effective operator parameter T^2  (non-orthogonal)
160 T2p: three-body effective operator parameter T^2' (orthogonalized T2)
161 T3:  three-body effective operator parameter T^3
162 T4:  three-body effective operator parameter T^4
163 T6:  three-body effective operator parameter T^6
164 T7:  three-body effective operator parameter T^7
165 T8:  three-body effective operator parameter T^8
166
167 T11p: three-body effective operator parameter T^11' (orthogonalized
    T11)
168 T12:  three-body effective operator parameter T^12
169 T14:  three-body effective operator parameter T^14
170 T15:  three-body effective operator parameter T^15
171 T16:  three-body effective operator parameter T^16
172 T17:  three-body effective operator parameter T^17
173 T18:  three-body effective operator parameter T^18
174 T19:  three-body effective operator parameter T^19
175
176 P0: pseudo-magnetic parameter P^0
177 P2: pseudo-magnetic parameter P^2
178 P4: pseudo-magnetic parameter P^4
179 P6: pseudo-magnetic parameter P^6
180
181 gs: electronic gyromagnetic ratio
182
183 α: Trees' parameter α describing configuration interaction via the
    Casimir operator of SO(3)
184 β: Trees' parameter β describing configuration interaction via the
    Casimir operator of G(2)
185 γ: Trees' parameter γ describing configuration interaction via the
    Casimir operator of SO(7)
186
187 B02: crystal field parameter B_0^2 (real)
188 B04: crystal field parameter B_0^4 (real)
189 B06: crystal field parameter B_0^6 (real)
190 B12: crystal field parameter B_1^2 (real)
191 B14: crystal field parameter B_1^4 (real)
192
193 B16: crystal field parameter B_1^6 (real)
194 B22: crystal field parameter B_2^2 (real)
195 B24: crystal field parameter B_2^4 (real)
196 B26: crystal field parameter B_2^6 (real)
197 B34: crystal field parameter B_3^4 (real)
198
199 B36: crystal field parameter B_3^6 (real)
200 B44: crystal field parameter B_4^4 (real)
201 B46: crystal field parameter B_4^6 (real)
202 B56: crystal field parameter B_5^6 (real)
203 B66: crystal field parameter B_6^6 (real)
204
205 S12: crystal field parameter S_1^2 (real)
206 S14: crystal field parameter S_1^4 (real)
207 S16: crystal field parameter S_1^6 (real)
208 S22: crystal field parameter S_2^2 (real)
209
210 S24: crystal field parameter S_2^4 (real)
211 S26: crystal field parameter S_2^6 (real)
212 S34: crystal field parameter S_3^4 (real)
213 S36: crystal field parameter S_3^6 (real)
214
215 S44: crystal field parameter S_4^4 (real)
```

```
S46: crystal field parameter S_4^6 (real)
S56: crystal field parameter S_5^6 (real)
S66: crystal field parameter S_6^6 (real)

\[Epsilon]: ground level baseline shift
t2Switch: controls the usage of the t2 operator beyond f7 (1 for f7
    or below, 0 for f8 or above)
wChErrA: If 1 then the type-A errors in Chen are used, if 0 then not.
wChErrB: If 1 then the type-B errors in Chen are used, if 0 then not.

Bx: x component of external magnetic field (in T)
By: y component of external magnetic field (in T)
Bz: z component of external magnetic field (in T)

\[CapitalOmega]2: Judd-Ofelt intensity parameter k=2 (in cm^2)
\[CapitalOmega]4: Judd-Ofelt intensity parameter k=4 (in cm^2)
\[CapitalOmega]6: Judd-Ofelt intensity parameter k=6 (in cm^2)

nE: number of electrons in a configuration

E0p: orthogonalized E0
E1p: orthogonalized E1
E2p: orthogonalized E2
E3p: orthogonalized E3
αp: orthogonalized α
βp: orthogonalized β
γp: orthogonalized γ
";
paramSymbols   = StringSplit[paramAtlas, "\n"];
paramSymbols   = Select[paramSymbols, # != ""& ];
paramSymbols   = ToExpression[StringSplit[#, ":"][[1]]] & /@
    paramSymbols;
Protect /@ paramSymbols;

(* Parameter families *)
Unprotect[racahSymbols, chenSymbols, slaterSymbols, controlSymbols,
    cfSymbols, TSymbols, pseudoMagneticSymbols, marvinSymbols,
    casimirSymbols, magneticSymbols, juddOfeltIntensitySymbols,
    mostlyOrthogonalSymbols];
racahSymbols   = {E0, E1, E2, E3};
chenSymbols    = {wChErrA, wChErrB};
slaterSymbols  = {F0, F2, F4, F6};
controlSymbols = {t2Switch, \[Sigma]SS};
cfSymbols      = {B02, B04, B06, B12, B14, B16, B22, B24, B26, B34,
    B36,
                  B44, B46, B56, B66,
                  S12, S14, S16, S22, S24, S26, S34, S36, S44, S46,
    S56, S66};
TSymbols       = {T2, T2p, T3, T4, T6, T7, T8, T11p, T12, T14, T15,
    T16, T17, T18, T19};
pseudoMagneticSymbols = {P0, P2, P4, P6};
marvinSymbols         = {M0, M2, M4};
magneticSymbols       = {Bx, By, Bz, gs, ζ};
casimirSymbols        = {α, β, γ};
juddOfeltIntensitySymbols = {\[CapitalOmega]2, \[CapitalOmega]4, \[
    CapitalOmega]6};
mostlyOrthogonalSymbols = Join[{E0p, E1p, E2p, E3p, αp, βp, γp},
  {T2p, T3, T4, T6, T7, T8, T11p, T12, T14, T15, T16, T17, T18, T19},
  cfSymbols];

paramFamilies         = Hold[{racahSymbols, chenSymbols,
    slaterSymbols, controlSymbols, cfSymbols, TSymbols,
    pseudoMagneticSymbols, marvinSymbols, casimirSymbols,
    magneticSymbols, juddOfeltIntensitySymbols,
    mostlyOrthogonalSymbols}];
ReleaseHold[Protect /@ paramFamilies];
crystalGroups = {"C1","Ci","C2","Cs","C2h","D2","C2v","D2h","C4","S4"
    ,"C4h","D4","C4v","D3d","D4h","C3","C3i","D3","C3v","D3d","C6","
    C3h","C6h","D6","C6v","D3h","D6h","T","Th","O","Td","Oh"};

(* Parameter usage *)
paramLines = Select[StringSplit[paramAtlas, "\n"], # != "" &];
usageTemplate = StringTemplate["`paramSymbol`::usage=\"`paramSymbol`
    : `paramUsage`\";"];
Do[(
  {paramString, paramUsage} = StringSplit[paramLine, ":"];
```

```
  paramUsage                = StringTrim[paramUsage];
  expressionString          = usageTemplate[<|"paramSymbol" ->
    paramString, "paramUsage" -> paramUsage|>];
  ToExpression[usageTemplate[<|"paramSymbol" -> paramString, "
    paramUsage" -> paramUsage|>]]
),
{paramLine, paramLines}
];

AllowedJ;
AllowedMforJ;
AllowedNKSLJMforJMTerms;
AllowedNKSLJMforJTerms;
AllowedNKSLJTerms;

AllowedNKSLTerms;
AllowedNKSLforJTerms;
AllowedSLJMTerms;
AllowedSLJTerms;
AllowedSLTerms;

AngularMomentumMatrices;
BasisLSJ;
BasisLSJMJ;
BasisTableGenerator;
Bqk;
CFP;

CFPAssoc;
CFPTable;
CFPTerms;
Carnall;
CasimirG2;

CasimirSO3;
CasimirSO7;
Cqk;
CrystalField;
CrystalFieldForm;

Dk;
EigenLever;
Electrostatic;
ElectrostaticConfigInteraction;
ElectrostaticTable;

EnergyLevelDiagram;
EnergyStates;
EtoF;
ExportMZip;
ExportmZip;

FindNKLSTerm;
FindSL;
FreeHam;
FreeIonTable;
FromArrayToTable;
FtoE;

GG2U;
GSO7W;
GenerateCFP;
GenerateCFPAssoc;
GenerateCFPTable;

GenerateCrystalFieldTable;
GenerateElectrostaticTable;
GenerateFreeIonTable;
GenerateReducedUkTable;
GenerateReducedV1kTable;
GenerateSOOandECSOLSTable;

GenerateSOOandECSOTable;
GenerateSpinOrbitTable;
GenerateSpinSpinTable;
GenerateT22Table;
```

```
GenerateThreeBodyTables;

Generator;
GroundMagDipoleOscillatorStrength;
HamMatrixAssembly;
HamiltonianForm;

HamiltonianMatrixPlot;
HoleElectronConjugation;
ImportMZip;
IonSolver;
JJBlockMagDip;

JJBlockMatrix;
JJBlockMatrixFileName;
JJBlockMatrixTable;
JuddOfeltUkSquared;
LabeledGrid;
LandeFactor;

LevelElecDipoleOscillatorStrength;
LevelJJBlockMagDipole;
LevelMagDipoleLineStrength;
LevelMagDipoleMatrixAssembly;
LevelMagDipoleOscillatorStrength;

LevelMagDipoleSpontaneousDecayRates;
LevelSimplerSymbolicHamMatrix;
LevelSolver;
ListRepeater;
LoadAll;

LoadCFP;
LoadCarnall;
LoadChenDeltas;
LoadElectrostatic;
LoadFreeIon;
LoadGuillotParameters;

LoadLaF3Parameters;
LoadLiYF4Parameters;
LoadSOOandECSO;
LoadSOOandECSOLS;
LoadSpinOrbit;
LoadSpinSpin;

LoadSymbolicHamiltonians;
LoadT11;
LoadT22;
LoadTermLabels;
LoadThreeBody;

LoadUk;
LoadV1k;
MagDipLineStrength;
MagDipoleMatrixAssembly;
MagDipoleRates;

MagneticInteractions;
MapToSparseArray;
MaxJ;
MinJ;
NKCFPPhase;

ParamPad;
ParseBenelli2015;
ParseStates;
ParseStatesByNumBasisVecs;
ParseStatesByProbabilitySum;

ParseTermLabels;
Phaser;
PrettySaundersSL;
PrettySaundersSLJ;
PrettySaundersSLJmJ;

```

```
PrintL;
PrintSLJ;
PrintSLJM;
ReducedSOOandECSOinf2;
ReducedSOOandECSOinfn;

ReducedT11inf2;
ReducedT22inf2;
ReducedT22infn;
ReducedUk;
ReducedUkTable;

ReducedV1kTable;
Reducedt11inf2;
ReplaceInSparseArray;
ScalarLSJMFromLS;
SOOandECSO;
SOOandECSOLSTable;

SOOandECSOTable;
ScalarOperatorProduct;
Seniority;
ShiftedLevels;
SimplerSymbolicHamMatrix;

SixJay;
SpinOrbit;
SpinOrbitTable;
SpinSpin;
SpinSpinTable;

Sqk;
SquarePrimeToNormal;
TPO;
TabulateJJBlockMagDipTable;
TabulateJJBlockMatrixTable;

TabulateManyJJBlockMagDipTables;
TabulateManyJJBlockMatrixTables;
ThreeBodyTable;
ThreeBodyTables;
ThreeJay;

TotalCFIters;
chenDeltas;
fK;
fnTermLabels;
fsubk;

fsupk;
moduleDir;
symbolicHamiltonians;

(* this selects the function that is applied to calculated matrix
   elements which helps keep down the complexity of the resulting
   algebraic expressions *)
SimplifyFun = Expand;

Begin["`Private`"]

  moduleDir = DirectoryName[$InputFileName];
  frontEndAvailable = (Head[$FrontEnd] === FrontEndObject);

  (* ############################################################ *)
  (* ########################### MISC ########################### *)

  TPO::usage = "TPO[x, y, ...] gives the product of 2x+1, 2y+1, ...";
  TPO[args__] := Times @@ ((2*# + 1) & /@ {args});

  Phaser::usage = "Phaser[x] gives (-1)^x.";
  Phaser[exponent_] := ((-1)^exponent);

  TriangleCondition::usage = "TriangleCondition[a, b, c] evaluates
    the triangle condition on a, b, and c.";
  TriangleCondition[a_, b_, c_] := (Abs[b - c] <= a <= (b + c));

```

```
TriangleAndSumCondition::usage = "TriangleAndSumCondition[a, b, c]
  evaluates the joint satisfaction of the triangle and sum
  conditions.";
TriangleAndSumCondition[a_, b_, c_] := (
  And[
    Abs[b - c] <= a <= (b + c),
    IntegerQ[a + b + c]
    ]
  );

SquarePrimeToNormal::usage = "SquarePrimeToNormal[squarePrime]
  evaluates the standard representation of a number from the squared
   prime representation given in the list squarePrime. For
  squarePrime of the form {c0, c1, c2, c3, ...} this function
  returns the number c0 * Sqrt[p1^c1 * p2^c2 * p3^c3 * ...] where pi
   is the ith prime number. Exceptionally some of the ci might be
  letters in which case they have to be one of \"A\", \"B\", \"C\",
  \"D\" with them corresponding to 10, 11, 12, and 13, respectively.
  ";
SquarePrimeToNormal[squarePrime_] :=
(
  radical = Product[Prime[idx1 - 1] ^ Part[squarePrime, idx1], {
  idx1, 2, Length[squarePrime]}];
  radical = radical /. {"A" -> 10, "B" -> 11, "C" -> 12, "D" ->
  13};
  val = squarePrime[[1]] * Sqrt[radical];
  Return[val];
);

ParamPad::usage = "ParamPad[params] takes an association params
  whose keys are a subset of paramSymbols. The function returns a
  new association where all the keys not present in paramSymbols,
  will now be included in the returned association with their values
   set to zero.
The function additionally takes an option \"Print\" that if set to
  True, will print the symbols that were not present in the given
  association. The default is True.";
Options[ParamPad] = {"PrintFun" -> PrintTemporary};
ParamPad[params_, OptionsPattern[]] := (
  notPresentSymbols = Complement[paramSymbols, Keys[params]];
  PrintFun = OptionValue["PrintFun"];
  PrintFun["Following symbols were not given and are being set to
  0: ",
    notPresentSymbols];
  newParams = Transpose[{paramSymbols, ConstantArray[0, Length[
  paramSymbols]]}];
  newParams = (#[[1]] -> #[[2]]) & /@ newParams;
  newParams = Association[newParams];
  newParams = Join[newParams, params];
  Return[newParams];
  )

(* ############################################################ *)
(* ##################### Angular Momentum ##################### *)

AngularMomentumMatrices::usage = "AngularMomentumMatrices[j] gives
  the matrix representation for the angular momentum operators Jx,
  Jy, and Jz for a given angular momentum j in the basis of
  eigenvectors of jz. j may be a half-integer or an integer.
The options are \"Sparse\" which defaults to False and \"Order\"
  which defaults to \"HighToLow\".
The option \"Order\" can be set to \"LowToHigh\" to get the
  matrices in the order from -jay to jay otherwise they are returned
   in the order jay to -jay.
The function returns a list {JxMatrix, JyMatrix, JzMatrix} with the
   matrix representations for the cartesian components of the
  angular momentum operator.";
Options[AngularMomentumMatrices] = {
  "Sparse" -> False,
  "Order" -> "HighToLow"};
AngularMomentumMatrices[jay_, OptionsPattern[]] := Module[
  {
    JxMatrix, JyMatrix, JzMatrix,
    JPlusMatrix, JMinusMatrix,
    m1, m2,
    ArrayInverter
```

```
    },
    (
      ArrayInverter = #[[-1 ;; 1 ;; -1, -1 ;; 1 ;; -1]] &;
      JPlusMatrix   = Table[
        If[m2 == m1 + 1,
          Sqrt[(jay - m1) (jay + m1 + 1)],
          0
          ],
        {m1, jay, -jay, -1},
        {m2, jay, -jay, -1}];
      JMinusMatrix = Table[
        If[m2 == m1 - 1,
          Sqrt[(jay + m1) (jay - m1 + 1)],
          0
          ],
        {m1, jay, -jay, -1},
        {m2, jay, -jay, -1}];
      JxMatrix = (JPlusMatrix + JMinusMatrix)/2;
      JyMatrix = (JMinusMatrix - JPlusMatrix)/(2 I);
      JzMatrix = DiagonalMatrix[Table[m, {m, jay, -jay, -1}]];
      If[OptionValue["Sparse"],
        {JxMatrix, JyMatrix, JzMatrix} = SparseArray /@ {JxMatrix,
    JyMatrix, JzMatrix}
      ];
      If[OptionValue["Order"] == "LowToHigh",
        {JxMatrix, JyMatrix, JzMatrix} = ArrayInverter /@ {JxMatrix,
    JyMatrix, JzMatrix};
      ];
      Return[{JxMatrix, JyMatrix, JzMatrix}];
    )
  ];

  LandeFactor::usage="LandeFactor[J, L, S] gives the Lande factor for
     a given total angular momentum J, orbital angular momentum L, and
     spin S.";
  LandeFactor[J_, L_, S_] := (3/2) + (S*(S + 1) - L*(L + 1))/(2*J*(J
    + 1));

  (* ##################### Angular Momentum ###################### *)
  (* ############################################################## *)


  (* ############################################################## *)
  (* ######################## Racah Algebra ######################### *)

  ReducedUk::usage = "ReducedUk[n, l, SL, SpLp, k] gives the reduced
    matrix element of the symmetric unit tensor operator U^(k). See
    equation 11.53 in TASS.";
  ReducedUk[numE_, l_, SL_, SpLp_, k_] := Module[
    {spin, orbital, Uk, S, L,
    Sp, Lp, Sb, Lb, parentSL,
    cfpSL, cfpSpLp, Ukval,
    SLparents, SLpparents,
    commonParents, phase},
      {spin, orbital} = {1/2, 3};
      {S, L}          = FindSL[SL];
      {Sp, Lp}        = FindSL[SpLp];
      If[Not[S == Sp],
        Return[0]
      ];
      cfpSL      = CFP[{numE, SL}];
      cfpSpLp    = CFP[{numE, SpLp}];
      SLparents  = First /@ Rest[cfpSL];
      SLpparents = First /@ Rest[cfpSpLp];
      commonParents = Intersection[SLparents, SLpparents];
      Uk = Sum[(
        {Sb, Lb} = FindSL[\[Psi]b];
        Phaser[Lb] *
          CFPAssoc[{numE, SL, \[Psi]b}] *
          CFPAssoc[{numE, SpLp, \[Psi]b}] *
          SixJay[{orbital, k, orbital}, {L, Lb, Lp}]
      ),
      {\[Psi]b, commonParents}
      ];
      phase     = Phaser[orbital + L + k];
      prefactor = numE * phase * Sqrt[TPO[L,Lp]];
```

```
    Ukval     = prefactor*Uk;
    Return[Ukval];
]

Ck::usage = "Ck[orbital, k] gives the diagonal reduced matrix
  element <l||C^(k)||l> where the Subscript[C, q]^(k) are
  renormalized spherical harmonics. See equation 11.23 in TASS with
  l=l'.";
Ck[orbital_, k_] := (-1)^orbital * TPO[orbital] * ThreeJay[{orbital
  , 0}, {k, 0}, {orbital, 0}];

SixJay::usage = "SixJay[{j1, j2, j3}, {j4, j5, j6}] provides the
  value for SixJSymbol[{j1, j2, j3}, {j4, j5, j6}] with memorization
   of computed values and short-circuiting values based on triangle
  conditions.";
SixJay[{j1_, j2_, j3_}, {j4_, j5_, j6_}] := (
  sixJayval = Which[
    Not[TriangleAndSumCondition[j1, j2, j3]],
    0,
    Not[TriangleAndSumCondition[j1, j5, j6]],
    0,
    Not[TriangleAndSumCondition[j4, j2, j6]],
    0,
    Not[TriangleAndSumCondition[j4, j5, j3]],
    0,
    True,
    SixJSymbol[{j1, j2, j3}, {j4, j5, j6}]];
  SixJay[{j1, j2, j3}, {j4, j5, j6}] = sixJayval);

ThreeJay::usage = "ThreeJay[{j1, m1}, {j2, m2}, {j3, m3}] gives the
   value of the Wigner 3j-symbol and memorizes the computed value.";
ThreeJay[{j1_, m1_}, {j2_, m2_}, {j3_, m3_}] := (
 threejval = Which[
   Not[(m1 + m2 + m3) == 0],
   0,
   Not[TriangleCondition[j1,j2,j3]],
   0,
   True,
   ThreeJSymbol[{j1, m1}, {j2, m2}, {j3, m3}]
   ];
 ThreeJay[{j1, m1}, {j2, m2}, {j3, m3}] = threejval);

ReducedV1k::usage = "ReducedV1k[n, l, SL, SpLp, k] gives the
  reduced matrix element of the spherical tensor operator V^(1k).
  See equation 2-101 in Wybourne 1965.";
ReducedV1k[numE_, SL_, SpLp_, k_] := Module[
  {Vk1, S, L, Sp, Lp,
  Sb, Lb, spin, orbital,
  cfpSL, cfpSpLp,
  SLparents, SpLpparents,
  commonParents, prefactor},
  (
    {spin, orbital} = {1/2, 3};
    {S, L}          = FindSL[SL];
    {Sp, Lp}        = FindSL[SpLp];
    cfpSL           = CFP[{numE, SL}];
    cfpSpLp         = CFP[{numE, SpLp}];
    SLparents       = First /@ Rest[cfpSL];
    SpLpparents     = First /@ Rest[cfpSpLp];
    commonParents = Intersection[SLparents, SpLpparents];
    Vk1 = Sum[(
        {Sb, Lb} = FindSL[\[Psi]b];
        Phaser[(Sb + Lb + S + L + orbital + k - spin)] *
        CFPAssoc[{numE, SL, \[Psi]b}] *
        CFPAssoc[{numE, SpLp, \[Psi]b}] *
        SixJay[{S, Sp, 1}, {spin, spin, Sb}] *
        SixJay[{L, Lp, k}, {orbital, orbital, Lb}]
      ),
    {\[Psi]b, commonParents}
    ];
    prefactor = numE * Sqrt[spin * (spin + 1) * TPO[spin, S, L, Sp,
   Lp] ];
    Return[prefactor * Vk1];
  )
];
```

```
GenerateReducedUkTable::usage = "GenerateReducedUkTable[numEmax] can be used to generate the association of reduced matrix elements for the unit tensor operators Uk from f^1 up to f^numEmax. If the option \"Export\" is set to True then the resulting data is saved to ./data/ReducedUkTable.m.";
Options[GenerateReducedUkTable] = {"Export" -> True, "Progress" -> True};
GenerateReducedUkTable[numEmax_Integer:7, OptionsPattern[]] := (
  numValues = Total[Length[AllowedNKSLTerms[#]]*Length[AllowedNKSLTerms[#]]&/@Range[1, numEmax]] * 4;
  Print["Calculating " <> ToString[numValues] <> " values for Uk k=0,2,4,6."];
  counter = 1;
  If[And[OptionValue["Progress"], frontEndAvailable],
  progBar = PrintTemporary[
      Dynamic[Row[{ProgressIndicator[counter, {0, numValues}], " ", counter}]]]
    ];
  ReducedUkTable = Table[
    (
      counter = counter+1;
      {numE, 3, SL, SpLp, k} -> SimplifyFun[ReducedUk[numE, 3, SL, SpLp, k]]
    ),
    {numE, 1, numEmax},
    {SL,   AllowedNKSLTerms[numE]},
    {SpLp, AllowedNKSLTerms[numE]},
    {k, {0, 2, 4, 6}}
  ];
  ReducedUkTable = Association[Flatten[ReducedUkTable]];
  ReducedUkTableFname = FileNameJoin[{moduleDir, "data", "ReducedUkTable.m"}];
  If[And[OptionValue["Progress"], frontEndAvailable],
    NotebookDelete[progBar]
  ];
  If[OptionValue["Export"],
    (
      Print["Exporting to file " <> ToString[ReducedUkTableFname]];
      Export[ReducedUkTableFname, ReducedUkTable];
    )
  ];
  Return[ReducedUkTable];
)

GenerateReducedV1kTable::usage = "GenerateReducedV1kTable[nmax] calculates values for Vk1 and returns an association where the keys are lists of the form {n, SL, SpLp, 1}. If the option \"Export\" is set to True then the resulting data is saved to ./data/ReducedV1kTable.m.";
Options[GenerateReducedV1kTable] = {"Export" -> True, "Progress" -> True};
GenerateReducedV1kTable[numEmax_Integer:7, OptionsPattern[]] := (
  numValues = Total[Length[AllowedNKSLTerms[#]]*Length[AllowedNKSLTerms[#]]&/@Range[1, numEmax]];
  Print["Calculating " <> ToString[numValues] <> " values for Vk1."];
  counter = 1;
  If[And[OptionValue["Progress"], frontEndAvailable],
  progBar = PrintTemporary[
      Dynamic[Row[{ProgressIndicator[counter, {0, numValues}], " ", counter}]]]
    ];
  ReducedV1kTable = Table[
    (
      counter = counter+1;
      {n, SL, SpLp, 1} -> SimplifyFun[ReducedV1k[n, SL, SpLp, 1]]
    ),
    {n, 1, numEmax},
    {SL, AllowedNKSLTerms[n]},
    {SpLp, AllowedNKSLTerms[n]}
  ];
  ReducedV1kTable = Association[ReducedV1kTable];
  If[And[OptionValue["Progress"], frontEndAvailable],
    NotebookDelete[progBar]
  ];
  exportFname = FileNameJoin[{moduleDir, "data", "ReducedV1kTable.m
```

```
    "}];
741   If[OptionValue["Export"],
742     (
743       Print["Exporting to file "<>ToString[exportFname]];
744       Export[exportFname, ReducedV1kTable];
745     )
746   ];
747   Return[ReducedV1kTable];
748 )
749
750 (* ###################### Racah Algebra ###################### *)
751 (* ########################################################## *)
752
753 (* ########################################################## *)
754 (* ###################### Electrostatic ###################### *)
755
756 fsubk::usage = "fsubk[numE, orbital, SL, SLp, k] gives the Slater
      integral f_k for the given configuration and pair of SL terms. See
       equation 12.17 in TASS.";
757 fsubk[numE_, orbital_, NKSL_, NKSLp_, k_] := Module[
758   {terms, S, L, Sp, Lp,
759   termsWithSameSpin, SL,
760   fsubkVal, spinMultiplicity,
761   prefactor, summand1, summand2},
762   (
763     {S, L}   = FindSL[NKSL];
764     {Sp, Lp} = FindSL[NKSLp];
765     terms    = AllowedNKSLTerms[numE];
766     (* sum for summand1 is over terms with same spin *)
767     spinMultiplicity  = 2*S + 1;
768     termsWithSameSpin = StringCases[terms, ToString[
      spinMultiplicity] ~~ __];
769     termsWithSameSpin = Flatten[termsWithSameSpin];
770     If[Not[{S, L} == {Sp, Lp}],
771       Return[0]
772     ];
773     prefactor = 1/2 * Abs[Ck[orbital, k]]^2;
774     summand1 = Sum[(
775         ReducedUkTable[{numE, orbital, SL, NKSL,  k}] *
776         ReducedUkTable[{numE, orbital, SL, NKSLp, k}]
777         ),
778       {SL, termsWithSameSpin}
779     ];
780     summand1 = 1 / TPO[L] * summand1;
781     summand2 = (
782       KroneckerDelta[NKSL, NKSLp] *
783         (numE *(4*orbital + 2 - numE)) /
784         ((2*orbital + 1) * (4*orbital + 1))
785       );
786     fsubkVal = prefactor*(summand1 - summand2);
787     Return[fsubkVal];
788   )
789 ];
790
791 fsupk::usage = "fsupk[numE, orbital, SL, SLp, k] gives the
      superscripted Slater integral f^k = Subscript[f, k] * Subscript[D,
       k].";
792 fsupk[numE_, orbital_, NKSL_, NKSLp_ ,k_] := (
793   Dk[k] * fsubk[numE, orbital, NKSL, NKSLp, k]
794   )
795
796 Dk::usage = "D[k] gives the ratio between the super-script and sub-
      scripted Slater integrals (F^k / F_k). k must be even. See table
      6-3 in TASS, and also section 2-7 of Wybourne (1965). See also
      equation 6.41 in TASS.";
797 Dk[k_] := {1, 225, 1089, 184041/25}[[k/2+1]];
798
799 FtoE::usage = "FtoE[F0, F2, F4, F6] calculates the Racah parameters
      {E0, E1, E2, E3} corresponding to the given Slater integrals.
800 See eqn. 2-80 in Wybourne.
801 Note that in that equation the subscripted Slater integrals are
      used but since this function assumes the the input values are
      superscripted Slater integrals, it is necessary to convert them
      using Dk.";
802 FtoE[F0_, F2_, F4_, F6_] := Module[
803   {E0, E1, E2, E3},
```

```
    (
      E0 = (F0 - 10*F2/Dk[2] - 33*F4/Dk[4] - 286*F6/Dk[6]);
      E1 = (70*F2/Dk[2] + 231*F4/Dk[4] + 2002*F6/Dk[6])/9;
      E2 = (F2/Dk[2] - 3*F4/Dk[4] + 7*F6/Dk[6])/9;
      E3 = (5*F2/Dk[2] + 6*F4/Dk[4] - 91*F6/Dk[6])/3;
      Return[{E0, E1, E2, E3}];
    )
  ];

  EtoF::usage = "EtoF[E0, E1, E2, E3] calculates the Slater integral
    parameters {F0, F2, F4, F6} corresponding to the given Racah
    parameters {E0, E1, E2, E3}. This is the inverse of the FtoE
    function.";
  EtoF[E0_, E1_, E2_, E3_] := Module[
    {F0, F2, F4, F6},
    (
      F0 = 1/7      (7 E0 + 9 E1);
      F2 = 75/14    (E1 + 143 E2 + 11 E3);
      F4 = 99/7     (E1 - 130 E2 + 4 E3);
      F6 = 5577/350 (E1 + 35 E2 - 7 E3);
      Return[{F0, F2, F4, F6}];
    )
  ];

  Electrostatic::usage = "Electrostatic[{numE, NKSL, NKSLp}] returns
    the LS reduced matrix element for repulsion matrix element for
    equivalent electrons. See equation 2-79 in Wybourne (1965). The
    option \"Coefficients\" can be set to \"Slater\" or \"Racah\". If
    set to \"Racah\" then E_k parameters and e^k operators are assumed
    , otherwise the Slater integrals F^k and operators f_k. The
    default is \"Slater\".";
  Options[Electrostatic] = {"Coefficients" -> "Slater"};
  Electrostatic[{numE_, NKSL_, NKSLp_}, OptionsPattern[]] := Module[
    {fsub0, fsub2, fsub4, fsub6,
     esub0, esub1, esub2, esub3,
     fsup0, fsup2, fsup4, fsup6,
     eMatrixVal, orbital},
    (
      orbital = 3;
      Which[
        OptionValue["Coefficients"] == "Slater",
        (
          fsub0 = fsubk[numE, orbital, NKSL, NKSLp, 0];
          fsub2 = fsubk[numE, orbital, NKSL, NKSLp, 2];
          fsub4 = fsubk[numE, orbital, NKSL, NKSLp, 4];
          fsub6 = fsubk[numE, orbital, NKSL, NKSLp, 6];
          eMatrixVal = fsub0*F0 + fsub2*F2 + fsub4*F4 + fsub6*F6;
        ),
        OptionValue["Coefficients"] == "Racah",
        (
          fsup0 = fsupk[numE, orbital, NKSL, NKSLp, 0];
          fsup2 = fsupk[numE, orbital, NKSL, NKSLp, 2];
          fsup4 = fsupk[numE, orbital, NKSL, NKSLp, 4];
          fsup6 = fsupk[numE, orbital, NKSL, NKSLp, 6];
          esub0 = fsup0;
          esub1 = 9/7*fsup0 +   1/42*fsup2 +   1/77*fsup4 +  1/462*
  fsup6;
          esub2 =             143/42*fsup2 - 130/77*fsup4 + 35/462*
  fsup6;
          esub3 =              11/42*fsup2 + 4/77*fsup4   -  7/462*
  fsup6;
          eMatrixVal = esub0*E0 + esub1*E1 + esub2*E2 + esub3*E3;
        )
      ];
      Return[eMatrixVal];
    )
  ];

  GenerateElectrostaticTable::usage = "GenerateElectrostaticTable[
    numEmax] can be used to generate the table for the electrostatic
    interaction from f^1 to f^numEmax. If the option \"Export\" is set
     to True then the resulting data is saved to ./data/
    ElectrostaticTable.m.";
  Options[GenerateElectrostaticTable] = {"Export" -> True, "
    Coefficients" -> "Slater"};
  GenerateElectrostaticTable[numEmax_Integer:7, OptionsPattern[]] :=
```

```
    (
    ElectrostaticTable = Table[
      {numE, SL, SpLp} -> SimplifyFun[Electrostatic[{numE, SL, SpLp},
     "Coefficients" -> OptionValue["Coefficients"]]],
      {numE, 1, numEmax},
      {SL, AllowedNKSLTerms[numE]},
      {SpLp, AllowedNKSLTerms[numE]}
    ];
    ElectrostaticTable = Association[Flatten[ElectrostaticTable]];
    If[OptionValue["Export"],
      Export[FileNameJoin[{moduleDir, "data", "ElectrostaticTable.m"
    }],
      ElectrostaticTable];
    ];
    Return[ElectrostaticTable];
  );

  (* ###################### Electrostatic ###################### *)
  (* ########################################################### *)

  (* ########################################################### *)
  (* ########################## Bases ########################## *)

  BasisTableGenerator::usage = "BasisTableGenerator[numE] returns an
    association whose keys are triples of the form {numE, J} and whose
     values are lists having the basis elements that correspond to {
    numE, J}.";
  BasisTableGenerator[numE_] := Module[
    {energyStatesTable, allowedJ, J, Jp},
    (
      energyStatesTable = <||>;
      allowedJ = AllowedJ[numE];
      Do[
      (
        energyStatesTable[{numE, J}] = EnergyStates[numE, J];
      ),
      {Jp, allowedJ},
      {J,  allowedJ}];
      Return[energyStatesTable]
    )
  ];

  BasisLSJMJ::usage = "BasisLSJMJ[numE] returns the ordered basis in
    L-S-J-MJ with the total orbital angular momentum L and total spin
    angular momentum S coupled together to form J. The function
    returns a list with each element representing the quantum numbers
    for each basis vector. Each element is of the form {SL (string in
    spectroscopic notation),J, MJ}.
  The option \"AsAssociation\" can be set to True to return the basis
     as an association with the keys corresponding to values of J and
    the values lists with the corresponding {L, S, J, MJ} list. The
    default of this option is False.
  ";
  Options[BasisLSJMJ] = {"AsAssociation" -> False};
  BasisLSJMJ[numE_, OptionsPattern[]] := Module[
    {energyStatesTable, basis, idx1},
    (
      energyStatesTable = BasisTableGenerator[numE];
      basis = Table[
        energyStatesTable[{numE, AllowedJ[numE][[idx1]]}],
        {idx1, 1, Length[AllowedJ[numE]]}];
      basis = Flatten[basis, 1];
      If[OptionValue["AsAssociation"],
        (
          Js    = AllowedJ[numE];
          basis = Table[(J -> Select[basis, #[[2]] == J &]), {J, Js
    }];
          basis = Association[basis];
        )
      ];
      Return[basis]
    )
  ];

  BasisLSJ::usage = "BasisLSJ[numE] returns the level basis LSJ. The
    function returns a list with each element representing the quantum
```

```
    numbers for each basis vector. Each element is of the form {SL (string in spectroscopic notation), J}.
The option \"AsAssociation\" can be set to True to return the basis as an association with the keys being the allowed J values. The default is False.
    ";
Options[BasisLSJ]={"AsAssociation"->False};
BasisLSJ[numE_,OptionsPattern[]] := Module[
  {Js, basis},
  (
    Js    = AllowedJ[numE];
    basis = BasisLSJMJ[numE,"AsAssociation"->False];
    basis = DeleteDuplicates[{#[[1]],#[[2]]}& /@ basis];
    If[OptionValue["AsAssociation"],
      (
        basis = Association @ Table[(J->Select[basis,#[[2]]==J&]),{J,Js}]
      )
    ];
    Return[basis];
  )
];

(* ########################## Bases ########################## *)
(* ########################################################### *)

(* ########################################################### *)
(* ########### Coefficients of Fracional Parentage ########### *)

GenerateCFP::usage = "GenerateCFP[] generates the association for the coefficients of fractional parentage. Result is exported to the file ./data/CFP.m. The coefficients of fractional parentage are taken beyond the half-filled shell using the phase convention determined by the option \"PhaseFunction\". The default is \"NK\" which corresponds to the phase convention of Nielson and Koster. The other option is \"Judd\" which corresponds to the phase convention of Judd.";
Options[GenerateCFP] = {"Export" -> True, "PhaseFunction"-> "NK"};
GenerateCFP[OptionsPattern[]] := (
  CFP = Table[
    {numE, NKSL} -> First[CFPTerms[numE, NKSL]],
    {numE, 1, 7},
    {NKSL, AllowedNKSLTerms[numE]}];
  CFP = Association[CFP];
  (* Go all the way to f14 *)
  CFP = CFPExpander["Export" -> False, "PhaseFunction"->OptionValue["PhaseFunction"]];
  If[OptionValue["Export"],
    Export[FileNameJoin[{moduleDir, "data", "CFPs.m"}], CFP];
  ];
  Return[CFP];
);

JuddCFPPhase::usage = "Phase between conjugate coefficients of fractional parentage according to Velkov's thesis, page 40.";
JuddCFPPhase[parent_, parentS_, parentL_, daughterS_, daughterL_, parentSeniority_, daughterSeniority_] := Module[
  {spin, orbital, expo, phase},
  (
    {spin, orbital} = {1/2, 3};
    expo = (
        (parentS + parentL + daughterS + daughterL) -
        (orbital + spin) +
        1/2 * (parentSeniority + daughterSeniority - 1)
    );
    phase = Phaser[-expo];
    Return[phase];
  )
];

NKCFPPhase::usage = "Phase between conjugate coefficients of fractional parentage according to Nielson and Koster page viii. Note that there is a typo on there the expression for zeta should be (-1)^((v-1)/2) instead of (-1)^(v - 1/2).";
NKCFPPhase[parent_, parentS_, parentL_, daughterS_, daughterL_, parentSeniority_, daughterSeniority_] := Module[
```

```
    {spin, orbital, expo, phase},
    (
      {spin, orbital} = {1/2, 3};
      expo = (
          (parentS + parentL + daughterS + daughterL) -
          (orbital + spin)
      );
      phase = Phaser[-expo];
      If[parent == 2*orbital,
          phase = phase * Phaser[(daughterSeniority-1)/2]];
      Return[phase];
    )
  ];

  Options[CFPExpander] = {"Export" -> True, "PhaseFunction" -> "NK"};
  CFPExpander::usage = "Using the coefficients of fractional
    parentage up to f7 this function calculates them up to f14.
  The coefficients of fractional parentage are taken beyond the half-
    filled shell using the phase convention determined by the option \
    "PhaseFunction\". The default is \"NK\" which corresponds to the
    phase convention of Nielson and Koster. The other option is \"Judd
    \" which corresponds to the phase convention of Judd. The result
    is exported to the file ./data/CFPs_extended.m.";
  CFPExpander[OptionsPattern[]] := Module[
    {orbital, halfFilled, fullShell, parentMax, PhaseFun,
    complementaryCFPs, daughter, conjugateDaughter,
    conjugateParent, parentTerms, daughterTerms,
    parentCFPs, daughterSeniority, daughterS, daughterL,
    parentCFP, parentTerm, parentCFPval,
    parentS, parentL, parentSeniority, phase, prefactor,
    newCFPval, key, extendedCFPs, exportFname},
    (
      orbital    = 3;
      halfFilled = 2 * orbital + 1;
      fullShell  = 2 * halfFilled;
      parentMax  = 2 * orbital;

      PhaseFun  = <|
          "Judd" -> JuddCFPPhase,
          "NK" -> NKCFPPhase|>[OptionValue["PhaseFunction"]];
      PrintTemporary["Calculating CFPs using the phase system from ",
     PhaseFun];
      (* Initialize everything with lists to be filled in the next Do
    *)
      complementaryCFPs =
          Table[
          ({numE, term} -> {term}),
          {numE, halfFilled + 1, fullShell - 1, 1},
          {term, AllowedNKSLTerms[numE]
          }];
      complementaryCFPs = Association[Flatten[complementaryCFPs]];
      Do[(
          daughter          = parent + 1;
          conjugateDaughter = fullShell - parent;
          conjugateParent   = conjugateDaughter - 1;
          parentTerms       = AllowedNKSLTerms[parent];
          daughterTerms     = AllowedNKSLTerms[daughter];
          Do[
          (
              parentCFPs             = Rest[CFP[{daughter,
    daughterTerm}]];
              daughterSeniority      = Seniority[daughterTerm];
              {daughterS, daughterL} = FindSL[daughterTerm];
              Do[
              (
                  {parentTerm, parentCFPval} = parentCFP;
                  {parentS, parentL}         = FindSL[parentTerm];
                  parentSeniority            = Seniority[parentTerm];
                  phase = PhaseFun[parent, parentS, parentL,
                                   daughterS, daughterL,
                                   parentSeniority, daughterSeniority
    ];
                  prefactor = (daughter * TPO[daughterS, daughterL])
    /
                              (conjugateDaughter * TPO[parentS,
    parentL]);
```

```
                    prefactor = Sqrt[prefactor];
                    newCFPval = phase * prefactor * parentCFPval;
                    key = {conjugateDaughter, parentTerm};
                    complementaryCFPs[key] = Append[complementaryCFPs[key], {daughterTerm, newCFPval}]
                ),
                {parentCFP, parentCFPs}
                ]
            ),
            {daughterTerm, daughterTerms}
            ]
            ),
        {parent, 1, parentMax}
        ];

        complementaryCFPs[{14, "1S"}] = {"1S", {"2F",1}};
        extendedCFPs          = Join[CFP, complementaryCFPs];
        If[OptionValue["Export"];,
        (
            exportFname = FileNameJoin[{moduleDir, "data", "CFPs_extended.m"}];
            Print["Exporting to ", exportFname];
            Export[exportFname, extendedCFPs];
        )
        ];
        Return[extendedCFPs];
    )
    ];

    GenerateCFPTable::usage = "GenerateCFPTable[] generates the table for the coefficients of fractional parentage. If the optional parameter \"Export\" is set to True then the resulting data is saved to ./data/CFPTable.m.
    The data being parsed here is the file attachment B1F_ALL.TXT which comes from Velkov's thesis.";
    Options[GenerateCFPTable] = {"Export" -> True};
    GenerateCFPTable[OptionsPattern[]] := Module[
      {rawText, rawLines, leadChar, configIndex, line, daughter,
      lineParts, numberCode, parsedNumber, toAppend, CFPTablefname},
      (
        CleanWhitespace[string_]      := StringReplace[string, RegularExpression["\\s+"]->" "];
        AddSpaceBeforeMinus[string_] := StringReplace[string, RegularExpression["(?<!\\s)-"]->" -"];
        ToIntegerOrString[list_]      := Map[If[StringMatchQ[#, NumberString], ToExpression[#], #] &, list];
        CFPTable       = ConstantArray[{},7];
        CFPTable[[1]] = {{"2F",{"1S",1}}};

        (* Cleaning before processing is useful *)
        rawText  = Import[FileNameJoin[{moduleDir, "data", "B1F_ALL.TXT"}]];
        rawLines = StringTrim/@StringSplit[rawText,"\n"];
        rawLines = Select[rawLines,#!=""&];
        rawLines = CleanWhitespace/@rawLines;
        rawLines = AddSpaceBeforeMinus/@rawLines;

        Do[(
          (* the first character can be used to identify the start of a block *)
          leadChar=StringTake[line,{1}];
          (* ..FN, N is at position 50 in that line *)
          If[leadChar=="[",
          (
            configIndex=ToExpression[StringTake[line,{50}]];
            Continue[];
          )
          ];
          (* Identify which daughter term is being listed *)
          If[StringContainsQ[line,"[DAUGHTER TERM]"],
            daughter=StringSplit[line,"["][[1]];
            CFPTable[[configIndex]]=Append[CFPTable[[configIndex]],{daughter}];
            Continue[];
          ];
          (* Once we get here we are already parsing a row with
```

```
    coefficient data *)
1108        lineParts    = StringSplit[line," "];
1109        parent       = lineParts[[1]];
1110        numberCode   = ToIntegerOrString[lineParts[[3;;]]];
1111        parsedNumber = SquarePrimeToNormal[numberCode];
1112        toAppend     = {parent,parsedNumber};
1113        CFPTable[[configIndex]][[-1]] = Append[CFPTable[[configIndex
    ]][[-1]], toAppend]
1114      ),
1115      {line,rawLines}];
1116      If[OptionValue["Export"],
1117        (
1118        CFPTablefname = FileNameJoin[{moduleDir, "data", "CFPTable.m"
    }];
1119        Export[CFPTablefname, CFPTable];
1120        )
1121      ];
1122      Return[CFPTable];
1123    )
1124  ];
1125
1126  GenerateCFPAssoc::usage = "GenerateCFPAssoc[export] converts the
      coefficients of fractional parentage into an association in which
      zero values are explicit. If \"Export\" is set to True, the
      association is exported to the file /data/CFPAssoc.m. This
      function requires that the association CFP be defined.";
1127  Options[GenerateCFPAssoc] = {"Export" -> True};
1128  GenerateCFPAssoc[OptionsPattern[]] := (
1129    CFPAssoc = Association[];
1130    Do[
1131      (daughterTerms = AllowedNKSLTerms[numE];
1132      parentTerms    = AllowedNKSLTerms[numE - 1];
1133      Do[
1134        (
1135        cfps = CFP[{numE, daughter}];
1136        cfps = cfps[[2 ;;]];
1137        parents = First /@ cfps;
1138        Do[
1139          (
1140          key = {numE, daughter, parent};
1141          cfp = If[
1142            MemberQ[parents, parent],
1143            (
1144              idx = Position[parents, parent][[1, 1]];
1145              cfps[[idx]][[2]]
1146            ),
1147            0
1148            ];
1149          CFPAssoc[key] = cfp;
1150          ),
1151          {parent, parentTerms}
1152          ]
1153        ),
1154        {daughter, daughterTerms}
1155        ]
1156      ),
1157      {numE, 1, 14}
1158      ];
1159    If[OptionValue["Export"],
1160      (
1161      CFPAssocfname = FileNameJoin[{moduleDir, "data", "CFPAssoc.m"
    }];
1162      Export[CFPAssocfname, CFPAssoc];
1163      )
1164    ];
1165    Return[CFPAssoc];
1166  );
1167
1168  CFPTerms::usage = "CFPTerms[numE] gives all the daughter and parent
      terms, together with the corresponding coefficients of fractional
      parentage, that correspond to the the f^n configuration.
1169  CFPTerms[numE, SL] gives all the daughter and parent terms,
      together with the corresponding coefficients of fractional
      parentage, that are compatible with the given string SL in the f^n
      configuration.
1170  CFPTerms[numE, L, S] gives all the daughter and parent terms,
```

```
    together with the corresponding coefficients of fractional
    parentage, that correspond to the given total orbital angular
    momentum L and total spin S n the f^n configuration. L being an
    integer, and S being integer or half-integer.
In all cases the output is in the shape of a list with enclosed
    lists having the format {daughter_term, {parent_term_1, CFP_1}, {
    parent_term_2, CFP_2}, ...}.
Only the one-body coefficients for f-electrons are provided.
In all cases it must be that 1 <= n <= 7.
These are according to the tables from Nielson & Koster.
";
CFPTerms[numE_] := Part[CFPTable, numE]
CFPTerms[numE_, SL_] := Module[
  {NKterms, CFPconfig},
  (
    NKterms   = {{}};
    CFPconfig = CFPTable[[numE]];
    Map[
      If[StringFreeQ[First[#], SL],
        Null,
        NKterms = Join[NKterms, {#}, 1]
      ] &,
    CFPconfig
    ];
    NKterms = DeleteCases[NKterms, {}]
  )
];
CFPTerms[numE_, L_, S_] := Module[
{NKterms, SL, CFPconfig},
(
  SL = StringJoin[ToString[2 S + 1], PrintL[L]];
  NKterms = {{}};
  CFPconfig = Part[CFPTable, numE];
  Map[
    If[StringFreeQ[First[#], SL],
      Null,
      NKterms = Join[NKterms, {#}, 1]
    ]&,
  CFPconfig
  ];
  NKterms = DeleteCases[NKterms, {}]
)
];

(* ########### Coefficients of Fracional Parentage ########### *)
(* ############################################################ *)

(* ############################################################ *)
(* ####################### Spin Orbit ######################### *)

SpinOrbit::usage = "SpinOrbit[numE, SL, SpLp, J] returns the LSJ
    reduced matrix element ζ <SL, J|L.S|SpLp, J>. These are given as a
     function of ζ. This function requires that the association
    ReducedV1kTable be defined.
See equations 2-106 and 2-109 in Wybourne (1965). Equivalently see
    eqn. 12.43 in TASS.";
SpinOrbit[numE_, SL_, SpLp_, J_] := Module[
  {S, L, Sp, Lp, orbital, sign, prefactor, val},
  (
    orbital   = 3;
    {S, L}    = FindSL[SL];
    {Sp, Lp}  = FindSL[SpLp];
    prefactor = Sqrt[orbital * (orbital+1) * (2*orbital+1)] *
                SixJay[{L, Lp, 1}, {Sp, S, J}];
    sign      = Phaser[J + L + Sp];
    val       = sign * prefactor * ζ * ReducedV1kTable[{numE, SL,
  SpLp, 1}];
    Return[val];
  )
];

GenerateSpinOrbitTable::usage = "GenerateSpinOrbitTable[nmax]
    computes the matrix elements for the spin-orbit interaction for f^
    n configurations up to n = nmax. The function returns an
    association whose keys are lists of the form {n, SL, SpLp, J}. If
    export is set to True, then the result is exported to the data
```

```
    subfolder for the folder in which this package is in. It requires
    ReducedV1kTable to be defined.";
1232  Options[GenerateSpinOrbitTable] = {"Export" -> True};
1233  GenerateSpinOrbitTable[nmax_Integer:7, OptionsPattern[]] := Module[
1234    {numE, J, SL, SpLp, exportFname},
1235    (
1236      SpinOrbitTable =
1237        Table[
1238          {numE, SL, SpLp, J} -> SpinOrbit[numE, SL, SpLp, J],
1239        {numE, 1, nmax},
1240        {J, MinJ[numE], MaxJ[numE]},
1241        {SL,   Map[First, AllowedNKSLforJTerms[numE, J]]},
1242        {SpLp, Map[First, AllowedNKSLforJTerms[numE, J]]}
1243        ];
1244      SpinOrbitTable = Association[SpinOrbitTable];
1245
1246      exportFname = FileNameJoin[{moduleDir, "data", "SpinOrbitTable.
    m"}];
1247      If[OptionValue["Export"],
1248        (
1249          Print["Exporting to file "<>ToString[exportFname]];
1250          Export[exportFname, SpinOrbitTable];
1251        )
1252      ];
1253      Return[SpinOrbitTable];
1254    )
1255  ];
1256
1257  (* ######################## Spin Orbit ########################### *)
1258  (* ############################################################## *)
1259
1260  (* ############################################################## *)
1261  (* ################## Three Body Operators ###################### *)
1262
1263  ParseJudd1984::usage = "This function parses the data from tables 1
      and 2 of Judd from Judd, BR, and MA Suskin. \"Complete Set of
    Orthogonal Scalar Operators for the Configuration f^3\". JOSA B 1,
      no. 2 (1984): 261-65.";
1264  Options[ParseJudd1984] = {"Export" -> False};
1265  ParseJudd1984[OptionsPattern[]] := (
1266    ParseJuddTab1[str_] := (
1267      strR = ToString[str];
1268      strR = StringReplace[strR, ".5" -> "^(1/2)"];
1269      num = ToExpression[strR];
1270      sign = Sign[num];
1271      num = sign*Simplify[Sqrt[num^2]];
1272      If[Round[num] == num, num = Round[num]];
1273      Return[num]);
1274
1275    (* Parse table 1 from Judd 1984 *)
1276    judd1984Fname1 = FileNameJoin[{moduleDir, "data", "Judd1984-1.csv
    "}];
1277    data = Import[judd1984Fname1, "CSV", "Numeric" -> False];
1278    headers = data[[1]];
1279    data = data[[2 ;;]];
1280    data = Transpose[data];
1281    \[Psi] = Select[data[[1]], # != "" &];
1282    \[Psi]p = Select[data[[2]], # != "" &];
1283    matrixKeys = Transpose[{\[Psi], \[Psi]p}];
1284    data = data[[3 ;;]];
1285    cols = Table[ParseJuddTab1 /@ Select[col, # != "" &], {col, data
    }];
1286    cols = Select[cols, Length[#] == 21 &];
1287    tab1 = Prepend[Prepend[cols, \[Psi]p], \[Psi]];
1288    tab1 = Transpose[Prepend[Transpose[tab1], headers]];
1289
1290    (* Parse table 2 from Judd 1984 *)
1291    judd1984Fname2 = FileNameJoin[{moduleDir, "data", "Judd1984-2.csv
    "}];
1292    data = Import[judd1984Fname2, "CSV", "Numeric" -> False];
1293    headers = data[[1]];
1294    data = data[[2 ;;]];
1295    data = Transpose[data];
1296    {operatorLabels, WUlabels, multiFactorSymbols, multiFactorValues}
     = data[[;; 4]];
1297    multiFactorValues = ParseJuddTab1 /@ multiFactorValues;
```

```
    multiFactorValues = AssociationThread[multiFactorSymbols ->
    multiFactorValues];

    (*scale values of table 1 given the values in table 2*)
    oppyS = {};
    normalTable =
      Table[header = col[[1]];
        If[StringContainsQ[header, " "],
          (
            multiplierSymbol = StringSplit[header, " "][[1]];
            multiplierValue = multiFactorValues[multiplierSymbol];
            operatorSymbol = StringSplit[header, " "][[2]];
            oppyS = Append[oppyS, operatorSymbol];
          ),
          (
            multiplierValue = 1;
            operatorSymbol = header;
          )
        ];
        normalValues = 1/multiplierValue*col[[2 ;;]];
        Join[{operatorSymbol}, normalValues], {col, tab1[[3 ;;]]}
      ];

    (*Create an association for the reduced matrix elements in the f
    ^3 config*)
    juddOperators = Association[];
    Do[(
      col      = normalTable[[colIndex]];
      opLabel  = col[[1]];
      opValues = col[[2 ;;]];
      opMatrix = AssociationThread[matrixKeys -> opValues];
      Do[(
        opMatrix[Reverse[mKey]] = opMatrix[mKey]
        ),
      {mKey, matrixKeys}
      ];
      juddOperators[{3, opLabel}] = opMatrix),
      {colIndex, 1, Length[normalTable]}
    ];

    (* special case of t2 in f3 *)
    (* this is the same as getting the reduced matrix elements from
    Judd 1966 *)
    numE = 3;
    e3Op      = juddOperators[{3, "e_{3}"}];
    t2prime   = juddOperators[{3, "t_{2}^{'}"}];
    prefactor = 1/(70 Sqrt[2]);
    t2Op = (# -> (t2prime[#] + prefactor*e3Op[#])) & /@ Keys[t2prime
    ];
    t2Op = Association[t2Op];
    juddOperators[{3, "t_{2}"}] = t2Op;

    (*Special case of t11 in f3*)
    t11 = juddOperators[{3, "t_{11}"}];
    eβprimeOp = juddOperators[{3, "e_{\\beta}^{'}"}];
    t11primeOp = (# -> (t11[#] + Sqrt[3/385] eβprimeOp[#])) & /@ Keys
    [t11];
    t11primeOp = Association[t11primeOp];
    juddOperators[{3, "t_{11}^{'}"}] = t11primeOp;
    If[OptionValue["Export"],
      (
        (*export them*)
        PrintTemporary["Exporting ..."];
        exportFname = FileNameJoin[{moduleDir, "data", "juddOperators
    .m"}];
        Export[exportFname, juddOperators];
      )
    ];
    Return[juddOperators];
  );

  GenerateThreeBodyTables::usage = "This function generates the
    reduced matrix elements for the three body operators using the
    coefficients of fractional parentage, including those beyond f^7."
    ;
  Options[GenerateThreeBodyTables] = {"Export" -> False};
```

```
GenerateThreeBodyTables[OptionsPattern[]] := (
  tiKeys        = (StringReplace[ToString[#], {"T" -> "t_{", "p" -> "}^{'"}] <> "}") & /@ TSymbols;
  TSymbolsAssoc = AssociationThread[tiKeys -> TSymbols];
  juddOperators = ParseJudd1984[];
  (* op3MatrixElement[SL, SpLp, opSymbol] returns the value for the reduced matrix element of the operator opSymbol for the terms {SL, SpLp} in the f^3 configuration. *)
  op3MatrixElement[SL_, SpLp_, opSymbol_] := (
    jOP = juddOperators[{3, opSymbol}];
    key = {SL, SpLp};
    val = If[MemberQ[Keys[jOP], key],
      jOP[key],
      0];
    Return[val];
  );
  (* ti: This is the implementation of formula (2) in Judd & Suskin 1984. It computes the reduced matrix elements of ti in f^n by using the reduced matrix elements in f^3 and the coefficients of fractional parentage. If the option \"Fast\" is set to True then the values for n>7 are simply computed as the negatives of the values in the complementary configuration; this except for t2 and t11 which are treated as special cases. *)
  Options[ti] = {"Fast" -> True};
  ti[nE_, SL_, SpLp_, tiKey_, opOrder_ : 3, OptionsPattern[]] :=
  Module[
    {nn, S, L, Sp, Lp,
    cfpSL, cfpSpLp,
    parentSL, parentSpLp,
    tnk, tnks},
    (
      {S, L}   = FindSL[SL];
      {Sp, Lp} = FindSL[SpLp];
      fast     = OptionValue["Fast"];
      numH     = 14 - nE;
      If[fast && Not[MemberQ[{"t_{2}","t_{11}"},tiKey]] && nE > 7,
        Return[-tktable[{numH, SL, SpLp, tiKey}]]
      ];
      If[(S == Sp && L == Lp),
      (
        cfpSL   = CFP[{nE, SL}];
        cfpSpLp = CFP[{nE, SpLp}];
        tnks = Table[(
            parentSL   = cfpSL[[nn, 1]];
            parentSpLp = cfpSpLp[[mm, 1]];
            cfpSL[[nn, 2]] * cfpSpLp[[mm, 2]] *
            tktable[{nE - 1, parentSL, parentSpLp, tiKey}]
            ),
            {nn, 2, Length[cfpSL]},
            {mm, 2, Length[cfpSpLp]}
            ];
        tnk = Total[Flatten[tnks]];
        ),
      tnk = 0;
      ];
      Return[ nE / (nE - opOrder) * tnk];
    )
  ];
  (* Calculate the reduced matrix elements of t^i for n up to 14 *)
  tktable = <||>;
  Do[(
    Do[(
      tkValue = Which[numE <= 2,
        (*Initialize n=1,2 with zeros*)
        0,
        numE == 3,
        (* Grab matrix elem in f^3 from Judd 1984 *)
        SimplifyFun[op3MatrixElement[SL, SpLp, opKey]],
        True,
        SimplifyFun[ti[numE, SL, SpLp, opKey, If[opKey == "e_{3}", 2, 3]]]
        ];
      tktable[{numE, SL, SpLp, opKey}] = tkValue;
      ),
    {SL, AllowedNKSLTerms[numE]},
    {SpLp, AllowedNKSLTerms[numE]},
```

```
      {opKey, Append[tiKeys, "e_{3}"]}
      ];
      PrintTemporary[StringJoin["\[ScriptF]", ToString[numE], "
    configuration complete"]];
      ),
      {numE, 1, 14}
    ];

    (* Now use those reduced matrix elements to determine their sum
    as weighted by their corresponding strengths Ti *)
    ThreeBodyTable = <||>;
    Do[
      Do[
      (
        ThreeBodyTable[{numE, SL, SpLp}] = (
          Sum[(
            If[tiKey == "t_{2}", t2Switch, 1] *
            tktable[{numE, SL, SpLp, tiKey}] *
            TSymbolsAssoc[tiKey] +
            If[tiKey == "t_{2}", 1 - t2Switch, 0] *
            (-tktable[{14 - numE, SL, SpLp, tiKey}]) *
            TSymbolsAssoc[tiKey]
            ),
          {tiKey, tiKeys}
          ]
        );
      ),
      {SL, AllowedNKSLTerms[numE]},
      {SpLp, AllowedNKSLTerms[numE]}
      ];
    PrintTemporary[StringJoin["\[ScriptF]", ToString[numE], " matrix
    complete"]];,
    {numE, 1, 7}
    ];

    ThreeBodyTables = Table[(
      terms = AllowedNKSLTerms[numE];
      singleThreeBodyTable =
        Table[
          {SL, SLp} -> ThreeBodyTable[{numE, SL, SLp}],
          {SL, terms},
          {SLp, terms}
        ];
      singleThreeBodyTable  = Flatten[singleThreeBodyTable];
      singleThreeBodyTables = Table[(
          notNullPosition = Position[TSymbols, notNullSymbol][[1,
    1]];
          reps = ConstantArray[0, Length[TSymbols]];
          reps[[notNullPosition]] = 1;
          rep = AssociationThread[TSymbols -> reps];
          notNullSymbol -> Association[(singleThreeBodyTable /. rep)]
          ),
        {notNullSymbol, TSymbols}
        ];
      singleThreeBodyTables = Association[singleThreeBodyTables];
      numE -> singleThreeBodyTables),
      {numE, 1, 7}
  ];

  ThreeBodyTables = Association[ThreeBodyTables];
  If[OptionValue["Export"],
    (
      threeBodyTablefname = FileNameJoin[{moduleDir, "data", "
    ThreeBodyTable.m"}];
      Export[threeBodyTablefname, ThreeBodyTable];
      threeBodyTablesfname = FileNameJoin[{moduleDir, "data", "
    ThreeBodyTables.m"}];
      Export[threeBodyTablesfname, ThreeBodyTables];
    )
   ];
  Return[{ThreeBodyTable, ThreeBodyTables}];
  );

  ScalarOperatorProduct::usage = "ScalarOperatorProduct[op1, op2,
    numE] calculated the innerproduct between the two scalar operators
     op1 and op2.";
```

```
ScalarOperatorProduct[op1_, op2_, numE_] := Module[
  {terms, S, L, factor, term1, term2},
  (
    terms = AllowedNKSLTerms[numE];
    Simplify[
      Sum[(
        {S, L} = FindSL[term1];
        factor = TPO[S, L];
        factor * op1[{term1, term2}] * op2[{term2, term1}]
        ),
      {term1, terms},
      {term2, terms}
      ]
    ]
  )
];

(* ################## Three Body Operators ##################### *)
(* ############################################################## *)

(* ############################################################## *)
(* ################### Reduced SOO and ECSO ##################### *)

ReducedT11inf2::usage = "ReducedT11inf2[SL, SpLp] returns the
  reduced matrix element of the scalar component of the double
  tensor T11 for the given SL terms SL, SpLp.
Data used here for m0, m2, m4 is from Table II of Judd, BR, HM
  Crosswhite, and Hannah Crosswhite. Intra-Atomic Magnetic
  Interactions for f Electrons. Physical Review 169, no. 1 (1968):
  130.
";
ReducedT11inf2[SL_, SpLp_] := Module[
  {T11inf2},
  (
    T11inf2 = <|
      {"1S", "3P"} -> 6 M0 + 2 M2 + 10/11 M4,
      {"3P", "3P"} -> -36 M0 - 72 M2 - 900/11 M4,
      {"3P", "1D"} -> -Sqrt[(2/15)] (27 M0 + 14 M2 + 115/11 M4),
      {"1D", "3F"} -> Sqrt[2/5] (23 M0 + 6 M2 - 195/11 M4),
      {"3F", "3F"} -> 2 Sqrt[14] (-15 M0 - M2 + 10/11 M4),
      {"3F", "1G"} -> Sqrt[11] (-6 M0 + 64/33 M2 - 1240/363 M4),
      {"1G", "3H"} -> Sqrt[2/5] (39 M0 - 728/33 M2 - 3175/363 M4),
      {"3H", "3H"} -> 8/Sqrt[55] (-132 M0 + 23 M2 + 130/11 M4),
      {"3H", "1I"} -> Sqrt[26] (-5 M0 - 30/11 M2 - 375/1573 M4)
      |>;
    Which[
      MemberQ[Keys[T11inf2],{SL,SpLp}],
        Return[T11inf2[{SL,SpLp}]],
      MemberQ[Keys[T11inf2],{SpLp,SL}],
        Return[T11inf2[{SpLp,SL}]],
      True,
        Return[0]
      ]
  )
];

Reducedt11inf2::usage = "Reducedt11inf2[SL, SpLp] returns the
  reduced matrix element in f^2 of the double tensor operator t11
  for the corresponding given terms {SL, SpLp}.
Values given here are those from Table VII of \"Judd, BR, HM
  Crosswhite, and Hannah Crosswhite. \"Intra-Atomic Magnetic
  Interactions for f Electrons.\" Physical Review 169, no. 1 (1968):
   130.\"
";
Reducedt11inf2[SL_, SpLp_] := Module[
  {t11inf2},
  (
    t11inf2 = <|
      {"1S", "3P"} -> -2 P0 - 105 P2 - 231 P4 - 429 P6,
      {"3P", "3P"} -> -P0 - 45 P2 - 33 P4 + 1287 P6,
      {"3P", "1D"} -> Sqrt[15/2] (P0 + 32 P2 - 33 P4 - 286 P6),
      {"1D", "3F"} -> Sqrt[10] (-P0 - 9/2 P2 + 66 P4 - 429/2 P6),
      {"3F", "3F"} -> Sqrt[14] (-P0 + 10 P2 + 33 P4 + 286 P6),
      {"3F", "1G"} -> Sqrt[11] (P0 - 20 P2 + 32 P4 - 104 P6),
      {"1G", "3H"} -> Sqrt[10] (-P0 + 55/2 P2 - 23 P4 - 65/2 P6),
      {"3H", "3H"} -> Sqrt[55] (-P0 + 25 P2 + 51 P4 + 13 P6),
```

```
        {"3H", "1I"} -> Sqrt[13/2] (P0 - 21 P4 - 6 P6)
        |>;
      Which[
        MemberQ[Keys[t11inf2],{SL,SpLp}],
          Return[t11inf2[{SL,SpLp}]],
        MemberQ[Keys[t11inf2],{SpLp,SL}],
          Return[t11inf2[{SpLp,SL}]],
        True,
          Return[0]
      ]
    )
];

ReducedSOOandECSOinf2::usage = "ReducedSOOandECSOinf2[SL, SpLp]
  returns the reduced matrix element corresponding to the operator (
  T11 + t11 - a13 * z13 / 6) for the terms {SL, SpLp}. This
  combination of operators corresponds to the spin-other-orbit plus
  ECSO interaction.
The T11 operator corresponds to the spin-other-orbit interaction,
  and the t11 operator (associated with electrostatically-correlated
   spin-orbit) originates from configuration interaction analysis.
  To their sum a factor proportional to the operator z13 is
  subtracted since its effect is redundant to the spin-orbit
  interaction. The factor of 1/6 is not on Judd's 1968 paper, but it
   is on \"Chen, Xueyuan, Guokui Liu, Jean Margerie, and Michael F
  Reid. \"A Few Mistakes in Widely Used Data Files for Fn
  Configurations Calculations.\" Journal of Luminescence 128, no. 3
  (2008): 421-27\".
The values for the reduced matrix elements of z13 are obtained from
   Table IX of the same paper. The value for a13 is from table VIII.
Rigurously speaking the Pk parameters here are subscripted. The
  conversion to superscripted parameters is performed elsewhere with
   the Prescaling replacement rules.
";
ReducedSOOandECSOinf2[SL_, SpLp_]  := Module[
  {a13, z13, z13inf2, matElement, redSOOandECSOinf2},
  (
    a13 = (-33 M0 + 3 M2 + 15/11 M4 -
          6 P0 + 3/2 (35 P2 + 77 P4 + 143 P6));
    z13inf2 = <|
        {"1S","3P"} -> 2,
        {"3P","3P"} -> 1,
        {"3P","1D"} -> -Sqrt[(15/2)],
        {"1D","3F"} -> Sqrt[10],
        {"3F","3F"} -> Sqrt[14],
        {"3F","1G"} -> -Sqrt[11],
        {"1G","3H"} -> Sqrt[10],
        {"3H","3H"} -> Sqrt[55],
        {"3H","1I"} -> -Sqrt[(13/2)]
        |>;
    matElement = Which[
      MemberQ[Keys[z13inf2],{SL,SpLp}],
        z13inf2[{SL,SpLp}],
      MemberQ[Keys[z13inf2],{SpLp,SL}],
        z13inf2[{SpLp,SL}],
      True,
        0
    ];
    redSOOandECSOinf2 = (
        ReducedT11inf2[SL, SpLp] +
        Reducedt11inf2[SL, SpLp] -
        a13 / 6 * matElement
    );
    redSOOandECSOinf2 = SimplifyFun[redSOOandECSOinf2];
    Return[redSOOandECSOinf2];
  )
];

ReducedSOOandECSOinfn::usage = "ReducedSOOandECSOinfn[numE, SL,
  SpLp] calculates the reduced matrix elements of the (spin-other-
  orbit + ECSO) operator for the f^numE configuration corresponding
  to the terms SL and SpLp. This is done recursively, starting from
  tabulated values for f^2 from \"Judd, BR, HM Crosswhite, and
  Hannah Crosswhite. \"Intra-Atomic Magnetic Interactions for f
  Electrons.\" Physical Review 169, no. 1 (1968): 130.\", and by
  using equation (4) of that same paper.
```

```
";
ReducedSOOandECSOinfn[numE_, SL_, SpLp_] := Module[
  {spin, orbital, t, S, L, Sp, Lp,
  idx1, idx2, cfpSL, cfpSpLp, parentSL,
  Sb, Lb, Sbp, Lbp, parentSpLp, funval},
  (
    {spin, orbital} = {1/2, 3};
    {S, L}   = FindSL[SL];
    {Sp, Lp} = FindSL[SpLp];
    t = 1;
    cfpSL    = CFP[{numE, SL}];
    cfpSpLp  = CFP[{numE, SpLp}];
    funval   = Sum[
      (
        parentSL = cfpSL[[idx2, 1]];
        parentSpLp = cfpSpLp[[idx1, 1]];
        {Sb, Lb}   = FindSL[parentSL];
        {Sbp, Lbp} = FindSL[parentSpLp];
        phase = Phaser[Sb + Lb + spin + orbital + Sp + Lp];
        (
          phase *
          cfpSpLp[[idx1, 2]] * cfpSL[[idx2, 2]] *
          SixJay[{S, t, Sp}, {Sbp, spin, Sb}] *
          SixJay[{L, t, Lp}, {Lbp, orbital, Lb}] *
          SOOandECSOLSTable[{numE - 1, parentSL, parentSpLp}]
        )
      ),
    {idx1, 2, Length[cfpSpLp]},
    {idx2, 2, Length[cfpSL]}
    ];
    funval *= numE / (numE - 2) * Sqrt[TPO[S, Sp, L, Lp]];
    Return[funval];
  )
];

GenerateSOOandECSOLSTable::usage = "GenerateSOOandECSOLSTable[nmax]
   generates the LS reduced matrix elements of the spin-other-orbit
  + ECSO for the f^n configurations up to n=nmax. The values for n=1
   and n=2 are taken from \"Judd, BR, HM Crosswhite, and Hannah
  Crosswhite. \"Intra-Atomic Magnetic Interactions for f Electrons.\
  " Physical Review 169, no. 1 (1968): 130.\", and the values for n
  >2 are calculated recursively using equation (4) of that same
  paper. The values are then exported to a file \"
  ReducedSOOandECSOLSTable.m\" in the data folder of this module.
  The values are also returned as an association.";
Options[GenerateSOOandECSOLSTable] = {"Progress" -> True, "Export"
  -> True};
GenerateSOOandECSOLSTable[nmax_Integer, OptionsPattern[]] := (
  If[And[OptionValue["Progress"], frontEndAvailable],
    (
      numItersai = Association[Table[numE->Length[AllowedNKSLTerms[
  numE]]^2, {numE, 1, nmax}]];
      counters   = Association[Table[numE->0, {numE, 1, nmax}]];
      totalIters = Total[Values[numItersai[[1;;nmax]]]];
      template1  = StringTemplate["Iteration `numiter` of `
  totaliter`"];
      template2  = StringTemplate["`remtime` min remaining"];
  template3 = StringTemplate["Iteration speed = `speed` ms/it"];
      template4  = StringTemplate["Time elapsed = `runtime` min"];
      progBar = PrintTemporary[
        Dynamic[
          Pane[
            Grid[{
                  {Superscript["f", numE]},
                  {template1[<|"numiter"->numiter, "totaliter"->
  totalIters|>]},
                  {template4[<|"runtime"->Round[QuantityMagnitude[
  UnitConvert[(Now-startTime), "min"]], 0.1]|>]},
                  {template2[<|"remtime"->Round[QuantityMagnitude[
  UnitConvert[(Now-startTime)/(numiter)*(totalIters-numiter), "min"
  ]], 0.1]|>]},
                  {template3[<|"speed"->Round[QuantityMagnitude[Now
  -startTime, "ms"]/(numiter), 0.01]|>]}, {ProgressIndicator[Dynamic
  [numiter], {1, totalIters}]}
                },
                Frame->All
```

```
            ],
            Full,
            Alignment->Center
          ]
        ]
      ];
    )
  ];
  SOOandECSOLSTable = <||>;
  numiter    = 1;
  startTime = Now;
  Do[
    (
      numiter+= 1;
      SOOandECSOLSTable[{numE, SL, SpLp}] = Which[
        numE==1,
        0,
        numE==2,
        SimplifyFun[ReducedSOOandECSOinf2[SL, SpLp]],
        True,
        SimplifyFun[ReducedSOOandECSOinfn[numE,  SL, SpLp]]
      ];
    ),
  {numE, 1, nmax},
  {SL, AllowedNKSLTerms[numE]},
  {SpLp, AllowedNKSLTerms[numE]}
  ];
  If[And[OptionValue["Progress"], frontEndAvailable],
    NotebookDelete[progBar]];
  If[OptionValue["Export"],
    (fname = FileNameJoin[{moduleDir, "data", "
 ReducedSOOandECSOLSTable.m"}];
    Export[fname, SOOandECSOLSTable];
    )
  ];
  Return[SOOandECSOLSTable];
);

(* ################## Reduced SOO and ECSO ################### *)
(* ############################################################ *)

(* ############################################################ *)
(* ######################## Spin-Spin ########################## *)

ReducedT22inf2::usage = "ReducedT22inf2[SL, SpLp] returns the
  reduced matrix element of the scalar component of the double
  tensor T22 for the terms SL, SpLp in f^2.
Data used here for m0, m2, m4 is from Table I of Judd, BR, HM
  Crosswhite, and Hannah Crosswhite. Intra-Atomic Magnetic
  Interactions for f Electrons. Physical Review 169, no. 1 (1968):
  130.
";
ReducedT22inf2[SL_, SpLp_] := Module[
  {statePosition, PsiPsipStates, m0, m2, m4, Tkk2m},
  (
    T22inf2 = <|
    {"3P", "3P"} -> -12 M0 - 24 M2 - 300/11 M4,
    {"3P", "3F"} -> 8/Sqrt[3] (3 M0 + M2 - 100/11 M4),
    {"3F", "3F"} -> 4/3 Sqrt[14] (-M0 + 8 M2 - 200/11 M4),
    {"3F", "3H"} -> 8/3 Sqrt[11/2] (2 M0 - 23/11 M2 - 325/121 M4),
    {"3H", "3H"} -> 4/3 Sqrt[143] (M0 - 34/11 M2 - (1325/1573) M4)
    |>;
    Which[
      MemberQ[Keys[T22inf2],{SL,SpLp}],
        Return[T22inf2[{SL,SpLp}]],
      MemberQ[Keys[T22inf2],{SpLp,SL}],
        Return[T22inf2[{SpLp,SL}]],
      True,
        Return[0]
    ]
  )
];

ReducedT22infn::usage = "ReducedT22infn[n, SL, SpLp] calculates the
   reduced matrix element of the T22 operator for the f^n
  configuration corresponding to the terms SL and SpLp. This is the
```

```
    operator corresponding to the inter-electron between spin.
1742  It does this by using equation (4) of \"Judd, BR, HM Crosswhite,
    and Hannah Crosswhite. \"Intra-Atomic Magnetic Interactions for f
    Electrons.\" Physical Review 169, no. 1 (1968): 130.\"
1743  ";
1744  ReducedT22infn[numE_, SL_, SpLp_] := Module[
1745    {spin, orbital, t, idx1, idx2, S, L,
1746    Sp, Lp, cfpSL, cfpSpLp, parentSL,
1747    parentSpLp, Sb, Lb, Tnkk, phase, Sbp, Lbp},
1748    (
1749      {spin, orbital} = {1/2, 3};
1750      {S, L}   = FindSL[SL];
1751      {Sp, Lp} = FindSL[SpLp];
1752      t = 2;
1753      cfpSL    = CFP[{numE, SL}];
1754      cfpSpLp  = CFP[{numE, SpLp}];
1755      Tnkk = Sum[(
1756        parentSL   = cfpSL[[idx2, 1]];
1757        parentSpLp = cfpSpLp[[idx1, 1]];
1758        {Sb, Lb}   = FindSL[parentSL];
1759        {Sbp, Lbp} = FindSL[parentSpLp];
1760        phase = Phaser[Sb + Lb + spin + orbital + Sp + Lp];
1761        (
1762          phase *
1763          cfpSpLp[[idx1, 2]] * cfpSL[[idx2, 2]] *
1764          SixJay[{S, t, Sp}, {Sbp, spin, Sb}] *
1765          SixJay[{L, t, Lp}, {Lbp, orbital, Lb}] *
1766          T22Table[{numE - 1, parentSL, parentSpLp}]
1767        )
1768      ),
1769      {idx1, 2, Length[cfpSpLp]},
1770      {idx2, 2, Length[cfpSL]}
1771      ];
1772      Tnkk *= numE / (numE - 2) * Sqrt[TPO[S,Sp,L,Lp]];
1773      Return[Tnkk];
1774    )
1775  ];
1776
1777  GenerateT22Table::usage = "GenerateT22Table[nmax] generates the LS
    reduced matrix elements for the double tensor operator T22 in f^n
    up to n=nmax. If the option \"Export\" is set to true then the
    resulting association is saved to the data folder. The values for
    n=1 and n=2 are taken from \"Judd, BR, HM Crosswhite, and Hannah
    Crosswhite. \"Intra-Atomic Magnetic Interactions for f Electrons.\
    " Physical Review 169, no. 1 (1968): 130.\", and the values for n
    >2 are calculated recursively using equation (4) of that same
    paper.
1778  This is an intermediate step to the calculation of the reduced
    matrix elements of the spin-spin operator.";
1779  Options[GenerateT22Table] = {"Export" -> True, "Progress" -> True};
1780  GenerateT22Table[nmax_Integer, OptionsPattern[]] := (
1781    If[And[OptionValue["Progress"], frontEndAvailable],
1782      (
1783        numItersai = Association[Table[numE->Length[AllowedNKSLTerms[
    numE]]^2, {numE, 1, nmax}]];
1784        counters = Association[Table[numE->0, {numE, 1, nmax}]];
1785        totalIters = Total[Values[numItersai[[1;;nmax]]]];
1786        template1 = StringTemplate["Iteration `numiter` of `totaliter
    `"];
1787        template2 = StringTemplate["`remtime` min remaining"];
    template3 = StringTemplate["Iteration speed = `speed` ms/it"];
1788        template4 = StringTemplate["Time elapsed = `runtime` min"];
1789        progBar = PrintTemporary[
1790          Dynamic[
1791            Pane[
1792              Grid[{{Superscript["f", numE]},
1793                    {template1[<|"numiter"->numiter, "totaliter"->
    totalIters|>]},
1794                    {template4[<|"runtime"->Round[QuantityMagnitude[
    UnitConvert[(Now-startTime), "min"]], 0.1]|>]},
1795                    {template2[<|"remtime"->Round[QuantityMagnitude[
    UnitConvert[(Now-startTime)/(numiter)*(totalIters-numiter), "min"
    ]], 0.1]|>]},
1796                    {template3[<|"speed"->Round[QuantityMagnitude[Now
    -startTime, "ms"]/(numiter), 0.01]|>]},
1797                    {ProgressIndicator[Dynamic[numiter], {1,
```

```
    totalIters}]}},
1798                    Frame->All],
1799                    Full,
1800                    Alignment->Center]
1801                ]
1802              ];
1803      )
1804    ];
1805    T22Table = <||>;
1806    startTime = Now;
1807    numiter = 1;
1808    Do[
1809      (
1810        numiter+= 1;
1811        T22Table[{numE, SL, SpLp}] = Which[
1812          numE==1,
1813          0,
1814          numE==2,
1815          SimplifyFun[ReducedT22inf2[SL, SpLp]],
1816          True,
1817          SimplifyFun[ReducedT22infn[numE,  SL, SpLp]]
1818        ];
1819      ),
1820    {numE, 1, nmax},
1821    {SL,   AllowedNKSLTerms[numE]},
1822    {SpLp, AllowedNKSLTerms[numE]}
1823    ];
1824    If[And[OptionValue["Progress"],frontEndAvailable],
1825      NotebookDelete[progBar]
1826    ];
1827    If[OptionValue["Export"],
1828      (
1829        fname = FileNameJoin[{moduleDir, "data", "ReducedT22Table.m"
    }];
1830        Export[fname, T22Table];
1831      )
1832    ];
1833    Return[T22Table];
1834  );
1835
1836  SpinSpin::usage = "SpinSpin[n, SL, SpLp, J] returns the matrix
      element <|SL,J|spin-spin|SpLp,J|> for the spin-spin operator
      within the configuration f^n. This matrix element is independent
      of MJ. This is obtained by querying the relevant reduced matrix
      element from the association T22Table, putting in the adequate
      phase, and 6-j symbol.
1837  This is calculated according to equation (3) in \"Judd, BR, HM
      Crosswhite, and Hannah Crosswhite. \"Intra-Atomic Magnetic
      Interactions for f Electrons.\" Physical Review 169, no. 1 (1968):
       130.\"
1838  \".
1839  ";
1840  SpinSpin[numE_, SL_, SpLp_, J_] := Module[
1841    {S, L, Sp, Lp, α, val},
1842    (
1843      α = 2;
1844      {S, L} = FindSL[SL];
1845      {Sp, Lp} = FindSL[SpLp];
1846      val = (
1847               Phaser[Sp + L + J] *
1848               SixJay[{Sp, Lp, J}, {L, S, α}] *
1849               T22Table[{numE, SL, SpLp}]
1850             );
1851      Return[val]
1852    )
1853  ];
1854
1855  GenerateSpinSpinTable::usage = "GenerateSpinSpinTable[nmax]
      generates the reduced matrix elements in the |LSJ> basis for the (
      spin-other-orbit + electrostatically-correlated-spin-orbit)
      operator. It returns an association where the keys are of the form
       {numE, SL, SpLp, J}. If the option \"Export\" is set to True then
       the resulting object is saved to the data folder. Since this is a
       scalar operator, there is no MJ dependence. This dependence only
      comes into play when the crystal field contribution is taken into
      account.";
```

```
Options[GenerateSpinSpinTable] = {"Export"->False};
GenerateSpinSpinTable[nmax_, OptionsPattern[]] :=
  (
    SpinSpinTable = <||>;
    PrintTemporary[Dynamic[numE]];
    Do[
      SpinSpinTable[{numE, SL, SpLp, J}] = (SpinSpin[numE, SL, SpLp
  , J]);,
    {numE, 1, nmax},
    {J, MinJ[numE], MaxJ[numE]},
    {SL,   First /@ AllowedNKSLforJTerms[numE, J]},
    {SpLp, First /@ AllowedNKSLforJTerms[numE, J]}
    ];
    If[OptionValue["Export"],
    (fname = FileNameJoin[{moduleDir, "data", "SpinSpinTable.m"}];
      Export[fname, SpinSpinTable];
      )
    ];
    Return[SpinSpinTable];
  );

(* ###################### Spin-Spin ########################### *)
(* ############################################################ *)

(*
  ######################################################################
   *)
(* ## Spin-Other-Orbit and Electrostatically-Correlated-Spin-Orbit
  ## *)

SOOandECSO::usage = "SOOandECSO[n, SL, SpLp, J] returns the matrix
  element <|SL,J|spin-spin|SpLp,J|> for the combined effects of the
  spin-other-orbit interaction and the electrostatically-correlated-
  spin-orbit (which originates from configuration interaction
  effects) within the configuration f^n. This matrix element is
  independent of MJ. This is obtained by querying the relevant
  reduced matrix element by querying the association
  SOOandECSOLSTable and putting in the adequate phase and 6-j symbol
  . The SOOandECSOLSTable puts together the reduced matrix elements
  from three operators.
This is calculated according to equation (3) in \"Judd, BR, HM
  Crosswhite, and Hannah Crosswhite. \"Intra-Atomic Magnetic
  Interactions for f Electrons.\" Physical Review 169, no. 1 (1968):
   130.\".
";
SOOandECSO[numE_, SL_, SpLp_, J_] := Module[
  {S, Sp, L, Lp, α, val},
  (
    α = 1;
    {S, L}   = FindSL[SL];
    {Sp, Lp} = FindSL[SpLp];
    val = (
            Phaser[Sp + L + J] *
            SixJay[{Sp, Lp, J}, {L, S, α}] *
            SOOandECSOLSTable[{numE, SL, SpLp}]
          );
    Return[val];
  )
];

Prescaling = {P2 -> P2/225, P4 -> P4/1089, P6 -> 25 * P6 / 184041};

GenerateSOOandECSOTable::usage = "GenerateSOOandECSOTable[nmax]
  generates the reduced matrix elements in the |LSJ> basis for the (
  spin-other-orbit + electrostatically-correlated-spin-orbit)
  operator. It returns an association where the keys are of the form
   {n, SL, SpLp, J}. If the option \"Export\" is set to True then
  the resulting object is saved to the data folder. Since this is a
  scalar operator, there is no MJ dependence. This dependence only
  comes into play when the crystal field contribution is taken into
  account.";
Options[GenerateSOOandECSOTable] = {"Export"->False};
GenerateSOOandECSOTable[nmax_, OptionsPattern[]] := (
  SOOandECSOTable = <||>;
  Do[
    SOOandECSOTable[{numE, SL, SpLp, J}] = (SOOandECSO[numE, SL,
```

```
    SpLp, J] /. Prescaling);,
    {numE, 1, nmax},
    {J, MinJ[numE], MaxJ[numE]},
    {SL,   First /@ AllowedNKSLforJTerms[numE, J]},
    {SpLp, First /@ AllowedNKSLforJTerms[numE, J]}
    ];
    If[OptionValue["Export"],
    (
      fname = FileNameJoin[{moduleDir, "data", "SOOandECSOTable.m"}];
      Export[fname, SOOandECSOTable];
    )
    ];
    Return[SOOandECSOTable];
  );

  (* ## Spin-Other-Orbit and Electrostatically-Correlated-Spin-Orbit
    ## *)
  (*
    ######################################################################
    *)

  (* ############################################################ *)
  (* ################## Magnetic Interactions ################## *)

  MagneticInteractions::usage = "MagneticInteractions[{numE, SL, SLp,
     J}] returns the matrix element of the magnetic interaction
    between the terms SL and SLp in the f^numE configuration for the
    given value of J. The interaction is given by the sum of the spin-
    spin, the spin-other-orbit,  and the electrostatically-correlated-
    spin-orbit interactions.
  The part corresponding to the spin-spin interaction is provided by
    SpinSpin[{numE, SL, SLp, J}].
  The part corresponding to SOO and ECSO is provided by the function
    SOOandECSO[{numE, SL, SLp, J}].
  The option \"ChenDeltas\" can be used to include or exclude the
    Chen deltas from the calculation. The default is to exclude them.
    If this option is used, then the chenDeltas association needs to
    be loaded into the session with LoadChen[].";
  Options[MagneticInteractions] = {"ChenDeltas" -> False};
  MagneticInteractions[{numE_, SL_, SLp_, J_}, OptionsPattern[]] :=
    Module[
    {key, ss, sooandecso, total,
    S, L, Sp, Lp, phase, sixjay,
    M0v, M2v, M4v,
    P2v, P4v, P6v},
    (
      key        = {numE, SL, SLp, J};
      ss         = \[Sigma]SS * SpinSpinTable[key];
      sooandecso = SOOandECSOTable[key];
      total = ss + sooandecso;
      total = SimplifyFun[total];
      If[
        Not[OptionValue["ChenDeltas"]],
        Return[total]
      ];
      (* In the type A errors the wrong values are different *)
      If[MemberQ[Keys[chenDeltas["A"]], {numE, SL, SLp}],
        (
          {S, L}   = FindSL[SL];
          {Sp, Lp} = FindSL[SLp];
          phase    = Phaser[Sp + L + J];
          sixjay   = SixJay[{Sp, Lp, J}, {L, S, 1}];
          {M0v, M2v, M4v, P2v, P4v, P6v} = chenDeltas["A"][{numE, SL,
     SLp}]["wrong"];
          total    = (
                          phase * sixjay *
                          (
                            M0v*M0 + M2v*M2 + M4v*M4 +
                            P2v*P2 + P4v*P4 + P6v*P6
                          )
                     );
          total    = wChErrA * total + (1 - wChErrA) * (ss +
    sooandecso)
        )
      ];
      (* In the type B errors the wrong values are zeros all around
```

```
    *)
      If[MemberQ[chenDeltas["B"], {numE, SL, SLp}],
        (
          total  = (1 - wChErrB) * (ss + sooandecso)
        )
      ];
      Return[total];
    )
  ];

  (* ################## Magnetic Interactions ################## *)
  (* ############################################################ *)

  (* ############################################################ *)
  (* ##################### Free-Ion Energies ##################### *)

  GenerateFreeIonTable::usage="GenerateFreeIonTable[] generates an
    association for free-ion energies in terms of Slater integrals Fk
    and spin-orbit parameter ζ. It returns an association where the
    keys are of the form {nE, SL, SpLp}. If the option \"Export\" is
    set to True then the resulting object is saved to the data folder.
     The free-ion Hamiltonian is the sum of the electrostatic and spin
    -orbit interactions. The electrostatic interaction is given by the
     function Electrostatic[{numE, SL, SpLp}] and the spin-orbit
    interaction is given by the function SpinOrbitTable[{numE, SL,
    SpLp}]. The values for the electrostatic interaction are taken
    from the data file ElectrostaticTable.m and the values for the
    spin-orbit interaction are taken from the data file SpinOrbitTable
    .m. The values for the free-ion Hamiltonian are then exported to a
     file \"FreeIonTable.m\" in the data folder of this module. The
    values are also returned as an association.";
  Options[GenerateFreeIonTable] = {"Export" -> False};
  GenerateFreeIonTable[OptionsPattern[]] := Module[
    {terms, numEH, zetaSign, fname, FreeIonTable},
    (
      If[Not[ValueQ[ElectrostaticTable]],
        LoadElectrostatic[]
        ];
      If[Not[ValueQ[SpinOrbitTable]],
        LoadSpinOrbit[]
      ];
      If[Not[ValueQ[ReducedUkTable]],
        LoadUk[]
      ];
      FreeIonTable = <||>;
      Do[
        (
          terms = AllowedNKSLJTerms[nE];
          numEH = Min[nE, 14 - nE];
          zetaSign = If[nE > 7, -1, 1];
          Do[
            FreeIonTable[{nE,term[[1]],term[[2]]}] = (
              Electrostatic[{numEH, term[[1]], term[[1]]}] +
                zetaSign * SpinOrbitTable[{numEH, term[[1]], term
    [[1]], term[[2]]}]
                ),
            {term, terms}];
        ),
        {nE, 1, 14}
      ];
      If[OptionValue["Export"],
        (
          fname = FileNameJoin[{moduleDir, "data", "FreeIonTable.m"
    }];
          Export[fname, FreeIonTable];
        )
      ];
      Return[FreeIonTable];
    )
  ];

  LoadFreeIon::usage = "LoadFreeIon[] loads the free-ion energies
    from the data folder. The values are stored in the association
    FreeIonTable.";
  LoadFreeIon[] := (
    If[ValueQ[FreeIonTable],
```

```
      Return[]
    ];
    PrintTemporary["Loading the association of free-ion energies ..."
    ];
    FreeIonTableFname = FileNameJoin[{moduleDir, "data", "
    FreeIonTable.m"}];
    FreeIonTable = If[!FileExistsQ[FreeIonTableFname],
      (
        PrintTemporary[">> FreeIonTable.m not found, generating ..."
    ];
        GenerateFreeIonTable["Export" -> True]
      ),
        Import[FreeIonTableFname]
    ];
  );

  (* ##################### Free-Ion Energies ##################### *)
  (* ############################################################ *)

  (* ############################################################ *)
  (* ###################### Crystal Field ####################### *)

  Cqk::usage = "Cqk[numE, q, k, NKSL, J, M, NKSLp, Jp, Mp]. In
    Wybourne (1965) see equations 6-3, 6-4, and 6-5. Also in TASS see
    equation 11.53.";
  Cqk[numE_, q_, k_, NKSL_, J_, M_, NKSLp_, Jp_, Mp_] := Module[
    {S, Sp, L, Lp, orbital, val},
    (
      orbital = 3;
      {S, L}   = FindSL[NKSL];
      {Sp, Lp} = FindSL[NKSLp];
      f1  = ThreeJay[{J, -M}, {k, q}, {Jp, Mp}];
      val =
        If[f1==0,
          0,
          (
            f2 = SixJay[{L, J, S}, {Jp, Lp, k}] ;
            If[f2==0,
              0,
              (
                f3 = ReducedUkTable[{numE, orbital, NKSL, NKSLp, k}];
                If[f3==0,
                  0,
                  (
                    (
                      Phaser[J - M + S + Lp + J + k] *
                      Sqrt[TPO[J, Jp]] *
                      f1 *
                      f2 *
                      f3 *
                      Ck[orbital, k]
                    )
                  )
                ]
              )
            ]
          )
        ];
      Return[val];
    )
  ];

  Bqk::usage = "Real part of the Bqk coefficients.";
  Bqk[q_, 2] := {B02/2, B12, B22}[[q + 1]];
  Bqk[q_, 4] := {B04/2, B14, B24, B34, B44}[[q + 1]];
  Bqk[q_, 6] := {B06/2, B16, B26, B36, B46, B56, B66}[[q + 1]];

  Sqk::usage = "Imaginary part of the Bqk coefficients.";
  Sqk[q_, 2] := {0, S12, S22}[[q + 1]];
  Sqk[q_, 4] := {0, S14, S24, S34, S44}[[q + 1]];
  Sqk[q_, 6] := {0, S16, S26, S36, S46, S56, S66}[[q + 1]];

  CrystalField::usage = "CrystalField[n, NKSL, J, M, NKSLp, Jp, Mp]
    gives the general expression for the matrix element of the crystal
     field Hamiltonian parametrized with Bqk and Sqk coefficients as a
     sum over spherical harmonics Cqk.
```

```
Sometimes this expression only includes Bqk coefficients, see for example eqn 6-2 in Wybourne (1965), but one may also split the coefficient into real and imaginary parts as is done here, in an expression that is patently Hermitian.";
CrystalField[numE_, NKSL_, J_, M_, NKSLp_, Jp_, Mp_] := (
  Sum[
    (
      cqk  = Cqk[numE,  q, k, NKSL, J, M, NKSLp, Jp, Mp];
      cmqk = Cqk[numE, -q, k, NKSL, J, M, NKSLp, Jp, Mp];
      Bqk[q, k]   * (cqk + (-1)^q * cmqk) +
      I*Sqk[q, k] * (cqk - (-1)^q * cmqk)
      ),
  {k, {2, 4, 6}},
  {q, 0, k}
  ]
)

TotalCFIters::usage = "TotalIters[i, j] returns total number of function evaluations for calculating all the matrix elements for the \!\(\*SuperscriptBox[\(f\), \(i\)]\) to the \!\(\*SuperscriptBox[\(f\), \(j\)]\) configurations.";
TotalCFIters[i_, j_] := (
  numIters = {196, 8281, 132496, 1002001, 4008004, 9018009, 11778624};
  Return[Total[numIters[[i ;; j]]]];
  )

GenerateCrystalFieldTable::usage = "GenerateCrystalFieldTable[{numEs}] computes the matrix values for the crystal field interaction for f^n configurations the given list of numE in numEs. The function calculates the association CrystalFieldTable with keys of the form {numE, NKSL, J, M, NKSLp, Jp, Mp}. If the option \"Export\" is set to True, then the result is exported to the data subfolder for the folder in which this package is in. If the option \"Progress\" is set to True then an interactive progress indicator is shown. If \"Compress\" is set to true the exported values are compressed when exporting.";
Options[GenerateCrystalFieldTable] = {"Export" -> False, "Progress" -> True, "Compress" -> True};
GenerateCrystalFieldTable[numEs_List:{1,2,3,4,5,6,7}, OptionsPattern[]] := (
  ExportFun =
  If[OptionValue["Compress"],
    ExportMZip,
    Export
  ];
  numiter = 1;
  template1 = StringTemplate["Iteration `numiter` of `totaliter`"];
  template2 = StringTemplate["`remtime` min remaining"];
  template3 = StringTemplate["Iteration speed = `speed` ms/it"];
  template4 = StringTemplate["Time elapsed = `runtime` min"];
  totalIter = Total[TotalCFIters[#, #] & /@ numEs];
  freebies = 0;
  startTime = Now;
  If[And[OptionValue["Progress"], frontEndAvailable],
    progBar = PrintTemporary[
      Dynamic[
        Pane[
          Grid[
            {
              {Superscript["f", numE]},
              {template1[<|"numiter" -> numiter, "totaliter" -> totalIter|>]},
              {template4[<|"runtime" -> Round[QuantityMagnitude[UnitConvert[(Now - startTime), "min"]], 0.1]|>]},
              {template2[<|"remtime" -> Round[QuantityMagnitude[UnitConvert[(Now - startTime)/(numiter - freebies) * (totalIter - numiter), "min"]], 0.1]|>]},
              {template3[<|"speed" -> Round[QuantityMagnitude[Now - startTime, "ms"]/(numiter-freebies), 0.01]|>]},
              {ProgressIndicator[Dynamic[numiter], {1, totalIter}]}
            },
          Frame -> All
          ],
        Full,
        Alignment -> Center
```

```
          ]
        ]
      ];
    ];
    Do[
      (
        exportFname = FileNameJoin[{moduleDir, "data", "
    CrystalFieldTable_f"<>ToString[numE]<>".m"}];
        If[FileExistsQ[exportFname],
          Print["File exists, skipping ..."];
          numiter+=  TotalCFIters[numE, numE];
          freebies+= TotalCFIters[numE, numE];
          Continue[];
        ];
        CrystalFieldTable = <||>;
        Do[
          (
            numiter+= 1;
            CrystalFieldTable[{numE, NKSL, J, M, NKSLp, Jp, Mp}] =
    CrystalField[numE, NKSL, J, M, NKSLp, Jp, Mp];
          ),
        {J, MinJ[numE], MaxJ[numE]},
        {Jp, MinJ[numE], MaxJ[numE]},
        {M, AllowedMforJ[J]},
        {Mp, AllowedMforJ[Jp]},
        {NKSL , First /@ AllowedNKSLforJTerms[numE, J]},
        {NKSLp, First /@ AllowedNKSLforJTerms[numE, Jp]}
        ];
        If[And[OptionValue["Progress"],frontEndAvailable],
          NotebookDelete[progBar]
        ];
        If[OptionValue["Export"],
          (
            Print["Exporting to file "<>ToString[exportFname]];
            ExportFun[exportFname, CrystalFieldTable];
          )
        ];
      ),
    {numE, numEs}
    ]
  )

  Options[ParseBenelli2015] = {"Export"->False};
  ParseBenelli2015[OptionsPattern[]] := Module[
    {fname, crystalSym,
    crystalSymmetries, parseFun,
    chars, qk, groupName, family,
    groupNum, params},
  (
    fname      = FileNameJoin[{moduleDir,"data","
    benelli_and_gatteschi_table3p3.csv"}];
    crystalSym = Import[fname][[2;;33]];
    crystalSymmetries = <||>;
    parseFun[txt_] := (
      chars = Characters[txt];
      qk    = chars[[-2;;]];
      If[chars[[1]]=="R",
      (
        Return[{ToExpression@StringJoin[{"B",qk[[1]],qk[[2]]}]}]
      ),
      (
        If[qk[[1]]=="0",
          Return[{ToExpression@StringJoin[{"B",qk[[1]],qk[[2]]}]}]
        ];
        Return[{
          ToExpression@StringJoin[{"B",qk[[1]],qk[[2]]}],
          ToExpression@StringJoin[{"S",qk[[1]],qk[[2]]}]
        }]
      )]
    );
    Do[
    (
      groupNum  = Round@ToExpression@row[[1]];
      groupName = row[[2]];
      family    = row[[3]];
      params    = Select[row[[4;;]],#=!=""&];
```

```
      params    = parseFun/@params;
      params    = <|"BqkSqk"->Sort@Flatten[params],
        "aliases"->{groupNum},
        "constraints"->{}|>;
      If[MemberQ[{"T","Th","O","Td","Oh"},groupName],
        params["constraints"] = {
            {B44 ->  Sqrt[5/14] B04, B46 -> -Sqrt[7/2] B06},
            {B44 -> -Sqrt[5/14] B04, B46 ->  Sqrt[7/2] B06}
            }
      ];
      If[StringContainsQ[groupName,","],
      (
        {alias1, alias2}          = StringSplit[groupName,","];
        crystalSymmetries[alias1] = params;
        crystalSymmetries[alias1]["aliases"] = {groupNum, alias2};
        crystalSymmetries[alias2] = params;
        crystalSymmetries[alias2]["aliases"] = {groupNum, alias1};
      ),
      (
      crystalSymmetries[groupName] = params;
      )
      ]
    ),
    {row,crystalSym}];
    crystalSymmetries["source"] = "Benelli and Gatteschi, 2015,
    Introduction to Molecular Magnetism, table 3.3.";
    If[OptionValue["Export"],
      Export[FileNameJoin[{moduleDir,"data","
    crystalFieldFunctionalForms.m"}],crystalSymmetries];
    ];
    Return[crystalSymmetries];
  )
  ]

  CrystalFieldForm::usage = "CrystalFieldForm[symmetryGroup] returns
    an association that describes the crystal field parameters that
    are necessary to desribe a crystal field for the given symmetry
    group.

  The symmetry group must be given as a string in Schoenflies
    notation and must be one of C1, Ci, S2, Cs, C1h, C2, C2h, C2v, D2,
     D2h, S4, C4, C4h, D2d, C4v, D4, D4h, C3, S6, C3h, C3v, D3, D3d,
    D3h, C6, C6h, C6v, D6, D6h, T, Th, Td, O, Oh.

  The returned association has three keys:
    \"BqkSqk\" whose values is a list with the nonzero Bqk and Sqk
    parameters;
    \"constraints\" whose value is either an empty list, or a lists
    of replacements rules that are constraints on the Bqk and Sqk
    parameters;
    \"simplifier\" whose value is an association that can be used to
    set to zero the crystal field parameters that are zero for the
    given symmetry group;
    \"aliases\" whose value is a list with the integer by which the
    point group is also known for and an alternate Schoenflies symbol
    if it exists.

  This uses data from table 3.3 in Benelli and Gatteschi, 2015.";
  CrystalFieldForm[symmetryGroupString_] := (
    If[Not@ValueQ[crystalFieldFunctionalForms],
      crystalFieldFunctionalForms = Import[FileNameJoin[{moduleDir, "
    data", "crystalFieldFunctionalForms.m"}]];
    ];
    cfForm = crystalFieldFunctionalForms[symmetryGroupString];
    simplifier = Association[(# -> 0) &/@ Complement[cfSymbols,
    cfForm["BqkSqk"]]];
    Return[Join[cfForm, <|"simplifier"->simplifier|>]];
  )

  (* ###################### Crystal Field ###################### *)
  (* ########################################################## *)

  (* ########################################################## *)
  (* #### Configuration-Interaction via Casimir Operators ###### *)

  CasimirSO3::usage = "CasimirSO3[SL, SpLp] returns LS reduced matrix
```

```
    element of the configuration interaction term corresponding to
    the Casimir operator of R3.";
2277  CasimirSO3[{SL_, SpLp_}] := (
2278    {S, L} = FindSL[SL];
2279    If[SL == SpLp,
2280      α * L * (L + 1),
2281      0
2282    ]
2283  )
2284
2285  GG2U::usage = "GG2U is an association whose keys are labels for the
      irreducible representations of group G2 and whose values are the
      eigenvalues of the corresponding Casimir operator.
2286  Reference: Wybourne, \"Spectroscopic Properties of Rare Earths\",
      table 2-6.";
2287  GG2U = Association[{
2288      "00" -> 0,
2289      "10" -> 6/12 ,
2290      "11" -> 12/12,
2291      "20" -> 14/12,
2292      "21" -> 21/12,
2293      "22" -> 30/12,
2294      "30" -> 24/12,
2295      "31" -> 32/12,
2296      "40" -> 36/12}
2297    ];
2298
2299  CasimirG2::usage = "CasimirG2[SL, SpLp] returns LS reduced matrix
      element of the configuration interaction term corresponding to the
      Casimir operator of G2.";
2300  CasimirG2[{SL_, SpLp_}] := (
2301    Ulabel = FindNKLSTerm[SL][[1]][[4]];
2302    If[SL==SpLp,
2303      β * GG2U[Ulabel],
2304      0
2305    ]
2306  )
2307
2308  GSO7W::usage = "GSO7W is an association whose keys are labels for
      the irreducible representations of group R7 and whose values are
      the eigenvalues of the corresponding Casimir operator.
2309  Reference: Wybourne, \"Spectroscopic Properties of Rare Earths\",
      table 2-7.";
2310  GSO7W := Association[
2311    {
2312      "000" -> 0,
2313      "100" -> 3/5,
2314      "110" -> 5/5,
2315      "111" -> 6/5,
2316      "200" -> 7/5,
2317      "210" -> 9/5,
2318      "211" -> 10/5,
2319      "220" -> 12/5,
2320      "221" -> 13/5,
2321      "222" -> 15/5
2322    }
2323  ];
2324
2325  CasimirSO7::usage = "CasimirSO7[SL, SpLp] returns the LS reduced
      matrix element of the configuration interaction term corresponding
      to the Casimir operator of R7.";
2326  CasimirSO7[{SL_, SpLp_}] := (
2327    Wlabel = FindNKLSTerm[SL][[1]][[3]];
2328    If[SL==SpLp,
2329      γ * GSO7W[Wlabel],
2330      0
2331    ]
2332  )
2333
2334  ElectrostaticConfigInteraction::usage = "
      ElectrostaticConfigInteraction[{SL, SpLp}] returns the matrix
      element for configuration interaction as approximated by the
      Casimir operators of the groups R3, G2, and R7. SL and SpLp are
      strings that represent terms under LS coupling.";
2335  ElectrostaticConfigInteraction[{SL_, SpLp_}] := Module[
2336    {S, L, val},
```

```
    (
      {S, L} = FindSL[SL];
      val = (
        If[SL == SpLp,
          CasimirSO3[{SL, SL}] +
          CasimirSO7[{SL, SL}] +
          CasimirG2[{SL, SL}],
          0
        ]
        );
      ElectrostaticConfigInteraction[{S, L}] = val;
      Return[val];
    )
  ];

  (* #### Configuration-Interaction via Casimir Operators ###### *)
  (* ############################################################ *)

  (* ############################################################ *)
  (* ###################### Block assembly ###################### *)

  JJBlockMatrix::usage = "For given J, J' in the f^n configuration
    JJBlockMatrix[numE, J, J'] determines all the SL S'L' terms that
    may contribute to them and using those it provides the matrix
    elements <J, LS | H | J', LS'>. H having contributions from the
    following interactions: Coulomb, spin-orbit, spin-other-orbit,
    electrostatically-correlated-spin-orbit, spin-spin, three-body
    interactions, and crystal-field.";
  Options[JJBlockMatrix] = {"Sparse"->True, "ChenDeltas"->False};
  JJBlockMatrix[numE_, J_, Jp_, CFTable_, OptionsPattern[]] := Module
    [
    {NKSLJMs, NKSLJMps, NKSLJM, NKSLJMp,
    SLterm, SpLpterm,
    MJ, MJp,
    subKron, matValue, eMatrix},
    (
      NKSLJMs  = AllowedNKSLJMforJTerms[numE, J];
      NKSLJMps = AllowedNKSLJMforJTerms[numE, Jp];
      eMatrix =
        Table[
          (*Condition for a scalar matrix op*)
          SLterm   = NKSLJM[[1]];
          SpLpterm = NKSLJMp[[1]];
          MJ       = NKSLJM[[3]];
          MJp      = NKSLJMp[[3]];
          subKron  = (
              KroneckerDelta[J, Jp] *
              KroneckerDelta[MJ, MJp]
                    );
          matValue =
            If[subKron==0,
              0,
                (
                  ElectrostaticTable[{numE, SLterm, SpLpterm}] +
                  ElectrostaticConfigInteraction[{SLterm, SpLpterm}]
    +
                  SpinOrbitTable[{numE, SLterm, SpLpterm, J}] +
                  MagneticInteractions[{numE, SLterm, SpLpterm, J},
                    "ChenDeltas" -> OptionValue["ChenDeltas"]] +
                  ThreeBodyTable[{numE, SLterm, SpLpterm}]
                )
            ];
          matValue += CFTable[{numE, SLterm, J, MJ, SpLpterm, Jp, MJp
    }];
          matValue,
        {NKSLJMp, NKSLJMps},
        {NKSLJM , NKSLJMs}
        ];
     If[OptionValue["Sparse"],
       eMatrix = SparseArray[eMatrix]
     ];
     Return[eMatrix]
  )
  ];

  EnergyStates::usage = "Alias for AllowedNKSLJMforJTerms. At some
```

```
  point may be used to redefine states used in basis.";
EnergyStates[numE_, J_]:= AllowedNKSLJMforJTerms[numE, J];

JJBlockMatrixFileName::usage = "JJBlockMatrixFileName[numE] gives
  the filename for the energy matrix table for an atom with numE f-
  electrons. The function admits an optional parameter \"
  FilenameAppendix\" which can be used to modify the filename.";
Options[JJBlockMatrixFileName] = {"FilenameAppendix" -> ""};
JJBlockMatrixFileName[numE_Integer, OptionsPattern[]] := (
  fileApp = OptionValue["FilenameAppendix"];
  fname = FileNameJoin[{moduleDir,
      "hams",
      StringJoin[{"f", ToString[numE], "_JJBlockMatrixTable",
  fileApp ,".m"}]}];
  Return[fname];
);

TabulateJJBlockMatrixTable::usage = "TabulateJJBlockMatrixTable[
  numE, I] returns a list with three elements {JJBlockMatrixTable,
  EnergyStatesTable, AllowedM}. JJBlockMatrixTable is an association
   with keys equal to lists of the form {numE, J, Jp}.
  EnergyStatesTable is an association with keys equal to lists of
  the form {numE, J}. AllowedM is another association with keys
  equal to lists of the form {numE, J} and values equal to lists
  equal to the corresponding values of MJ. It's unnecessary (and it
  won't work in this implementation) to give numE > 7 given the
  equivalency between electron and hole configurations.";
Options[TabulateJJBlockMatrixTable] = {"Sparse"->True, "ChenDeltas"
  ->False};
TabulateJJBlockMatrixTable[numE_, CFTable_, OptionsPattern[]] := (
  JJBlockMatrixTable = <||>;
  totalIterations = Length[AllowedJ[numE]]^2;
  template1 = StringTemplate["Iteration `numiter` of `totaliter`"];
  template2 = StringTemplate["`remtime` min remaining"];
  template4 = StringTemplate["Time elapsed = `runtime` min"];
  numiter   = 0;
  startTime = Now;
  If[$FrontEnd =!= Null,
    (
      temp = PrintTemporary[
        Dynamic[
          Grid[
            {
              {template1[<|"numiter"->numiter, "totaliter"->
  totalIterations|>]},
              {template2[<|"remtime"->Round[QuantityMagnitude[
  UnitConvert[(Now-startTime)/(Max[1,numiter])*(totalIterations-
  numiter), "min"]], 0.1]|>]},
              {template4[<|"runtime"->Round[QuantityMagnitude[
  UnitConvert[(Now-startTime), "min"]], 0.1]|>]},
              {ProgressIndicator[numiter, {1, totalIterations}]}
            }
          ]
      ]
      ];
    )
  ];
  Do[
    (
      JJBlockMatrixTable[{numE, J, Jp}] = JJBlockMatrix[numE, J, Jp
  , CFTable, "Sparse"->OptionValue["Sparse"], "ChenDeltas" ->
  OptionValue["ChenDeltas"]];
      numiter += 1;
    ),
  {Jp, AllowedJ[numE]},
  {J, AllowedJ[numE]}
  ];
  If[$FrontEnd =!= Null,
    NotebookDelete[temp]
  ];
  Return[JJBlockMatrixTable];
);

TabulateManyJJBlockMatrixTables::usage = "
  TabulateManyJJBlockMatrixTables[{n1, n2, ...}] calculates the
  tables of matrix elements for the requested f^n_i configurations.
```

```
    The function does not return the matrices themselves. It instead
    returns an association whose keys are numE and whose values are
    the filenames where the output of TabulateJJBlockMatrixTables was
    saved to.The output consists of an association whose keys are of
    the form {n, J, Jp} and whose values are rectangular arrays given
    the values of <|LSJMJa|H|L'S'J'MJ'a'|>.";
Options[TabulateManyJJBlockMatrixTables] = {"Overwrite"->False, "
    Sparse"->True, "ChenDeltas"->False, "FilenameAppendix"-> "", "
    Compressed" -> False};
TabulateManyJJBlockMatrixTables[ns_, OptionsPattern[]] := (
    overwrite = OptionValue["Overwrite"];
    fNames = <||>;
    fileApp = OptionValue["FilenameAppendix"];
    ExportFun = If[OptionValue["Compressed"], ExportMZip, Export];
    Do[
      (
        CFdataFilename = FileNameJoin[{moduleDir, "data", "
    CrystalFieldTable_f"<>ToString[numE]<>".zip"}];
        PrintTemporary["Importing CrystalFieldTable from ",
    CFdataFilename, " ..."];
        CrystalFieldTable = ImportMZip[CFdataFilename];

        PrintTemporary["#------- numE = ", numE, " -------#"];
        exportFname = JJBlockMatrixFileName[numE, "FilenameAppendix"
    -> fileApp];
        fNames[numE] = exportFname;
        If[FileExistsQ[exportFname] && Not[overwrite],
          Continue[]
        ];
        JJBlockMatrixTable = TabulateJJBlockMatrixTable[numE,
    CrystalFieldTable, "Sparse"->OptionValue["Sparse"], "ChenDeltas"
    -> OptionValue["ChenDeltas"]];
        If[FileExistsQ[exportFname]&&overwrite,
          DeleteFile[exportFname]
        ];
        ExportFun[exportFname, JJBlockMatrixTable];

        ClearAll[CrystalFieldTable];
      ),
    {numE, ns}
    ];
    Return[fNames];
);

HamMatrixAssembly::usage = "HamMatrixAssembly[numE] returns the
    Hamiltonian matrix for the f^n_i configuration. The matrix is
    returned as a SparseArray.
The function admits an optional parameter \"FilenameAppendix\"
    which can be used to modify the filename to which the resulting
    array is exported to.
It also admits an optional parameter \"IncludeZeeman\" which can be
    used to include the Zeeman interaction. The default is False.
The option \"Set t2Switch\" can be used to toggle on or off setting
    the t2 selector automatically or not, the default is True, which
    replaces the parameter according to numE.
The option \"ReturnInBlocks\" can be used to return the matrix in
    block or flattened form. The default is to return it in flattened
    form.";
Options[HamMatrixAssembly] = {
      "FilenameAppendix" -> "",
      "IncludeZeeman"    -> False,
      "Set t2Switch"     -> True,
      "ReturnInBlocks"   -> False,
      "OperatorBasis"    -> "Legacy"};
HamMatrixAssembly[nf_, OptionsPattern[]] := Module[
    {numE, ii, jj, howManyJs, Js, blockHam, opBasis},
    (
      (*####################################*)
      ImportFun = ImportMZip;
      opBasis = OptionValue["OperatorBasis"];
      If[Not[MemberQ[{"Legacy", "MostlyOrthogonal", "Orthogonal"},
    opBasis]],
        Print["Operator basis ", opBasis, " not recognized, using \"
    Legacy\" basis."];
        opBasis = "Legacy";
      ];
```

```
2509      If[opBasis == "Orthogonal",
2510        Print["Operator basis \"Orthogonal\" not implemented yet,
      aborting ..."];
2511        Return[Null];
2512      ];
2513      (*####################################*)
2514      If[opBasis == "MostlyOrthogonal",
2515        (
2516          blockHam = HamMatrixAssembly[nf,
2517          "OperatorBasis" -> "Legacy",
2518          "FilenameAppendix" -> OptionValue["FilenameAppendix"],
2519          "IncludeZeeman" -> OptionValue["IncludeZeeman"],
2520          "Set t2Switch" -> OptionValue["Set t2Switch"],
2521          "ReturnInBlocks" -> OptionValue["ReturnInBlocks"]];
2522          paramChanger = Which[
2523            nf < 7,
2524            <|
2525              F0 -> 1/91 (54 E1p+91 E0p+78 γp),
2526              F2 -> (15/392 *
2527                (
2528                  140   E1p  +
2529                  20020 E2p +
2530                  1540  E3p  +
2531                  770 αp -
2532                  70 γp  +
2533                  22 Sqrt[2] T2p -
2534                  11 Sqrt[2] nf T2p t2Switch -
2535                  11 Sqrt[2] (14-nf) T2p (1 - t2Switch)
2536                )
2537              ),
2538              F4 -> (99/490 *
2539                (
2540                  70   E1p -
2541                  9100 E2p +
2542                  280  E3p +
2543                  140 αp -
2544                  35 γp  +
2545                  4 Sqrt[2] T2p -
2546                  2 Sqrt[2] nf T2p t2Switch -
2547                  2 Sqrt[2] (14-nf) T2p (1-t2Switch)
2548                )
2549                ),
2550              F6 -> (5577/7000 *
2551                (
2552                  20  E1p +
2553                  700 E2p -
2554                  140 E3p -
2555                  70 αp -
2556                  10 γp -
2557                  2 Sqrt[2] T2p +
2558                  Sqrt[2] nf T2p t2Switch +
2559                  Sqrt[2] (14-nf) T2p (1-t2Switch)
2560                )
2561                ),
2562              ζ  -> ζ,
2563              α -> (5 αp)/4,
2564              β  -> -6  (5 αp + βp),
2565              γ -> 5/2 (2 βp + 5 γp),
2566              T2 -> 0
2567            |>,
2568          nf >= 7,
2569            <|
2570              F0 -> 1/91 (54 E1p+91 E0p+78 γp),
2571              F2 -> (15/392 *
2572                (
2573                  140   E1p  +
2574                  20020 E2p +
2575                  1540  E3p  +
2576                  770 αp -
2577                  70 γp  +
2578                  22 Sqrt[2] T2p -
2579                  11 Sqrt[2] nf T2p
2580                )
2581              ),
2582              F4 -> (99/490 *
2583                (
```

```
                  70   E1p -
                  9100 E2p +
                  280  E3p +
                  140 αp -
                  35 γp  +
                  4 Sqrt[2] T2p -
                  2 Sqrt[2] nf T2p
                )
                ),
              F6 -> (5577/7000 *
                (
                  20  E1p +
                  700 E2p -
                  140 E3p -
                  70 αp -
                  10 γp -
                  2 Sqrt[2] T2p +
                  Sqrt[2] nf T2p
                )
                ),
              ζ  -> ζ,
              α -> (5 αp)/4,
              β  -> -6  (5 αp + βp),
              γ -> 5/2 (2 βp + 5 γp),
              T2 -> 0
            |>
        ];
        blockHamM0 = Which[
          OptionValue["ReturnInBlocks"] == False,
          ReplaceInSparseArray[blockHam, paramChanger],
          OptionValue["ReturnInBlocks"] == True,
          Map[ReplaceInSparseArray[#, paramChanger]&, blockHam,
{2}]
        ];
        Return[blockHamM0];
      )
    ];
    (*######################################*)

    (*hole-particle equivalence enforcement*)
    numE = nf;
    allVars = {E0, E1, E2, E3, ζ, F0, F2, F4, F6, M0, M2, M4, T2,
T2p,
      T3, T4, T6, T7, T8, P0, P2, P4, P6, gs,
      α, β, γ, B02, B04, B06, B12, B14, B16,
      B22, B24, B26, B34, B36, B44, B46, B56, B66, S12, S14, S16,
S22,
      S24, S26, S34, S36, S44, S46, S56, S66, T11p, T12, T14, T15,
T16,
      T17, T18, T19, Bx, By, Bz};
    params0 = AssociationThread[allVars, allVars];
    If[nf > 7,
      (
        numE = 14 - nf;
        params = HoleElectronConjugation[params0];
        If[OptionValue["Set t2Switch"], params[t2Switch] = 0];
      ),
      params = params0;
      If[OptionValue["Set t2Switch"], params[t2Switch] = 1];
    ];
    (* Load symbolic expressions for LS,J,J' energy sub-matrices.
*)
    emFname = JJBlockMatrixFileName[numE, "FilenameAppendix" ->
OptionValue["FilenameAppendix"]];
    JJBlockMatrixTable = ImportFun[emFname];
    (*Patch together the entire matrix representation using J,J'
blocks.*)
    PrintTemporary["Patching JJ blocks ..."];
    Js        = AllowedJ[numE];
    howManyJs = Length[Js];
    blockHam  = ConstantArray[0, {howManyJs, howManyJs}];
    Do[
      blockHam[[jj, ii]] = JJBlockMatrixTable[{numE, Js[[ii]], Js[[
jj]]}];,
    {ii, 1, howManyJs},
    {jj, 1, howManyJs}
```

```
2652        ];
2653
2654        (* Once the block form is created flatten it *)
2655        If[Not[OptionValue["ReturnInBlocks"]],
2656          (blockHam = ArrayFlatten[blockHam];
2657          blockHam  = ReplaceInSparseArray[blockHam, params];
2658          ),
2659          (blockHam  = Map[ReplaceInSparseArray[#, params]&,blockHam
    ,{2}];)
2660        ];
2661
2662        If[OptionValue["IncludeZeeman"],
2663          (
2664            PrintTemporary["Including Zeeman terms ..."];
2665            {magx, magy, magz} = MagDipoleMatrixAssembly[numE, "
    ReturnInBlocks" -> OptionValue["ReturnInBlocks"]];
2666            blockHam += - teslaToKayser * (Bx * magx + By * magy + Bz *
     magz);
2667          )
2668        ];
2669        Return[blockHam];
2670      )
2671    ];
2672
2673    SimplerSymbolicHamMatrix::usage = "SimplerSymbolicHamMatrix[numE,
      simplifier] is a simple addition to HamMatrixAssembly that applies
       a given simplification to the full Hamiltonian. simplifier is a
      list of replacement rules.
2674    If the option \"Export\" is set to True, then the function also
      exports the resulting sparse array to the ./hams/ folder.
2675    The option \"PrependToFilename\" can be used to append a string to
      the filename to which the function may export to.
2676    The option \"Return\" can be used to choose whether the function
      returns the matrix or not.
2677    The option \"Overwrite\" can be used to overwrite the file if it
      already exists, if this options is set to False then this function
       simply reloads a file that it assumed to be present already in
      the ./hams folder.
2678    The option \"IncludeZeeman\" can be used to toggle the inclusion of
       the Zeeman interaction with an external magnetic field.
2679    The option \"OperatorBasis\" can be used to choose the basis in
      which the operator is expressed. The default is the \"Legacy\"
      basis. Order alternatives being: \"MostlyOrthogonal\" and \"
      Orthogonal\". In the \"Legacy\" alternative the operators used are
       the same as in Carnall's work. In the \"MostlyOrthogonal\"  all
      operators are orthogonal except those corresponding to the Mk and
      Pk parameters. In the \"Orthogonal\" basis all operators are
      orthogonal, with the operators corresponding to the Mk and Pk
      parameters replaced by zi operators and accompanying ai
      coefficients. The \"Orthogonal\" option has not been implemented
      yet.";
2680    Options[SimplerSymbolicHamMatrix] = {
2681      "Export"->True,
2682      "PrependToFilename"->"",
2683      "EorF"->"F",
2684      "Overwrite" -> False,
2685      "Return" -> True,
2686      "Set t2Switch" -> False,
2687      "IncludeZeeman" -> False,
2688      "OperatorBasis" -> "Legacy"
2689      };
2690    SimplerSymbolicHamMatrix[numE_Integer, simplifier_List,
      OptionsPattern[]] := Module[
2691      {thisHam,fname, fnamemx},
2692      (
2693        If[Not[ValueQ[ElectrostaticTable]],
2694          LoadElectrostatic[]
2695          ];
2696        If[Not[ValueQ[SOOandECSOTable]],
2697          LoadSOOandECSO[]
2698        ];
2699        If[Not[ValueQ[SpinOrbitTable]],
2700          LoadSpinOrbit[]
2701        ];
2702        If[Not[ValueQ[SpinSpinTable]],
2703          LoadSpinSpin[]
```

```
2704      ];
2705      If[Not[ValueQ[ThreeBodyTable]],
2706        LoadThreeBody[]
2707      ];
2708
2709      opBasis = OptionValue["OperatorBasis"];
2710      If[Not[MemberQ[{"Legacy", "MostlyOrthogonal", "Orthogonal"},
      opBasis]],
2711        Print["Operator basis ", opBasis, " not recognized, using \"
      Legacy\" basis."];
2712        opBasis = "Legacy";
2713      ];
2714      If[opBasis == "Orthogonal",
2715        Print["Operator basis \"Orthogonal\" not implemented yet,
      aborting ..."];
2716        Return[Null];
2717      ];
2718
2719      fnamePrefix = Which[
2720        opBasis == "Legacy",
2721        "SymbolicMatrix-f",
2722        opBasis == "MostlyOrthogonal",
2723        "SymbolicMatrix-mostly-orthogonal-f",
2724        opBasis == "Orthogonal",
2725        "SymbolicMatrix-orthogonal-f"
2726      ];
2727      fname   = FileNameJoin[{moduleDir, "hams",
2728        OptionValue["PrependToFilename"] <> fnamePrefix <> ToString[
      numE] <> ".m"}];
2729      fnamemx = FileNameJoin[{moduleDir,"hams",
2730        OptionValue["PrependToFilename"] <> fnamePrefix <> ToString[
      numE] <> ".mx"}];
2731      If[Or[FileExistsQ[fname], FileExistsQ[fnamemx]]
2732        && Not[OptionValue["Overwrite"]],
2733        (
2734          If[OptionValue["Return"],
2735            (
2736              Which[
2737                FileExistsQ[fnamemx],
2738                (
2739                  Print["File ", fnamemx, " already exists, and
      option \"Overwrite\" is set to False, loading file ..."];
2740                  thisHam = Import[fnamemx];
2741                  Return[thisHam];
2742                ),
2743                FileExistsQ[fname],
2744                (
2745                  Print["File ", fname, " already exists, and option
      \"Overwrite\" is set to False, loading file ..."];
2746                  thisHam = Import[fname];
2747                  Print["Exporting to file ", fnamemx, " for quicker
      loading."];
2748                  Export[fnamemx, thisHam];
2749                  Return[thisHam];
2750                )
2751              ]
2752            ),
2753            (
2754              Print["File ", fname, " already exists, skipping ..."];
2755              Return[Null];
2756            )
2757          ]
2758        )
2759      ];
2760
2761      thisHam = HamMatrixAssembly[numE, "Set t2Switch" -> OptionValue
      ["Set t2Switch"], "IncludeZeeman" -> OptionValue["IncludeZeeman"],
       "OperatorBasis" -> opBasis];
2762      If[Length[simplifier] > 0,
2763        thisHam = ReplaceInSparseArray[thisHam, simplifier];
2764      ];
2765      (* This removes zero entries from being included in the sparse
      array *)
2766      thisHam = SparseArray[thisHam];
2767      If[OptionValue["Export"],
2768      (
```

```
2769          Print["Exporting to file ", fname, " and to ", fnamemx];
2770          Export[fname,   thisHam];
2771          Export[fnamemx, thisHam];
2772        )
2773        ];
2774        If[OptionValue["Return"],
2775          Return[thisHam],
2776          Return[Null]
2777        ];
2778      )
2779    ];
2780
2781    ScalarLSJMFromLS::usage = "ScalarLSJMFromLS[numE,
        LSReducedMatrixElements]. Given the LS-reduced matrix elements
        LSReducedMatrixElements of a scalar operator, this function
        returns the corresponding LSJM representation. This is returned as
         a SparseArray.";
2782    ScalarLSJMFromLS[numE_, LSReducedMatrixElements_] := Module[
2783      {jjBlocktable, NKSLJMs, NKSLJMps, J, Jp, eMatrix, SLterm,
        SpLpterm,
2784      MJ, MJp, subKron, matValue, Js, howManyJs, blockHam, ii, jj},
2785      (
2786      SparseDiagonalArray[diagonalElements_] := SparseArray[
2787        Table[{i, i} -> diagonalElements[[i]],
2788          {i, 1, Length[diagonalElements]}]
2789        ];
2790      SparseZeroArray[width_, height_] := (
2791        SparseArray[
2792          Join[
2793          Table[{1, i} -> 0, {i, 1, width}],
2794          Table[{i, 1} -> 0, {i, 1, height}]
2795          ]
2796          ]
2797        );
2798      jjBlockTable = <||>;
2799      Do[
2800        NKSLJMs  = AllowedNKSLJMforJTerms[numE, J];
2801        NKSLJMps = AllowedNKSLJMforJTerms[numE, Jp];
2802        If[J != Jp,
2803          jjBlockTable[{numE, J, Jp}] = SparseZeroArray[Length[NKSLJMs
        ], Length[NKSLJMps]];
2804        Continue[];
2805        ];
2806        eMatrix = Table[
2807          (* Condition for a scalar matrix op *)
2808          SLterm = NKSLJM[[1]];
2809          SpLpterm = NKSLJMp[[1]];
2810          MJ = NKSLJM[[3]];
2811          MJp = NKSLJMp[[3]];
2812          subKron = (KroneckerDelta[MJ, MJp]);
2813          matValue = If[subKron == 0,
2814          0,
2815            (
2816              Which[MemberQ[Keys[LSReducedMatrixElements], {numE,
        SLterm, SpLpterm}],
2817                LSReducedMatrixElements[{numE, SLterm, SpLpterm}],
2818                MemberQ[Keys[LSReducedMatrixElements], {numE, SpLpterm,
         SLterm}],
2819                LSReducedMatrixElements[{numE, SpLpterm, SLterm}],
2820                True,
2821                0
2822              ]
2823            )
2824          ];
2825          matValue,
2826          {NKSLJMp, NKSLJMps},
2827          {NKSLJM, NKSLJMs}
2828          ];
2829        jjBlockTable[{numE, J, Jp}] = SparseArray[eMatrix],
2830        {J, AllowedJ[numE]},
2831        {Jp, AllowedJ[numE]}
2832        ];
2833
2834      Js = AllowedJ[numE];
2835      howManyJs = Length[Js];
2836      blockHam = ConstantArray[0, {howManyJs, howManyJs}];
```

```
    Do[blockHam[[jj, ii]] = jjBlockTable[{numE, Js[[ii]], Js[[jj]]}];,
      {ii, 1, howManyJs},
      {jj, 1, howManyJs}];
    blockHam = ArrayFlatten[blockHam];
    blockHam = SparseArray[blockHam];
    Return[blockHam];
    )
    ];

(* ###################### Block assembly ###################### *)
(* ############################################################ *)

(* ############################################################ *)
(* ##################### Level Description ##################### *)

FreeHam::usage = "FreeHam[JJBlocks, numE] given the JJ blocks of the Hamiltonian for f^n, this function returns a list with all the scalar-simplified versions of the blocks.";
FreeHam[JJBlocks_List, numE_Integer] := Module[
  {Js, basisJ, pivot, freeHam, idx, J,
  thisJbasis, shrunkBasisPositions, theBlock},
  (
    Js       = AllowedJ[numE];
    basisJ   = BasisLSJMJ[numE, "AsAssociation" -> True];
    pivot    = If[OddQ[numE], 1/2, 0];
    freeHam = Table[(
      J           = Js[[idx]];
      theBlock    = JJBlocks[[idx]];
      thisJbasis = basisJ[J];
      (* find the basis vectors that end with pivot *)
      shrunkBasisPositions = Flatten[Position[thisJbasis, {_ ..., pivot}]];
      (* take only those rows and columns *)
      theBlock[[shrunkBasisPositions, shrunkBasisPositions]]
    ),
    {idx, 1, Length[Js]}
    ];
    Return[freeHam];
  )
];

ListRepeater::usage = "ListRepeater[list, reps] repeats each element of list reps times.";
ListRepeater[list_List, repeats_Integer] := (
  Flatten[ConstantArray[#, repeats] & /@ list]
);

ListLever::usage = "ListLever[vecs, multiplicity] takes a list of vectors and returns all interleaved shifted versions of them.";
ListLever[vecs_, multiplicity_] := Module[
{uppityVecs, uppityVec},
(
  uppityVecs = Table[(
    uppityVec = PadRight[{#}, multiplicity] & /@ vec;
    uppityVec = Permutations /@ uppityVec;
    uppityVec = Transpose[uppityVec];
    uppityVec = Flatten /@ uppityVec
    ),
  {vec, vecs}
  ];
  Return[Flatten[uppityVecs, 1]];
)
];

EigenLever::usage = "EigenLever[eigenSys, multiplicity] takes a list eigenSys of the form {eigenvalues, eigenvectors} and returns the eigenvalues repeated multiplicity times and the eigenvectors interleaved and shifted accordingly.";
EigenLever[eigenSys_, multiplicity_] := Module[
  {eigenVals, eigenVecs,
  leveledEigenVecs, leveledEigenVals},
  (
    {eigenVals, eigenVecs} = eigenSys;
    leveledEigenVals       = ListRepeater[eigenVals, multiplicity];
    leveledEigenVecs       = ListLever[eigenVecs, multiplicity];
```

```
      Return[{Flatten[leveledEigenVals], leveledEigenVecs}]
    )
];


LevelSimplerSymbolicHamMatrix::usage = "
  LevelSimplerSymbolicHamMatrix[numE] is a variation of
  HamMatrixAssembly that returns the diagonal JJ Hamiltonian blocks
  applying a simplifier and with simplifications adequate for the
  level description. The keys of the given association correspond to
   the different values of J that are possible for f^numE, the
  values are sparse array that are meant to be interpreted in the
  basis provided by BasisLSJ.
The option \"Simplifier\" is a list of symbols that are set to zero
  . At a minimum this has to include the crystal field parameters.
  By default this includes everything except the Slater parameters
  Fk and the spin orbit coupling ζ.
The option \"Export\" controls whether the resulting association is
   saved to disk, the default is True and the resulting file is
  saved to the ./hams/ folder. A hash is appended to the filename
  that corresponds to the simplifier used in the resulting
  expression. If the option \"Overwrite\" is set to False then these
   files may be used to quickly retrieve a previously computed case.
   The file is saved both in .m and .mx format.
The option \"PrependToFilename\" can be used to append a string to
  the filename to which the function may export to.
The option \"Return\" can be used to choose whether the function
  returns the matrix or not.
The option \"Overwrite\" can be used to overwrite the file if it
  already exists.";
Options[LevelSimplerSymbolicHamMatrix] = {
  "Export" -> True,
  "PrependToFilename" -> "",
  "Overwrite" -> False,
  "Return" -> True,
  "Simplifier" -> Join[
    {F0,\[Sigma]SS},
    cfSymbols,
    TSymbols,
    casimirSymbols,
    pseudoMagneticSymbols,
    marvinSymbols,
    DeleteCases[magneticSymbols,ζ]
  ]
};
LevelSimplerSymbolicHamMatrix[numE_Integer, OptionsPattern[]] :=
  Module[
  {thisHamAssoc, Js, fname,
  fnamemx, hash, simplifier},
  (
    simplifier = (#->0)&/@Sort[OptionValue["Simplifier"]];
    hash       = Hash[simplifier];
    If[Not[ValueQ[ElectrostaticTable]],LoadElectrostatic[]];
    If[Not[ValueQ[SOOandECSOTable]],LoadSOOandECSO[]];
    If[Not[ValueQ[SpinOrbitTable]],LoadSpinOrbit[]];
    If[Not[ValueQ[SpinSpinTable]],LoadSpinSpin[]];
    If[Not[ValueQ[ThreeBodyTable]],LoadThreeBody[]];
    fname   = FileNameJoin[{moduleDir,"hams",OptionValue["
  PrependToFilename"]<>"Level-SymbolicMatrix-f"<>ToString[numE]<>"-"
  <>ToString[hash]<>".m"}];
    fnamemx = FileNameJoin[{moduleDir,"hams",OptionValue["
  PrependToFilename"]<>"Level-SymbolicMatrix-f"<>ToString[numE]<>"-"
  <>ToString[hash]<>".mx"}];
    If[Or[FileExistsQ[fname],FileExistsQ[fnamemx]]&&Not[OptionValue
  ["Overwrite"]],
    (
      If[OptionValue["Return"],
      (
        Which[FileExistsQ[fnamemx],
        (
          Print["File ",fnamemx," already exists, and option \"
  Overwrite\" is set to False, loading file ..."];
          thisHamAssoc=Import[fnamemx];
          Return[thisHamAssoc];
        ),
        FileExistsQ[fname],
```

```
2954            (
2955              Print["File ",fname," already exists, and option \"
      Overwrite\" is set to False, loading file ..."];
2956              thisHamAssoc=Import[fname];
2957              Print["Exporting to file ",fnamemx," for quicker loading.
      "];
2958              Export[fnamemx,thisHamAssoc];
2959              Return[thisHamAssoc];
2960            )
2961            ]
2962          ),
2963          (
2964            Print["File ",fname," already exists, skipping ..."];
2965            Return[Null];
2966          )
2967          ]
2968        )
2969        ];
2970        Js           = AllowedJ[numE];
2971        thisHamAssoc = HamMatrixAssembly[numE,
2972          "Set t2Switch"->True,
2973          "IncludeZeeman"->False,
2974          "ReturnInBlocks"->True
2975        ];
2976        thisHamAssoc = Diagonal[thisHamAssoc];
2977        thisHamAssoc = Map[SparseArray[ReplaceInSparseArray[#,
      simplifier]]&,thisHamAssoc,{1}];
2978        thisHamAssoc = FreeHam[thisHamAssoc,numE];
2979        thisHamAssoc = AssociationThread[Js->thisHamAssoc];
2980        If[OptionValue["Export"],
2981          (
2982            Print["Exporting to file ",fname," and to ",fnamemx];
2983            Export[fname,thisHamAssoc];
2984            Export[fnamemx,thisHamAssoc];
2985          )
2986        ];
2987        If[OptionValue["Return"],
2988          Return[thisHamAssoc],
2989          Return[Null]
2990        ];
2991      )
2992    ];
2993
2994    LevelSolver::usage = "LevelSolver[numE, params] puts together (or
        retrieves from disk) the symbolic level Hamiltonian for the f^numE
         configuration and solves it for the given params returning the
        resultant energies and eigenstates.
2995    If the option \"Return as states\" is set to False, then the
        function returns an association whose keys are values for J in f^
        numE, and whose values are lists with two elements. The first
        element being equal to the ordered basis for the corresponding
        subpsace, given as a list of lists of the form {LS string, J}. The
         second element being another list of two elements, the first
        element being equal to the energies and the second being equal to
        the corresponding normalized eigenvectors. The energies given have
         been subtracted the energy of the ground state.
2996    If the option \"Return as states\" is set to True, then the
        function returns a list with three elements. The first element is
        the global level basis for the f^numE configuration, given as a
        list of lists of the form {LS string, J}. The second element are
        the mayor LSJ components in the returned eigenstates. The third
        element is a list of lists with three elements, in each list the
        first element being equal to the energy, the second being equal to
         the value of J, and the third being equal to the corresponding
        normalized eigenvector (given as a row). The energies given have
        been subtracted the energy of the ground state, and the states
        have been sorted in order of increasing energy.
2997    The following options are admitted:
2998      - \"Overwrite Hamiltonian\", if set to True the function will
        overwrite the symbolic Hamiltonian. Default is False.
2999      - \"Return as states\", see description above. Default is True.
3000      - \"Simplifier\", this is a list with symbols that are set to
        zero for defining the parameters kept in the level  description.
3001    ";
3002    Options[LevelSolver] = {
3003      "Overwrite Hamiltonian" -> False,
```

```
    "Return as states" -> True,
    "Simplifier" -> Join[
      cfSymbols,
      TSymbols,
      casimirSymbols,
      pseudoMagneticSymbols,
      marvinSymbols,
      DeleteCases[magneticSymbols,ζ]
    ],
    "PrintFun" -> PrintTemporary
  };
LevelSolver[numE_Integer, params0_Association, OptionsPattern[]] := Module[
  {ln, simplifier, simpleHam, basis,
  numHam, eigensys, startTime, endTime,
  diagonalTime, params=params0, globalBasis,
  eigenVectors, eigenEnergies, eigenJs,
  states, groundEnergy, allEnergies, PrintFun},
  (
    ln          = theLanthanides[[numE]];
    basis       = BasisLSJ[numE, "AsAssociation"->True];
    simplifier  = OptionValue["Simplifier"];
    PrintFun    = OptionValue["PrintFun"];
    PrintFun["> LevelSolver for ",ln," with ",numE," f-electrons."];
    PrintFun["> Loading the symbolic level Hamiltonian ..."];
    simpleHam   = LevelSimplerSymbolicHamMatrix[numE,
      "Simplifier" -> simplifier,
      "Overwrite"  -> OptionValue["Overwrite Hamiltonian"]
    ];
    (* Everything that is not given is set to zero *)
    PrintFun["> Setting to zero every parameter not given ..."];
    params     = ParamPad[params, "PrintFun" -> PrintFun];
    PrintFun[params];
    (* Create the numeric hamiltonian *)
    PrintFun["> Replacing parameters in the J-blocks of the Hamiltonian to produce numeric arrays ..."];
    numHam     = N /@ Map[ReplaceInSparseArray[#, params]&, simpleHam];
    Clear[simpleHam];
    (* Eigensolver *)
    PrintFun["> Diagonalizing the numerical Hamiltonian within each separate J-subspace ..."];
    startTime     = Now;
    eigensys      = Eigensystem /@ numHam;
    endTime       = Now;
    diagonalTime  = QuantityMagnitude[endTime-startTime,"Seconds"];
    allEnergies   = Flatten[First/@Values[eigensys]];
    groundEnergy  = Min[allEnergies];
    eigensys      = Map[Chop[{#[[1]]-groundEnergy,#[[2]]}]&,eigensys];
    eigensys      = Association@KeyValueMap[#1->{basis[#1],#2} &, eigensys];
    PrintFun[">> Diagonalization took ",diagonalTime," seconds."];
    If[OptionValue["Return as states"],
      (
        PrintFun["> Padding the eigenvectors to correspond to the level basis ..."];
        eigenVectors  = SparseArray @ BlockDiagonalMatrix[Values[#[[2, 2]] & /@ eigensys]];
        globalBasis   = Flatten[Values[basis], 1];
        eigenEnergies = Flatten[Values[#[[2, 1]] & /@ eigensys]];
        eigenJs       = Flatten[KeyValueMap[ConstantArray[#1, Length[#2[[2, 2]]]] &, eigensys]];
        states        = Transpose[{eigenEnergies, eigenJs, eigenVectors}];
        states        = SortBy[states, First];
        eigenVectors  = Last /@ states;
        LSJmultiplets = (RemoveTrailingDigits[#[[1]]] <> ToString[InputForm[#[[2]]]]) & /@ globalBasis;
        majorComponentIndices = Ordering[Abs[#]][[-1]] & /@ eigenVectors;
        levelLabels           = LSJmultiplets[[majorComponentIndices]];
        Return[{globalBasis, levelLabels, states}];
      ),
```

```
        Return[{basis, eigensys}]
      ];
    )
];


(* ################### Level Description ################### *)
(* ######################################################### *)

(* ######################################################### *)
(* ################### Optical Operators ################### *)

magOp = <||>;

JJBlockMagDip::usage = "JJBlockMagDip[numE, J, Jp] returns an array
   for the LSJM matrix elements of the magnetic dipole operator
  between states with given J and Jp. The option \"Sparse\" can be
  used to return a sparse matrix. The default is to return a sparse
  matrix.
See eqn 15.7 in TASS.
Here it is provided in atomic units in which the Bohr magneton is
  1/2.
\[Mu] = -(1/2) (L + gs S)
We are using the Racah convention for the reduced matrix elements
  in the Wigner-Eckart theorem. See TASS eqn 11.15.
";
Options[JJBlockMagDip]={"Sparse"->True};
JJBlockMagDip[numE_, braJ_, ketJ_, OptionsPattern[]] := Module[
  {braSLJs, ketSLJs,
  braSLJ,   ketSLJ,
  braSL,    ketSL,
  braS,     braL,
  ketS,     ketL,
  braMJ,    ketMJ,
  matValue, magMatrix,
  summand1, summand2,
  threejays},
  (
    braSLJs  = AllowedNKSLJMforJTerms[numE,braJ];
    ketSLJs  = AllowedNKSLJMforJTerms[numE,ketJ];
    magMatrix  = Table[
      braSL     = braSLJ[[1]];
      ketSL     = ketSLJ[[1]];
      {braS, braL}   = FindSL[braSL];
      {ketS, ketL}   = FindSL[ketSL];
      braMJ       = braSLJ[[3]];
      ketMJ       = ketSLJ[[3]];
      summand1    = If[Or[braJ  != ketJ,
                          braSL != ketSL],
        0,
        Sqrt[braJ*(braJ+1)*TPO[braJ]]
      ];
      (* looking at the string includes checking L=L', S=S', and \
  alpha=\alpha'*)
      summand2 = If[braSL!= ketSL,
        0,
        (gs-1) *
          Phaser[braS+braL+ketJ+1] *
            Sqrt[TPO[braJ]*TPO[ketJ]] *
              SixJay[{braJ,1,ketJ},{braS,braL,braS}] *
                Sqrt[braS(braS+1)TPO[braS]]
      ];
      matValue = summand1 + summand2;
      (* We are using the Racah convention for red matrix elements
  in Wigner-Eckart *)
      threejays = (ThreeJay[{braJ, -braMJ}, {1, #}, {ketJ, ketMJ}]
  &) /@ {-1,0,1};
      threejays *= Phaser[braJ-braMJ];
      matValue  = - 1/2 * threejays * matValue;
      matValue,
    {braSLJ, braSLJs},
    {ketSLJ, ketSLJs}
    ];
    If[OptionValue["Sparse"],
      magMatrix = SparseArray[magMatrix]
    ];
```

```
      Return[magMatrix];
    )
  ];

  Options[TabulateJJBlockMagDipTable]={"Sparse"->True};
  TabulateJJBlockMagDipTable[numE_,OptionsPattern[]] := (
    JJBlockMagDipTable=<||>;
    Js=AllowedJ[numE];
    Do[
      (
        JJBlockMagDipTable[{numE,braJ,ketJ}]=
          JJBlockMagDip[numE,braJ,ketJ,"Sparse"->OptionValue["Sparse"]]
      ),
    {braJ, Js},
    {ketJ, Js}
    ];
    Return[JJBlockMagDipTable]
  );

  TabulateManyJJBlockMagDipTables::usage = "
    TabulateManyJJBlockMagDipTables[{n1, n2, ...}] calculates the
    tables of matrix elements for the requested f^n_i configurations.
    The function does not return the matrices themselves. It instead
    returns an association whose keys are numE and whose values are
    the filenames where the output of TabulateManyJJBlockMagDipTables
    was saved to. The output consists of an association whose keys are
     of the form {n, J, Jp} and whose values are rectangular arrays
    given the values of <|LSJMJa|H_dip|L'S'J'MJ'a'|>.";
  Options[TabulateManyJJBlockMagDipTables]={"FilenameAppendix"->"","
    Overwrite"->False,"Compressed"->True};
  TabulateManyJJBlockMagDipTables[ns_,OptionsPattern[]] := (
    fnames=<||>;
    Do[
    (
      ExportFun=If[OptionValue["Compressed"],ExportMZip,Export];
      PrintTemporary["#------- numE = ",numE," -------#"];
      appendTo     = (OptionValue["FilenameAppendix"]<>"-magDip");
      exportFname  = JJBlockMatrixFileName[numE,"FilenameAppendix"->
    appendTo];
      fnames[numE] = exportFname;
      If[FileExistsQ[exportFname]&&Not[OptionValue["Overwrite"]],
        Continue[]
      ];
      JJBlockMatrixTable = TabulateJJBlockMagDipTable[numE];
      If[FileExistsQ[exportFname]&&OptionValue["Overwrite"],
        DeleteFile[exportFname]
      ];
      ExportFun[exportFname,  JJBlockMatrixTable];
    ),
    {numE,ns}
    ];
    Return[fnames];
  );

  MagDipoleMatrixAssembly::usage = "MagDipoleMatrixAssembly[numE]
    returns the matrix representation of the operator - 1/2 (L + gs S)
     in the f^numE configuration. The function returns a list with
    three elements corresponding to the x,y,z components of this
    operator. The option \"FilenameAppendix\" can be used to append a
    string to the filename from which the function imports from in
    order to patch together the array. For numE beyond 7 the function
    returns the same as for the complementary configuration. The
    option \"ReturnInBlocks\" can be used to return the matrices in
    blocks. The default is to return the matrices in flattened form
    and as sparse array.";
  Options[MagDipoleMatrixAssembly]={
    "FilenameAppendix"->"",
    "ReturnInBlocks"->False};
  MagDipoleMatrixAssembly[nf_Integer, OptionsPattern[]] := Module[
    {ImportFun, numE, appendTo,
    emFname, JJBlockMagDipTable,
    Js, howManyJs, blockOp,
    rowIdx, colIdx},
    (
      ImportFun = ImportMZip;
```

```
3188      numE      = nf;
3189      numH      = 14 - numE;
3190      numE      = Min[numE,numH];
3191
3192      appendTo  = (OptionValue["FilenameAppendix"]<>"-magDip");
3193      emFname   = JJBlockMatrixFileName[numE,"FilenameAppendix"->
          appendTo];
3194      JJBlockMagDipTable = ImportFun[emFname];
3195
3196      Js        = AllowedJ[numE];
3197      howManyJs = Length[Js];
3198      blockOp   = ConstantArray[0,{howManyJs,howManyJs}];
3199      Do[
3200        blockOp[[rowIdx,colIdx]] = JJBlockMagDipTable[{numE,Js[[
          rowIdx]],Js[[colIdx]]}],
3201        {rowIdx,1,howManyJs},
3202        {colIdx,1,howManyJs}
3203      ];
3204      If[OptionValue["ReturnInBlocks"],
3205        (
3206          opMinus = Map[#[[1]]&, blockOp, {4}];
3207          opZero  = Map[#[[2]]&, blockOp, {4}];
3208          opPlus  = Map[#[[3]]&, blockOp, {4}];
3209          opX =   (opMinus - opPlus)/Sqrt[2];
3210          opY = I (opPlus + opMinus)/Sqrt[2];
3211          opZ = opZero;
3212        ),
3213          blockOp = ArrayFlatten[blockOp];
3214          opMinus = blockOp[[;; , ;; , 1]];
3215          opZero  = blockOp[[;; , ;; , 2]];
3216          opPlus  = blockOp[[;; , ;; , 3]];
3217          opX =   (opMinus - opPlus)/Sqrt[2];
3218          opY = I (opPlus + opMinus)/Sqrt[2];
3219          opZ = opZero;
3220      ];
3221      Return[{opX, opY, opZ}];
3222    )
3223  ];
3224
3225  MagDipLineStrength::usage = "MagDipLineStrength[theEigensys, numE]
       takes the eigensystem of an ion and the number numE of f-electrons
        that correspond to it and calculates the line strength array Stot
       .
3226  The option \"Units\" can be set to either \"SI\" (so that the units
        of the returned array are (A m^2)^2) or to \"Hartree\".
3227  The option \"States\" can be used to limit the states for which the
        line strength is calculated. The default, All, calculates the
       line strength for all states. A second option for this is to
       provide an index labelling a specific state, in which case only
       the line strengths between that state and all the others are
       computed.
3228  The returned array should be interpreted in the eigenbasis of the
       Hamiltonian. As such the element Stot[[i,i]] corresponds to the
       line strength states between states |i> and |j>.";
3229  Options[MagDipLineStrength]={"Reload MagOp" -> False, "Units"->"SI"
       , "States" -> All};
3230  MagDipLineStrength[theEigensys_List, numE0_Integer, OptionsPattern
       []] := Module[
3231    {numE, allEigenvecs, Sx, Sy, Sz, Stot, factor},
3232    (
3233      numE = Min[14-numE0, numE0];
3234      (*If not loaded then load it, *)
3235      If[Or[
3236          Not[MemberQ[Keys[magOp], numE]],
3237          OptionValue["Reload MagOp"]],
3238      (
3239        magOp[numE] = ReplaceInSparseArray[#, {gs->2}]& /@
          MagDipoleMatrixAssembly[numE];
3240      )
3241      ];
3242      allEigenvecs = Transpose[Last /@ theEigensys];
3243      Which[OptionValue["States"] === All,
3244        (
3245          {Sx, Sy, Sz} = (ConjugateTranspose[allEigenvecs].#.
          allEigenvecs) & /@ magOp[numE];
3246          Stot         = Abs[Sx]^2+Abs[Sy]^2+Abs[Sz]^2;
```

```
3247          ),
3248          IntegerQ[OptionValue["States"]],
3249          (
3250            singleState  = theEigensys[[OptionValue["States"],2]];
3251            {Sx, Sy, Sz} = (ConjugateTranspose[allEigenvecs].#.
    singleState) & /@ magOp[numE];
3252            Stot         = Abs[Sx]^2+Abs[Sy]^2+Abs[Sz]^2;
3253          )
3254        ];
3255        Which[
3256          OptionValue["Units"] == "SI",
3257            Return[4 \[Mu]B^2 * Stot],
3258          OptionValue["Units"] == "Hartree",
3259            Return[Stot],
3260          True,
3261          (
3262            Print["Invalid option for \"Units\". Options are \"SI\" and
     \"Hartree\"."];
3263            Abort[];
3264          )
3265        ];
3266      )
3267    ];
3268
3269    MagDipoleRates::usage = "MagDipoleRates[eigenSys, numE] calculates
      the magnetic dipole transition rate array for the provided
      eigensystem. The option \"Units\" can be set to \"SI\" or to \"
      Hartree\". If the option \"Natural Radiative Lifetimes\" is set to
       true then the reciprocal of the rate is returned instead.
      eigenSys is a list of lists with two elements, in each list the
      first element is the energy and the second one the corresponding
      eigenvector.
3270  Based on table 7.3 of Thorne 1999, using g2=1.
3271  The energy unit assumed in eigenSys is kayser.
3272  The returned array should be interpreted in the eigenbasis of the
      Hamiltonian. As such the element AMD[[i,i]] corresponds to the
      transition rate (or the radiative lifetime, depending on options)
      between eigenstates |i> and |j>.
3273  By default this assumes that the refractive index is unity, this
      may be changed by setting the option \"RefractiveIndex\" to the
      desired value.
3274  The option \"Lifetime\" can be used to return the reciprocal of the
       transition rates. The default is to return the transition rates."
      ;
3275  Options[MagDipoleRates]={"Units"->"SI", "Lifetime"->False, "
      RefractiveIndex"->1};
3276  MagDipoleRates[eigenSys_List, numE0_Integer,OptionsPattern[]] :=
      Module[
3277    {AMD, Stot, eigenEnergies,
3278    transitionWaveLengthsInMeters, nRefractive},
3279    (
3280      nRefractive    = OptionValue["RefractiveIndex"];
3281      numE           = Min[14-numE0, numE0];
3282      Stot           = MagDipLineStrength[eigenSys, numE, "Units" ->
    OptionValue["Units"]];
3283      eigenEnergies = Chop[First/@eigenSys];
3284      energyDiffs    = Outer[Subtract, eigenEnergies, eigenEnergies];
3285      energyDiffs    = ReplaceDiagonal[energyDiffs, Indeterminate];
3286      (* Energies assumed in kayser.*)
3287      transitionWaveLengthsInMeters = 0.01/energyDiffs;
3288
3289      unitFactor     = Which[
3290      OptionValue["Units"]=="Hartree",
3291      (
3292        (* The bohrRadius factor in SI needed to convert the
    wavelengths which are assumed in m*)
3293        16 \[Pi]^3 (\[Mu]0Hartree /(3 hPlanckFine)) * bohrRadius^3
3294      ),
3295      OptionValue["Units"]=="SI",
3296      (
3297        16 \[Pi]^3 \[Mu]0/(3 hPlanck)
3298      ),
3299      True,
3300      (
3301        Print["Invalid option for \"Units\". Options are \"SI\" and \
    "Hartree\"."];
```

```
3302          Abort[];
3303        )
3304        ];
3305        AMD = unitFactor / transitionWaveLengthsInMeters^3 * Stot *
      nRefractive^3;
3306        Which[OptionValue["Lifetime"],
3307          Return[1/AMD],
3308          True,
3309          Return[AMD]
3310        ]
3311      )
3312    ];
3313
3314    GroundMagDipoleOscillatorStrength::usage = "
      GroundMagDipoleOscillatorStrength[eigenSys, numE] calculates the
      magnetic diople oscillator strengths between the ground state and
      the excited states as given by eigenSys.
3315  Based on equation 8 of Carnall 1965, removing the 2J+1 factor since
       this degeneracy has been removed by the crystal field.
3316  eigenSys is a list of lists with two elements, in each list the
      first element is the energy and the second one the corresponding
      eigenvector.
3317  The energy unit assumed in eigenSys is Kayser.
3318  The oscillator strengths are dimensionless.
3319  The returned array should be interpreted in the eigenbasis of the
      Hamiltonian. As such the element fMDGS[[i]] corresponds to the
      oscillator strength between ground state and eigenstate |i>.
3320  By default this assumes that the refractive index is unity, this
      may be changed by setting the option \"RefractiveIndex\" to the
      desired value.";
3321  Options[GroundMagDipoleOscillatorStrength]={"RefractiveIndex"->1};
3322  GroundMagDipoleOscillatorStrength[eigenSys_List, numE_Integer,
      OptionsPattern[]] := Module[
3323    {eigenEnergies, SMDGS, GSEnergy, energyDiffs,
3324    transitionWaveLengthsInMeters, unitFactor, nRefractive},
3325    (
3326      eigenEnergies    = First/@eigenSys;
3327      nRefractive      = OptionValue["RefractiveIndex"];
3328      SMDGS            = MagDipLineStrength[eigenSys,numE, "Units"->"
      SI", "States"->1];
3329      GSEnergy         = eigenSys[[1,1]];
3330      energyDiffs      = eigenEnergies-GSEnergy;
3331      energyDiffs[[1]] = Indeterminate;
3332      transitionWaveLengthsInMeters = 0.01/energyDiffs;
3333      unitFactor       = (8\[Pi]^2 me)/(3 hPlanck eCharge^2 cLight);
3334      fMDGS            = unitFactor / transitionWaveLengthsInMeters *
      SMDGS * nRefractive;
3335      Return[fMDGS];
3336    )
3337  ];
3338
3339  (* ##################### Optical Operators ##################### *)
3340  (* ############################################################ *)
3341
3342  (* ############################################################ *)
3343  (* ##################### Printers and Labels ##################### *)
3344
3345  PrintL::usage = "PrintL[L] give the string representation of a
      given angular momentum.";
3346  PrintL[L_] := If[StringQ[L], L, StringTake[specAlphabet, {L + 1}]]
3347
3348  FindSL::usage = "FindSL[LS] gives the spin and orbital angular
      momentum that corresponds to the provided string LS.";
3349  FindSL[SL_] := (
3350    FindSL[SL] =
3351    If[StringQ[SL],
3352      {
3353        (ToExpression[StringTake[SL, 1]]-1)/2,
3354        StringPosition[specAlphabet, StringTake[SL, {2}]][[1, 1]]-1
3355      },
3356      SL
3357    ]
3358  );
3359
3360  PrintSLJ::usage = "Given a list with three elements {S, L, J} this
      function returns a symbol where the spin multiplicity is presented
```

```
    as a superscript, the orbital angular momentum as its
   corresponding spectroscopic letter, and J as a subscript. Function
    does not check to see if the given J is compatible with the given
    S and L.";
PrintSLJ[SLJ_] := (
  RowBox[{
    SuperscriptBox[" ", 2 SLJ[[1]] + 1],
    SubscriptBox[PrintL[SLJ[[2]]], SLJ[[3]]]
    }
    ] // DisplayForm
  );

PrintSLJM::usage = "Given a list with four elements {S, L, J, MJ}
  this function returns a symbol where the spin multiplicity is
  presented as a superscript, the orbital angular momentum as its
  corresponding spectroscopic letter, and {J, MJ} as a subscript. No
   attempt is made to guarantee that the given input is consistent."
  ;
PrintSLJM[SLJM_] := (
  RowBox[{
    SuperscriptBox[" ", 2 SLJM[[1]] + 1],
    SubscriptBox[PrintL[SLJM[[2]]], {SLJM[[3]], SLJM[[4]]}]
    }
    ] // DisplayForm
  );

(* ################### Printers and Labels ################### *)
(* ############################################################ *)

(* ############################################################ *)
(* ###################### Term management ###################### *)

AllowedSLTerms::usage = "AllowedSLTerms[numE] returns a list with
  the allowed terms in the f^numE configuration, the terms are given
   as lists in the format {S, L}. This list may have redundancies
  which are compatible with the degeneracies that might correspond
  to the given case.";
AllowedSLTerms[numE_] := Map[FindSL[First[#]] &, CFPTerms[Min[numE,
   14-numE]]];

AllowedNKSLTerms::usage = "AllowedNKSLTerms[numE] returns a list
  with the allowed terms in the f^numE configuration, the terms are
  given as strings in spectroscopic notation. The integers in the
  last positions are used to distinguish cases with degeneracy.";
AllowedNKSLTerms[numE_] := Map[First, CFPTerms[Min[numE, 14-numE
  ]]];
AllowedNKSLTerms[0]  = {"1S"};
AllowedNKSLTerms[14] = {"1S"};

MaxJ::usage = "MaxJ[numE] gives the maximum J = S+L that
  corresponds to the configuration f^numE.";
MaxJ[numE_] := Max[Map[Total, AllowedSLTerms[Min[numE, 14-numE]]]];

MinJ::usage = "MinJ[numE] gives the minimum J = S+L that
  corresponds to the configuration f^numE.";
MinJ[numE_] := Min[Map[Abs[Part[#, 1] - Part[#, 2]] &,
  AllowedSLTerms[Min[numE, 14-numE]]]]

AllowedSLJTerms::usage = "AllowedSLJTerms[numE] returns a list with
   the allowed {S, L, J} terms in the f^n configuration, the terms
  are given as lists in the format {S, L, J}. This list may have
  repeated elements which account for possible degeneracies of the
  related term.";
AllowedSLJTerms[numE_] := Module[
  {idx1, allowedSL, allowedSLJ},
  (
    allowedSL = AllowedSLTerms[numE];
    allowedSLJ = {};
    For[
      idx1 = 1,
      idx1 <= Length[allowedSL],
      termSL = allowedSL[[idx1]];
      termsSLJ =
        Table[
          {termSL[[1]], termSL[[2]], J},
        {J,Abs[termSL[[1]] - termSL[[2]]], Total[termSL]}
```

```
3412            ];
3413          allowedSLJ = Join[allowedSLJ, termsSLJ];
3414          idx1++
3415        ];
3416        SortBy[allowedSLJ, Last]
3417      )
3418    ];
3419
3420    AllowedNKSLJTerms::usage = "AllowedNKSLJTerms[numE] returns a list
        with the allowed {SL, J} terms in the f^n configuration, the terms
         are given as lists in the format {SL, J} where SL is a string in
        spectroscopic notation.";
3421    AllowedNKSLJTerms[numE_] := Module[
3422      {allowedSL, allowedNKSL, allowedSLJ, nn},
3423      (
3424        allowedNKSL = AllowedNKSLTerms[numE];
3425        allowedSL = AllowedSLTerms[numE];
3426        allowedSLJ = {};
3427        For[
3428          nn = 1,
3429          nn <= Length[allowedSL],
3430          (
3431            termSL = allowedSL[[nn]];
3432            termNKSL = allowedNKSL[[nn]];
3433            termsSLJ =
3434              Table[{termNKSL, J},
3435              {J, Abs[termSL[[1]] - termSL[[2]]], Total[termSL]}
3436              ];
3437            allowedSLJ = Join[allowedSLJ, termsSLJ];
3438            nn++
3439          )
3440        ];
3441        SortBy[allowedSLJ, Last]
3442      )
3443    ];
3444
3445    AllowedNKSLforJTerms::usage = "AllowedNKSLforJTerms[numE, J] gives
        the terms that correspond to the given total angular momentum J in
         the f^n configuration. The result is a list whose elements are
        lists of length 2, the first element being the SL term in
        spectroscopic notation, and the second element being J.";
3446    AllowedNKSLforJTerms[numE_, J_] := Module[
3447      {allowedSL, allowedNKSL, allowedSLJ,
3448      nn, termSL, termNKSL, termsSLJ},
3449      (
3450        allowedNKSL = AllowedNKSLTerms[numE];
3451        allowedSL = AllowedSLTerms[numE];
3452        allowedSLJ = {};
3453        For[
3454          nn = 1,
3455          nn <= Length[allowedSL],
3456          (
3457            termSL = allowedSL[[nn]];
3458            termNKSL = allowedNKSL[[nn]];
3459            termsSLJ = If[Abs[termSL[[1]] - termSL[[2]]] <= J <= Total[
    termSL],
3460              {{termNKSL, J}},
3461              {}
3462              ];
3463            allowedSLJ = Join[allowedSLJ, termsSLJ];
3464            nn++
3465          )
3466        ];
3467        Return[allowedSLJ]
3468      )
3469    ];
3470
3471    AllowedSLJMTerms::usage = "AllowedSLJMTerms[numE] returns a list
        with all the states that correspond to the configuration f^n. A
        list is returned whose elements are lists of the form {S, L, J, MJ
        }.";
3472    AllowedSLJMTerms[numE_] := Module[
3473      {allowedSLJ, allowedSLJM,
3474      termSLJ, termsSLJM, nn},
3475      (
3476      allowedSLJ = AllowedSLJTerms[numE];
```

```
3477      allowedSLJM = {};
3478      For[
3479        nn = 1,
3480        nn <= Length[allowedSLJ],
3481        nn++,
3482        (
3483          termSLJ = allowedSLJ[[nn]];
3484          termsSLJM =
3485            Table[{termSLJ[[1]], termSLJ[[2]], termSLJ[[3]], M},
3486            {M, - termSLJ[[3]], termSLJ[[3]]}
3487            ];
3488          allowedSLJM = Join[allowedSLJM, termsSLJM];
3489        )
3490      ];
3491      Return[SortBy[allowedSLJM, Last]];
3492      )
3493    ];
3494
3495    AllowedNKSLJMforJMTerms::usage = "AllowedNKSLJMforJMTerms[numE, J,
       MJ] returns a list with all the terms that contain states of the f
       ^n configuration that have a total angular momentum J, and a
       projection along the z-axis MJ. The returned list has elements of
       the form {SL (string in spectroscopic notation), J, MJ}.";
3496    AllowedNKSLJMforJMTerms[numE_, J_, MJ_] := Module[
3497      {allowedSL, allowedNKSL,
3498      allowedSLJM, nn},
3499      (
3500        allowedNKSL = AllowedNKSLTerms[numE];
3501        allowedSL   = AllowedSLTerms[numE];
3502        allowedSLJM = {};
3503        For[
3504          nn = 1,
3505          nn <= Length[allowedSL],
3506          termSL = allowedSL[[nn]];
3507          termNKSL = allowedNKSL[[nn]];
3508          termsSLJ = If[(Abs[termSL[[1]] - termSL[[2]]]
3509                         <= J
3510                         <= Total[termSL]
3511                         && (Abs[MJ] <= J)
3512                         ),
3513                         {{termNKSL, J, MJ}},
3514                         {}];
3515          allowedSLJM = Join[allowedSLJM, termsSLJ];
3516          nn++
3517        ];
3518        Return[allowedSLJM];
3519      )
3520    ];
3521
3522    AllowedNKSLJMforJTerms::usage = "AllowedNKSLJMforJTerms[numE, J]
       returns a list with all the states that have a total angular
       momentum J. The returned list has elements of the form {{SL (
       string in spectroscopic notation), J}, MJ}, and if the option \"
       Flat\" is set to True then the returned list has element of the
       form {SL (string in spectroscopic notation), J, MJ}.";
3523    AllowedNKSLJMforJTerms[numE_, J_] := Module[
3524      {MJs, labelsAndMomenta, termsWithJ},
3525      (
3526        MJs = AllowedMforJ[J];
3527        (* Pair LS labels and their {S,L} momenta *)
3528        labelsAndMomenta = ({#, FindSL[#]}) & /@ AllowedNKSLTerms[numE
       ];
3529        (* A given term will contain J if |L-S|<=J<=L+S *)
3530        ContainsJ[{SL_String, {S_, L_}}] := (Abs[S - L] <= J <= (S + L)
       );
3531        (* Keep just the terms that satisfy this condition *)
3532        termsWithJ = Select[labelsAndMomenta, ContainsJ];
3533        (* We don't want to keep the {S,L} *)
3534        termsWithJ = {#[[1]], J} & /@ termsWithJ;
3535        (* This is just a quick way of including up all the MJ values
       *)
3536        Return[Flatten /@ Tuples[{termsWithJ, MJs}]]
3537      )
3538    ];
3539
3540    AllowedMforJ::usage = "AllowedMforJ[J] is shorthand for Range[-J, J
```

```
  , 1].";
AllowedMforJ[J_] := Range[-J, J, 1];

AllowedJ::usage = "AllowedJ[numE] returns the total angular momenta
   J that appear in the f^numE configuration.";
AllowedJ[numE_] := Table[J, {J, MinJ[numE], MaxJ[numE]}];

Seniority::usage = "Seniority[LS] returns the seniority of the
  given term.";
Seniority[LS_] := FindNKLSTerm[LS][[1, 2]];

FindNKLSTerm::usage = "Given the string LS FindNKLSTerm[SL] returns
   all the terms that are compatible with it. This is only for f^n
  configurations. The provided terms might belong to more than one
  configuration. The function returns a list with elements of the
  form {LS, seniority, W, U}.";
FindNKLSTerm[SL_] := Module[
  {NKterms, n},
  (
    n = 7;
    NKterms = {{}};
    Map[
      If[! StringFreeQ[First[#], SL],
        If[ToExpression[Part[#, 2]] <= n,
          NKterms = Join[NKterms, {#}, 1]
        ]
      ] &,
    fnTermLabels
    ];
    NKterms = DeleteCases[NKterms, {}];
    NKterms
  )
];

ParseTermLabels::usage = "ParseTermLabels[] parses the labels for
  the terms in the f^n configurations based on the labels for the f6
   and f7 configurations. The function returns a list whose elements
   are of the form {LS, seniority, W, U}.";
Options[ParseTermLabels] = {"Export" -> True};
ParseTermLabels[OptionsPattern[]] := Module[
  {labelsTextData, fNtextLabels, nielsonKosterLabels,
  seniorities, RacahW, RacahU},
  (
    labelsTextData = FileNameJoin[{moduleDir, "data", "
NielsonKosterLabels_f6_f7.txt"}];
    fNtextLabels   = Import[labelsTextData];
    nielsonKosterLabels = Partition[StringSplit[fNtextLabels], 3];
    termLabels = Map[Part[#, {1}] &, nielsonKosterLabels];
    seniorities = Map[ToExpression[Part[# , {2}]] &,
nielsonKosterLabels];
    racahW =
      Map[
        StringTake[
          Flatten[StringCases[Part[# , {3}],
            "(" ~~ DigitCharacter ~~ DigitCharacter ~~
DigitCharacter ~~ ")"]],
          {2, 4}
        ] &,
      nielsonKosterLabels];
    racahU =
      Map[
        StringTake[
          Flatten[StringCases[Part[# , {3}],
            "(" ~~ DigitCharacter ~~ DigitCharacter ~~ ")"]],
          {2, 3}
        ] &,
      nielsonKosterLabels];
    fnTermLabels = Join[termLabels, seniorities, racahW, racahU,
2];
    fnTermLabels = Sort[fnTermLabels];
    If[OptionValue["Export"],
      (
        broadFname = FileNameJoin[{moduleDir,"data","fnTerms.m"}];
        Export[broadFname, fnTermLabels];
      )
    ];
```

```
3603        Return[fnTermLabels];
3604      )
3605    ];
3606
3607    (* ###################### Term management ###################### *)
3608    (* ############################################################ *)
3609
3610    LoadLaF3Parameters::usage = "LoadLaF3Parameters[ln] takes a string
          with the symbol the element of a trivalent lanthanide ion and
          returns model parameters for it. It is based on the data for LaF3.
           If the option \"Free Ion\" is set to True then the function sets
          all crystal field parameters to zero. Through the option \"gs\" it
           allows modyfing the electronic gyromagnetic ratio. For
          completeness this function also computes the E parameters using
          the F parameters quoted on Carnall.";
3611    Options[LoadLaF3Parameters] = {
3612        "Free Ion"->False,
3613        "gs"->2.002319304386,
3614        "With Uncertainties"->False
3615        };
3616    LoadLaF3Parameters[Ln_String, OptionsPattern[]] := Module[
3617      {params, uncertain,
3618      uncertainKeys, uncertainRules},
3619      (
3620        If[Not[ValueQ[Carnall]],
3621          LoadCarnall[];
3622        ];
3623        params = Association[Carnall["data"][Ln]];
3624        (*If a free ion then all the parameters from the crystal field
        are set to zero*)
3625        If[OptionValue["Free Ion"],
3626          Do[params[cfSymbol] = 0, {cfSymbol, cfSymbols}]
3627        ];
3628        params[F0] = 0;
3629        params[M2] = 0.56 * params[M0]; (*See Carnall 1989,Table I,
        caption,probably fixed based on HF values*)
3630        params[M4] = 0.31 * params[M0]; (*See Carnall 1989,Table I,
        caption,probably fixed based on HF values*)
3631        params[P0] = 0;
3632        params[P4] = 0.5 * params[P2];  (*See Carnall 1989,Table I,
        caption,probably fixed based on HF values*)
3633        params[P6] = 0.1 * params[P2];  (*See Carnall 1989,Table I,
        caption,probably fixed based on HF values*)
3634        params[gs] = OptionValue["gs"];
3635        {params[E0], params[E1], params[E2], params[E3]} = FtoE[params[
        F0], params[F2], params[F4], params[F6]];
3636        params[E0] = 0;
3637        If[
3638          Not[OptionValue["With Uncertainties"]],
3639          Return[params],
3640          (
3641            uncertain     = Association[Carnall["annotations"][Ln]];
3642            uncertainKeys = Keys[uncertain];
3643            uncertain     = If[# == "Not allowed to vary in fitting."
        || # == "Interpolated",
3644                0., #] & /@ uncertain;
3645            paramKeys = Keys[params];
3646            uncertainVals = Sort[Intersection[paramKeys, uncertainKeys
        ]] /. Association[uncertain];
3647            uncertainRules = MapThread[Rule, {Sort[uncertainKeys],
        uncertainVals}];
3648            Which[
3649              MemberQ[{"Ce", "Yb"}, Ln],
3650              (
3651                subsetL = {F0};
3652                subsetR = {0};
3653              ),
3654              True,
3655              (
3656              subsetL = {F0, M2, M4, P0, P4, P6, E0, E1, E2, E3};
3657              subsetR = {0, M0*0.65, M0*0.31, 0, P2*0.5, P2*0.1,
3658                0,
3659                Sqrt[(196 F2^2)/164025 + (49 F4^2)/88209 + (122500 F6
        ^2)/134165889],
3660                Sqrt[F2^2/4100625 + F4^2/10673289 + (30625 F6^2)
        /2743558264161],
```

```
3661              Sqrt[F2^2/18225 + (4 F4^2)/1185921 + (30625 F6^2)
    /1803785841]};
3662            )
3663          ];
3664          uncertainRules = Join[uncertainRules, MapThread[Rule, {
    subsetL,subsetR /. uncertainRules}]];
3665          uncertainRules = Association[uncertainRules];
3666          Which[
3667            Ln == "Eu",
3668            (
3669              uncertainRules[F4] = 12.121;
3670              uncertainRules[F6] = 15.872;
3671            ),
3672            Ln == "Gd",
3673            (
3674              uncertainRules[F4] = 12.07;
3675            ),
3676            Ln == "Tb",
3677            (
3678              uncertainRules[F4] = 41.006;
3679            )
3680          ];
3681          If[MemberQ[{"Eu","Gd","Tb"},Ln],
3682            (
3683              uncertainRules[E1] = Sqrt[(196 F2^2)/164025 + (49 F4^2)
    /88209 + (122500 F6^2)/134165889] /. uncertainRules;
3684              uncertainRules[E2] = Sqrt[F2^2/4100625 + F4^2/10673289
    + (30625 F6^2)/2743558264161] /. uncertainRules;
3685              uncertainRules[E3] = Sqrt[F2^2/18225 + (4 F4^2)/1185921
     + (30625 F6^2)/1803785841] /. uncertainRules;
3686            )
3687          ];
3688          uncertainKeys = First /@ Normal[uncertainRules];
3689          fullParams = Association[MapThread[Rule, {uncertainKeys,
    MapThread[Around, {uncertainKeys /. params, uncertainKeys /.
    uncertainRules}]}]];
3690          Return[Join[params, fullParams]]
3691        )
3692        ];
3693    )
3694  ];
3695
3696  LoadLiYF4Parameters::usage="LoadLiYF4Parameters[ln] takes a string
    with the symbol the element of a trivalent lanthanide ion and
    returns model parameters for it. It return the data for LiYF4 from
     Cheng et al.";
3697  LoadLiYF4Parameters[ln_, OptionsPattern[]]:=(
3698    If[!ValueQ[paramsLiYF4],
3699      paramsChengLiYF4 = Import[FileNameJoin[{moduleDir,"data","
    chengLiYF4.m"}]];
3700    ];
3701    Return[paramsChengLiYF4[ln]];
3702  )
3703
3704  HoleElectronConjugation::usage = "HoleElectronConjugation[params]
    takes the parameters (as an association) that define a
    configuration and converts them so that they may be interpreted as
     corresponding to a complentary hole configuration. Some of this
    can be simply done by changing the sign of the model parameters.
    In the case of the effective three body interaction the
    relationship is more complex and is controlled by the value of the
     t2Switch variable.";
3705  HoleElectronConjugation[params_] := Module[
3706    {newparams = params},
3707    (
3708      flipSignsOf = Join[{ζ} , cfSymbols, TSymbols];
3709      flipped = Table[
3710        (
3711          flipper -> - newparams[flipper]
3712        ),
3713        {flipper, flipSignsOf}
3714        ];
3715      nonflipped = Table[
3716        (
3717          flipper -> newparams[flipper]
3718        ),
```

```
3719          {flipper, Complement[Keys[newparams], flipSignsOf]}
3720          ];
3721        flippedParams = Association[Join[nonflipped, flipped]];
3722        flippedParams = Select[flippedParams, FreeQ[#, Missing]&];
3723        Return[flippedParams];
3724      )
3725    ];
3726
3727    IonSolver::usage = "IonSolver[numE, params, host] puts together (or
        retrieves from disk) the symbolic Hamiltonian for the f^numE
      configuration and solves it for the given params.
3728    params is an Association with keys equal to parameter symbols and
      values their numerical values. The function will replace the
      symbols in the symbolic Hamiltonian with their numerical values
      and then diagonalize the resulting matrix. Any parameter that is
      not defined in the params Association is assumed to be zero.
3729    host is an optional string that may be used to prepend the filename
        of the symbolic Hamiltonian that is saved to disk. The default is
        \"Ln\".
3730    The function returns the eigensystem as a list of lists where in
      each list the first element is the energy and the second element
      the corresponding eigenvector.
3731    Tha ordered basis in which this eigenvector is to be interpreted is
        the one corresponding to BasisLSJMJ[numE].
3732    The function admits the following options:
3733    \"Include Spin-Spin\" (bool) : If True then the spin-spin
      interaction is included as a contribution to the m_k operators.
      The default is True.
3734    \"Overwrite Hamiltonian\" (bool) : If True then the function will
      overwrite the symbolic Hamiltonian that is saved to disk to
      expedite calculations. The default is False. The symbolic
      Hamiltonian is saved to disk to the ./hams/ folder preceded by the
        string host.
3735    \"Zeroes\" (list) : A list with symbols assumed to be zero.
3736    ";
3737    Options[IonSolver] = {
3738      "Include Spin-Spin" -> True,
3739      "Overwrite Hamiltonian" -> False,
3740      "Zeroes" -> {}
3741    };
3742    IonSolver[numE_Integer, params0_Association, host_String:"Ln",
      OptionsPattern[]] := Module[
3743      {ln, simplifier, simpleHam, numHam, eigensys,
3744      startTime, endTime, diagonalTime,
3745      params=params0, zeroSymbols},
3746      (
3747        ln = theLanthanides[[numE]];
3748
3749        (* This could be done when replacing values, but this produces
      smaller saved arrays. *)
3750        simplifier = (#-> 0) & /@ OptionValue["Zeroes"];
3751        simpleHam = SimplerSymbolicHamMatrix[numE,
3752          simplifier,
3753          "PrependToFilename" -> host,
3754          "Overwrite" -> OptionValue["Overwrite Hamiltonian"]
3755        ];
3756
3757        (* Note that we don't have to flip signs of parameters for fn
      beyond f7 since the matrix produced
3758        by SimplerSymbolicHamMatrix has already accounted for this. *)
3759
3760        (* Everything that is not given is set to zero *)
3761        params = ParamPad[params];
3762        PrintFun[params];
3763
3764        (* Enforce the override to the spin-spin contribution to the
      magnetic interactions *)
3765        params[\[Sigma]SS] = If[OptionValue["Include Spin-Spin"], 1,
      0];
3766
3767        (* Create the numeric hamiltonian *)
3768        numHam = ReplaceInSparseArray[simpleHam, params];
3769        Clear[simpleHam];
3770
3771        (* Eigensolver *)
3772        PrintFun["> Diagonalizing the numerical Hamiltonian ..."];
```

```
3773        startTime = Now;
3774        eigensys  = Eigensystem[numHam];
3775        endTime   = Now;
3776        diagonalTime = QuantityMagnitude[endTime - startTime, "Seconds"
      ];
3777        PrintFun[">> Diagonalization took ", diagonalTime, " seconds."
      ];
3778        eigensys = Chop[eigensys];
3779        eigensys = Transpose[eigensys];
3780
3781        (* Shift the baseline energy *)
3782        eigensys = ShiftedLevels[eigensys];
3783        (* Sort according to energy *)
3784        eigensys = SortBy[eigensys, First];
3785        Return[eigensys];
3786      )
3787    ];
3788
3789    ShiftedLevels::usage = "ShiftedLevels[eigenSys] takes a list of
        levels of the form
3790    {{energy_1, coeff_vector_1}, {energy_2, coeff_vector_2},...}} and
        returns the same input except that now to every energy the minimum
         of all of them has been subtracted.";
3791    ShiftedLevels[originalLevels_] := Module[
3792      {groundEnergy, shifted},
3793      (
3794        groundEnergy = Sort[originalLevels][[1,1]];
3795        shifted      = Map[{#[[1]] - groundEnergy, #[[2]]} &,
      originalLevels];
3796        Return[shifted];
3797      )
3798    ];
3799
3800
3801    (* ############################################################ *)
3802    (* ############## Optical Transitions for Levels ############## *)
3803
3804    JuddOfeltUkSquared::usage = "JuddOfeltUkSquared[numE, params]
        calculates the matrix elements of the Uk operator in the level
        basis. These are calculated according to equation (7) in Carnall
        1965.
3805    The function returns a list with the following elements:
3806      - basis : A list with the allowed {SL, J} terms in the f^numE
        configuration. Equal to BasisLSJ[numE].
3807      - eigenSys : A list with the eigensystem of the Hamiltonian for
        the f^n configuration.
3808      - levelLabels : A list with the labels of the major components of
         the level eigenstates.
3809      - LevelUkSquared : An association with the squared matrix
        elements of the Uk operators in the level eigenbasis. The keys
        being {2, 4, 6} corresponding to the rank of the Uk operator. The
        basis in which the matrix elements are given is the one
        corresponding to the level eigenstates given in eigenSys and whose
         major SLJ components are given in levelLabels. The matrix is
        symmetric and given as a SymmetrizedArray.
3810      The function admits the following options:
3811      \"PrintFun\" : A function that will be used to print the progress
         of the calculations. The default is PrintTemporary.";
3812    Options[JuddOfeltUkSquared] = {"PrintFun" -> PrintTemporary};
3813    JuddOfeltUkSquared[numE_, params_, OptionsPattern[]] := Module[
3814      {eigenChanger, numEH, basis, eigenSys,
3815      Js, Ukmat, LevelUkSquared, kRank,
3816      S, L, Sp, Lp, J, Jp, phase,
3817      braTerm, ketTerm, levelLabels,
3818      eigenVecs, majorComponentIndices},
3819      (
3820        If[Not[ValueQ[ReducedUkTable]],
3821          LoadUk[]
3822        ];
3823        numEH = Min[numE, 14-numE];
3824        PrintFun = OptionValue["PrintFun"];
3825        PrintFun["> Calculating the levels for the given parameters ...
      "];
3826        {basis, majorComponents, eigenSys} = LevelSolver[numE, params];
3827        (* The change of basis matrix to the eigenstate basis *)
3828        eigenChanger = Transpose[Last /@ eigenSys];
```

```
3829      PrintFun["Calculating the matrix elements of Uk in the physical
      coupling basis ..."];
3830      LevelUkSquared = <||>;
3831      Do[(
3832        Ukmat = Table[(
3833            {S, L}   = FindSL[braTerm[[1]]];
3834            J        = braTerm[[2]];
3835            Jp       = ketTerm[[2]];
3836            {Sp, Lp} = FindSL[ketTerm[[1]]];
3837            phase    = Phaser[S + Lp + J + kRank];
3838            Simplify @ (
3839              phase *
3840              Sqrt[TPO[J]*TPO[Jp]] *
3841              SixJay[{J, Jp, kRank}, {Lp, L, S}] *
3842              ReducedUkTable[{numEH, 3, braTerm[[1]], ketTerm[[1]],
    kRank}]
3843              )
3844            ),
3845          {braTerm, basis},
3846          {ketTerm, basis}
3847        ];
3848        Ukmat = (Transpose[eigenChanger] . Ukmat . eigenChanger)^2;
3849        Ukmat = Chop@Ukmat;
3850        LevelUkSquared[kRank] = SymmetrizedArray[Ukmat, Dimensions[
    eigenChanger], Symmetric[{1, 2}]];
3851        ),
3852        {kRank, {2, 4, 6}}
3853      ];
3854      LSJmultiplets = (RemoveTrailingDigits[#[[1]]] <> ToString[
    InputForm[#[[2]]]]) & /@ basis;
3855      eigenVecs = Last /@ eigenSys;
3856      majorComponentIndices = Ordering[Abs[#]][[-1]] & /@ eigenVecs;
3857      levelLabels = LSJmultiplets[[majorComponentIndices]];
3858      Return[{basis, eigenSys, levelLabels, LevelUkSquared}];
3859    )
3860  ];
3861
3862  LevelElecDipoleOscillatorStrength::usage = "
      LevelElecDipoleOscillatorStrength[numE, levelParams,
      juddOfeltParams] uses Judd-Ofelt theory to estimate the forced
      electric dipole oscillator strengths ions whose level description
      is determined by levelParams.
3863  The third parameter juddOfeltParams is an asssociation with keys
      equal to the three Judd-Ofelt intensity parameters {\[CapitalOmega
      ]2, \[CapitalOmega]4, \[CapitalOmega]6} and corresponding values
      in cm^2.
3864  The local field correction implemented here corresponds to the one
      given by the virtual cavity model of Lorentz.
3865  The function returns a list with the following elements:
3866    - basis : A list with the allowed {SL, J} terms in the f^numE
      configuration. Equal to BasisLSJ[numE].
3867    - eigenSys : A list with the eigensystem of the Hamiltonian for
      the f^n configuration in the level description.
3868    - levelLabels : A list with the labels of the major components of
       the calculated levels.
3869    - oStrengthArray : A square array whose elements represent the
      oscillator strengths between levels such that the element
      oStrengthArray[[i,j]] is the oscillator strength between the
      levels |Subscript[\[Psi], i]> and |Subscript[\[Psi], j]>. In this
      array, the elements below the diagonal represent emission
      oscillator strengths, and elements above the diagonal represent
      absorption oscillator strengths.
3870  The function admits the following three options:
3871    \"PrintFun\" : A function that will be used to print the progress
       of the calculations. The default is PrintTemporary.
3872    \"RefractiveIndex\" : The refractive index of the medium where
      the transitions are taking place. This may be a number or a
      function. If a number then the oscillator strengths are calculated
       for assuming a wavelength-independent refractive index. If a
      function then the refractive indices are calculated accordingly to
       the wavelength of each transition (the function must admit a
      single argument equal to the wavelength in nm). The default is 1.
3873    \"LocalFieldCorrection\" : The local field correction to be used.
       The default is \"VirtualCavity\". The options are: \"
      VirtualCavity\" and \"EmptyCavity\".
3874  The equation implemented here is the one given in eqn. 29 from the
```

```
    review article of Hehlen (2013). See that same article for a
    discussion on the local field correction.
3875 ";
3876 Options[LevelElecDipoleOscillatorStrength]={
3877   "PrintFun"        -> PrintTemporary,
3878   "RefractiveIndex" -> 1,
3879   "LocalFieldCorrection" -> "VirtualCavity"
3880 };
3881 LevelElecDipoleOscillatorStrength[numE_, levelParams_Association,
    juddOfeltParams_Association, OptionsPattern[]] := Module[
3882   {PrintFun, basis, eigenSys, levelLabels,
3883   LevelUkSquared, eigenEnergies, energyDiffs,
3884   oStrengthArray, nRef, \[Chi], nRefs,
3885   \[Chi]OverN, groundLevel, const,
3886   transitionFrequencies, wavelengthsInNM,
3887   fieldCorrectionType},
3888   (
3889     PrintFun = OptionValue["PrintFun"];
3890     nRef     = OptionValue["RefractiveIndex"];
3891     PrintFun["Calculating the Uk^2 matrix elements for the given
    parameters ..."];
3892     {basis, eigenSys, levelLabels, LevelUkSquared} =
    JuddOfeltUkSquared[numE, levelParams, "PrintFun"->PrintFun];
3893     eigenEnergies = First/@eigenSys;
3894     (* converted to cm^-2 *)
3895     const         = (8\[Pi]^2)/3 me/hPlanck * 10^(-4);
3896     energyDiffs   = Transpose@Outer[Subtract,eigenEnergies,
    eigenEnergies];
3897     (* since energies are assumed in Kayser, speed of light needs
    to be in cm/s, so that the frequencies are in 1/s *)
3898     transitionFrequencies = energyDiffs*cLight*100;
3899     (* grab the J for each level *)
3900     levelJs        = #[[2]] & /@ eigenSys;
3901     oStrengthArray = (
3902       juddOfeltParams[\[CapitalOmega]2]*LevelUkSquared[2]+
3903       juddOfeltParams[\[CapitalOmega]4]*LevelUkSquared[4]+
3904       juddOfeltParams[\[CapitalOmega]6]*LevelUkSquared[6]
3905     );
3906     oStrengthArray = Abs@(const * transitionFrequencies *
    oStrengthArray);
3907     (* it is necessary to divide each oscillator strength by the
    degeneracy of the initial level *)
3908     oStrengthArray = MapIndexed[1/(2 levelJs[[#2[[1]]]]+1) #1 &,
    oStrengthArray,{2}];
3909     (* including the effects of the refractive index *)
3910     fieldCorrectionType = OptionValue["LocalFieldCorrection"];
3911     Which[
3912       nRef === 1,
3913         True,
3914       NumberQ[nRef],
3915       (
3916         \[Chi] = Which[
3917           fieldCorrectionType == "VirtualCavity",
3918           (
3919             ( (nRef^2 + 2) / 3 )^2
3920           ),
3921           fieldCorrectionType == "EmptyCavity",
3922           (
3923             ( 3 * nRef^2 / ( 2 * nRef^2 + 1 ) )^2
3924           )
3925         ];
3926         \[Chi]OverN    = \[Chi] / nRef;
3927         oStrengthArray = \[Chi]OverN * oStrengthArray;
3928         (* the refractive index participates differently in
    absorption and in emission *)
3929         aFunction      = If[#2[[1]]>#2[[2]], #1 * nRef^2, #1]&;
3930         oStrengthArray = MapIndexed[aFunction, oStrengthArray,
    {2}];
3931       ),
3932       True,
3933       (
3934         wavelengthsInNM = Abs[1 / energyDiffs] * 10^7;
3935         nRefs           = Map[nRef, wavelengthsInNM];
3936         Print["Calculating the oscillator strengths for the given
    refractive index ..."];
3937         \[Chi] = Which[
```

```
            fieldCorrectionType == "VirtualCavity",
            (
              ( (nRefs^2 + 2) / 3 )^2
            ),
            fieldCorrectionType == "EmptyCavity",
            (
              ( 3 * nRefs^2 / ( 2*nRefs^2 + 1 ) )^2
            )
          ];
          \[Chi]OverN     = \[Chi] / nRefs;
          oStrengthArray  = \[Chi]OverN * oStrengthArray
        )
      ];
      Return[{basis, eigenSys, levelLabels, oStrengthArray}];
    )
  ];

  LevelJJBlockMagDipole::usage = "LevelJJBlockMagDipole[numE, J, Jp]
    returns an array of the LSJ reduced matrix elements of the
    magnetic dipole operator between states with given J and Jp. The
    option \"Sparse\" can be used to return a sparse matrix. The
    default is to return a sparse matrix.";
  Options[LevelJJBlockMagDipole] = {"Sparse"->True};
  LevelJJBlockMagDipole[numE_, braJ_, ketJ_, OptionsPattern[]] :=
    Module[
    {
      braSLJs, ketSLJs,
      braSLJ, ketSLJ,
      braSL, ketSL,
      braS, braL,
      ketS, ketL,
      matValue, magMatrix,
      summand1, summand2
    },
  (
    braSLJs     = AllowedNKSLforJTerms[numE,braJ];
    ketSLJs     = AllowedNKSLforJTerms[numE,ketJ];
    magMatrix   = Table[
      (
        braSL       = braSLJ[[1]];
        ketSL       = ketSLJ[[1]];
        {braS,braL} = FindSL[braSL];
        {ketS,ketL} = FindSL[ketSL];
        summand1    = If[Or[braJ!=ketJ,braSL!=ketSL],
          0,
          Sqrt[braJ*(braJ+1)*TPO[braJ]
          ]
        ];
        (*looking at the string includes checking L=L',S=S',and\alpha
    =\alpha'*)
        summand2    = If[braSL!=ketSL,
          0,
          (gs-1)*
          Phaser[braS+braL+ketJ+1]*
          Sqrt[TPO[braJ]*TPO[ketJ]]*
          SixJay[{braJ,1,ketJ},{braS,braL,braS}]*
          Sqrt[braS(braS+1)TPO[braS]]
        ];
        matValue = summand1 + summand2;
        matValue = -1/2 * matValue;
        matValue
      ),
      {braSLJ,braSLJs},
      {ketSLJ,ketSLJs}
    ];
    If[OptionValue["Sparse"],
      magMatrix = SparseArray[magMatrix]];
    Return[magMatrix];
  )
  ];

  LevelMagDipoleMatrixAssembly::usage = "LevelMagDipoleMatrixAssembly
    [numE] puts together an array with the reduced matrix elements of
    the magnetic dipole operator in the level basis for the f^numE
    configuration. The function admits the two following options:
    \"Flattened\": If True then the returned matrix is flattened. The
```

```
    default is True.
  \"gs\": The electronic gyromagnetic ratio. The default is 2.";
Options[LevelMagDipoleMatrixAssembly] = {
  "Flattened" -> True,
  gs -> 2
};
LevelMagDipoleMatrixAssembly[numE_, OptionsPattern[]] := Module[
  {Js, magDip, braJ, ketJ},
  (
    Js      = AllowedJ[numE];
    magDip  = Table[
      ReplaceInSparseArray[LevelJJBlockMagDipole[numE, braJ, ketJ],
   {gs -> OptionValue[gs]}],
      {braJ,Js},
      {ketJ,Js}
    ];
    If[OptionValue["Flattened"],
      magDip = ArrayFlatten[magDip];
    ];
    Return[magDip];
  )
];

LevelMagDipoleLineStrength::usage = "LevelMagDipoleLineStrength[
  eigenSys, numE] calculates the magnetic dipole line strengths for
  an ion whose level description is determined by levelParams. The
  function returns a square array whose elements represent the
  magnetic dipole line strengths between the levels given in
  eigenSys such that the element magDipoleLineStrength[[i,j]] is the
   line strength between the levels |Subscript[\[Psi], i]> and |
  Subscript[\[Psi], j]>. Eigensys must be such that it consists of a
   lists of lists where in each list the last element corresponds to
   the eigenvector of a level (given as a row) in the standard basis
   for levels of the f^numE configuration.
The function admits the following options:
  \"Units\" : The units in which the line strengths are given. The
  default is \"SI\". The options are \"SI\" and \"Hartree\". If \"SI
  \" then the unit of the line strength is (A m^2)^2 = (J/T)^2. If \
  "Hartree\" then the line strength is given in units of 2 \[Mu]B.";
Options[LevelMagDipoleLineStrength] = {
  "Units"->"SI"
  };
LevelMagDipoleLineStrength[theEigensys_List, numE0_Integer,
  OptionsPattern[]] := Module[
  {numE, levelMagOp, allEigenvecs, magDipoleLineStrength, units},
  (
    numE          = Min[14-numE0,numE0];
    levelMagOp    = LevelMagDipoleMatrixAssembly[numE];
    allEigenvecs  = Transpose[Last/@theEigensys];
    units         = OptionValue["Units"];
    magDipoleLineStrength          = Transpose[allEigenvecs].
  levelMagOp.allEigenvecs;
    magDipoleLineStrength          = Abs[magDipoleLineStrength]^2;
    Which[
      units=="SI",
        Return[4 \[Mu]B^2 * magDipoleLineStrength],
      units=="Hartree",
        Return[magDipoleLineStrength]
    ];
  )
];

LevelMagDipoleOscillatorStrength::usage = "
  LevelMagDipoleOscillatorStrength[eigenSys, numE] calculates the
  magnetic dipole oscillator strengths for an ion whose level
  description is determined by levelParams. The refractive index of
  the medium is relevant, but here it is assumed to be 1, this can
  be changed through the option \"RefractiveIndex\". eigenSys must
  consist of a lists of lists with three elements: the first element
   being the energy of the level, the second element being the J of
  the level, and the third element being the eigenvector of the
  level.
   The function returns a list with the following elements:
    - basis : A list with the allowed {SL, J} terms in the f^numE
  configuration. Equal to BasisLSJ[numE].
    - eigenSys : A list with the eigensystem of the Hamiltonian for
```

```
    the f^n configuration in the level description.
      - levelLabels : A list with the labels of the major components
    of the calculated levels.
      - magDipoleOstrength : A square array whose elements represent
    the magnetic dipole oscillator strengths between the levels given
    in eigenSys such that the element magDipoleOstrength[[i,j]] is the
     oscillator strength between the levels |Subscript[\[Psi], i]> and
     |Subscript[\[Psi], j]>. In this array the elements below the
    diagonal represent emission oscillator strengths, and elements
    above the diagonal represent absorption oscillator strengths. The
    emission oscillator strengths are negative. The oscillator
    strength is a dimensionless quantity.
    The function admits the following option:
      \"RefractiveIndex\" : The refractive index of the medium where
    the transitions are taking place. This may be a number or a
    function. If a number then the oscillator strengths are calculated
     assuming a wavelength-independent refractive index as given. If a
     function then the refractive indices are calculated accordingly
    to the vaccum wavelength of each transition (the function must
    admit a single argument equal to the wavelength in nm). The
    default is 1.
    For reference see equation (27.8) in Rudzikas (2007). The
    expression for the line strenght is the simplest when using atomic
     units, (27.8) is missing a factor of α^2.";
Options[LevelMagDipoleOscillatorStrength]={
  "RefractiveIndex" -> 1
};
LevelMagDipoleOscillatorStrength[eigenSys_, numE_, OptionsPattern
  []] := Module[
  {eigenEnergies, eigenVecs, levelJs,
  energyDiffs, magDipoleOstrength, nRef,
  wavelengthsInNM, nRefs, degenDivisor},
  (
    basis          = BasisLSJ[numE];
    eigenEnergies  = First/@eigenSys;
    nRef           = OptionValue["RefractiveIndex"];
    eigenVecs      = Last/@eigenSys;
    levelJs        = #[[2]]&/@eigenSys;
    energyDiffs    = -Outer[Subtract,eigenEnergies,eigenEnergies];
    energyDiffs  *= kayserToHartree;
    magDipoleOstrength = LevelMagDipoleLineStrength[eigenSys, numE,
   "Units"->"Hartree"];
    magDipoleOstrength = 2/3 * αFine^2 * energyDiffs *
  magDipoleOstrength;
    degenDivisor       = #1 / ( 2 * levelJs[[#2[[1]]]] + 1 ) &;
    magDipoleOstrength = MapIndexed[degenDivisor,
  magDipoleOstrength, {2}];
    Which[nRef===1,
      True,
      NumberQ[nRef],
        (
          magDipoleOstrength = nRef * magDipoleOstrength;
        ),
      True,
        (
          wavelengthsInNM    = Abs[kayserToHartree / energyDiffs] *
   10^7;
          nRefs              = Map[nRef, wavelengthsInNM];
          magDipoleOstrength = nRefs * magDipoleOstrength;
        )
    ];
    Return[{basis, eigenSys, magDipoleOstrength}];
  )
];

LevelMagDipoleSpontaneousDecayRates::usage = "
  LevelMagDipoleSpontaneousDecayRates[eigenSys, numE] calculates the
   spontaneous emission rates for the magnetic dipole transitions
  between the levels given in eigenSys. The function returns a
  square array whose elements represent the spontaneous emission
  rates between the levels given in eigenSys such that the element
  [[i,j]] of the returned array is the rate of spontaneous emission
  from the level |Subscript[\[Psi], i]> to the level |Subscript[\[
  Psi], j]>. In this array the elements below the diagonal represent
   emission rates, and elements above the diagonal are identically
  zero.
```

```
The function admits two optional arguments:
  + \"Units\" : The units in which the rates are given. The default
   is \"SI\". The options are \"SI\" and \"Hartree\". If \"SI\" then
   the rates are given in s^-1. If \"Hartree\" then the rates are
  given in the atomic unit of frequency.
  + \"RefractiveIndex\" : The refractive index of the medium where
  the transitions are taking place. This may be a number or a
  function. If a number then the rates are calculated assuming a
  wavelength-independent refractive index as given. If a function
  then the refractive indices are calculated accordingly to the
  vaccum wavelength of each transition (the function must admit a
  single argument equal to the wavelength in nm). The default is 1."
  ;
Options[LevelMagDipoleSpontaneousDecayRates] = {
  "Units" -> "SI",
  "RefractiveIndex" -> 1};
LevelMagDipoleSpontaneousDecayRates[eigenSys_List, numE_Integer,
  OptionsPattern[]] := Module[
  {levMDlineStrength, eigenEnergies, energyDiffs,
  levelJs, spontaneousRatesInHartree, spontaneousRatesInSI,
  degenDivisor, units, nRef, nRefs, wavelengthsInNM},
  (
    nRef              = OptionValue["RefractiveIndex"];
    units             = OptionValue["Units"];
    levMDlineStrength =
  LowerTriangularize@LevelMagDipoleLineStrength[eigenSys,numE,"Units
  "->"Hartree"];
    levMDlineStrength = SparseArray[levMDlineStrength];
    eigenEnergies     = First /@ eigenSys;
    energyDiffs       = Outer[Subtract, eigenEnergies,
  eigenEnergies];
    energyDiffs       = kayserToHartree * energyDiffs;
    energyDiffs       = SparseArray[LowerTriangularize[energyDiffs
  ]];
    levelJs           = #[[2]]& /@ eigenSys;
    spontaneousRatesInHartree = 4/3 αFine^5 * energyDiffs^3 *
  levMDlineStrength;
    degenDivisor              = #1 / (2*levelJs[[#2[[1]]]] + 1)&;
    spontaneousRatesInHartree = MapIndexed[degenDivisor,
  spontaneousRatesInHartree, {2}];
    Which[nRef===1,
      True,
      NumberQ[nRef],
        (
          spontaneousRatesInHartree = nRef^3 *
  spontaneousRatesInHartree;
        ),
      True,
        (
          wavelengthsInNM    = Abs[kayserToHartree / energyDiffs] *
   10^7;
          nRefs              = Map[nRef, wavelengthsInNM];
          spontaneousRatesInHartree = nRefs^3 *
  spontaneousRatesInHartree;
        )
    ];
    If[units == "SI",
      (
        spontaneousRatesInSI = 1/hartreeTime *
  spontaneousRatesInHartree;
        Return[SparseArray@spontaneousRatesInSI];
      ),
      Return[SparseArray@spontaneousRatesInHartree];
    ];
  )
];

(* ############# Optical Transitions for Levels ############## *)
(* ############################################################ *)

(* ############################################################ *)
(* ################## Eigensystem analysis ################### *)

PrettySaundersSL::usage = "PrettySaundersSL[SL] produces a human-
  redeable symbol for the spectroscopic term SL. SL can be either a
  string (in RS notation for the term) or a list of two numbers {S,
```

```
    L}. The option \"Representation\" can be used to specify whether
    the output is given as a symbol or as a ket. The default is \"Ket\
    ".";
4149  Options[PrettySaundersSL] = {"Representation" -> "Ket"};
4150  PrettySaundersSL[SL_, OptionsPattern[]] := (
4151    If[StringQ[SL],
4152    (
4153      {S,L} = FindSL[SL];
4154      L     = StringTake[SL,{2,-1}];
4155    ),
4156      {S,L}=SL
4157    ];
4158    pretty = RowBox[{
4159        AdjustmentBox[Style[2*S+1,Smaller], BoxBaselineShift->-1,
    BoxMargins->0],
4160        AdjustmentBox[PrintL[L]]
4161      }
4162    ];
4163    pretty = DisplayForm[pretty];
4164    pretty = Which[
4165      OptionValue["Representation"]=="Ket",
4166        Ket[pretty],
4167      OptionValue["Representation"]=="Symbol",
4168        pretty
4169    ];
4170    Return[pretty];
4171  );
4172
4173  PrettySaundersSLJmJ::usage = "PrettySaundersSLJmJ[{SL, J, mJ}]
    produces a human-redeable symbol for the given basis vector {SL, J
    , mJ}.";
4174  Options[PrettySaundersSLJmJ] = {"Representation" -> "Ket"};
4175  PrettySaundersSLJmJ[{SL_, J_, mJ_}, OptionsPattern[]] := (If[
4176    StringQ[SL],
4177    ({S, L} = FindSL[SL];
4178      L = StringTake[SL, {2, -1}];
4179      ),
4180    {S, L} = SL];
4181    pretty = RowBox[{AdjustmentBox[Style[2*S + 1, Smaller],
4182        BoxBaselineShift -> -1, BoxMargins -> 0],
4183        AdjustmentBox[PrintL[L], BoxMargins -> -0.2],
4184        AdjustmentBox[
4185        Style[Row[{InputForm[J], ",", mJ}], Small],
4186        BoxBaselineShift -> 1,
4187        BoxMargins -> {{0.7, 0}, {0.4, 0.4}}]}];
4188    pretty = DisplayForm[pretty];
4189    If[OptionValue["Representation"] == "Ket",
4190      pretty = Ket[pretty]
4191    ];
4192    Return[pretty];
4193  );
4194
4195  PrettySaundersSLJ::usage = "PrettySaundersSLJ[{SL, J}] produces a
    human-redeable symbol for the given basis vector {SL, J}. SL can
    be either a list of two numbers representing S and L or a string
    representing the spin multiplicity and the total orbital angular
    momentum J in spectroscopic notation. The option \"Representation\
    " can be used to specify whether the output is given as a symbol
    or as a ket. The default is \"Ket\".";
4196  Options[PrettySaundersSLJ] = {"Representation"->"Ket"};
4197  PrettySaundersSLJ[{SL_,J_},OptionsPattern[]] := (
4198    If[StringQ[SL],
4199    (
4200      {S,L}=FindSL[SL];
4201      L=StringTake[SL,{2,-1}];
4202    ),
4203      {S,L}=SL
4204    ];
4205    pretty = RowBox[{
4206        AdjustmentBox[Style[2*S+1,Smaller],BoxBaselineShift->-1,
    BoxMargins->0],
4207        AdjustmentBox[PrintL[L],BoxMargins->-0.2],
4208        AdjustmentBox[Style[InputForm[J],Small,FontTracking->"Narrow"
    ],BoxBaselineShift->1,BoxMargins->{{0.7,0},{0.4,0.4}}]
4209      }
4210    ];
```

```
    pretty = DisplayForm[pretty];
    pretty = Which[
      OptionValue["Representation"]=="Ket",
        Ket[pretty],
      OptionValue["Representation"]=="Symbol",
        pretty
    ];
    Return[pretty];
  );

  BasisVecInRusselSaunders::usage = "BasisVecInRusselSaunders[
    basisVec] takes a basis vector in the format {LSstring, Jval,
    mJval} and returns a human-readable symbol for the corresponding
    Russel-Saunders term.";
  BasisVecInRusselSaunders[basisVec_] := (
    {LSstring, Jval, mJval} = basisVec;
    Ket[PrettySaundersSLJmJ[basisVec]]
  );

  LSJMJTemplate =
    StringTemplate[
    "\!\(\*TemplateBox[{\nRowBox[{\"`LS`\", \",\", \nRowBox[{\"J\", \
  \"=\", \"`J`\"}], \",\", \nRowBox[{\"mJ\", \"=\", \"`mJ`\"}]}]},\n\
  \"Ket\"]\)"];

  BasisVecInLSJMJ::usage = "BasisVecInLSJMJ[basisVec] takes a basis
    vector in the format {{{LSstring, Jval}, mJval}, nucSpin} and
    returns a human-readable symbol for the corresponding LSJMJ term
    in the form |LS, J=..., mJ=...>.";
  BasisVecInLSJMJ[basisVec_] := (
    {LSstring, Jval, mJval} = basisVec;
    LSJMJTemplate[<|
      "LS" -> LSstring,
      "J" -> ToString[Jval, InputForm],
      "mJ" -> ToString[mJval, InputForm]|>]
  );

  ParseStates::usage = "ParseStates[eigenSys, basis] takes a list of
    eigenstates in terms of their coefficients in the given basis and
    returns a list of the same states in terms of their energy, LSJMJ
    symbol, J, mJ, S, L, LSJ symbol, and LS symbol. eigenSys is a list
     of lists with two elements, in each list the first element is the
     energy and the second one the corresponding eigenvector. The LS
    symbol returned corresponds to the term with the largest
    coefficient in the given basis.";
  ParseStates[states_, basis_, OptionsPattern[]] := Module[
    {parsedStates},
    (
      parsedStates = Table[(
        {energy, eigenVec}      = state;
        maxTermIndex            = Ordering[Abs[eigenVec]][[-1]];
        {LSstring, Jval, mJval} = basis[[maxTermIndex]];
        LSJsymbol               = Subscript[LSstring, {Jval, mJval}];
        LSJMJsymbol             = LSstring <> ToString[Jval,
    InputForm];
        {S, L}                  = FindSL[LSstring];
        {energy, LSstring, Jval, mJval, S, L, LSJsymbol, LSJMJsymbol}
      ),
      {state, states}
      ];
      Return[parsedStates];
    )
  ];

  ParseStatesByNumBasisVecs::usage = "ParseStatesByNumBasisVecs[
    eigenSys, basis, numBasisVecs, roundTo] takes a list of
    eigenstates (given in eigenSys) in terms of their coefficients in
    the given basis and returns a list of the same states in terms of
    their energy and the coefficients at most numBasisVecs basis
    vectors. By default roundTo is 0.01 and this is the value used to
    round the amplitude coefficients. eigenSys is a list of lists with
     two elements, in each list the first element is the energy and
    the second one the corresponding eigenvector.
  The option \"Coefficients\" can be used to specify whether the
    coefficients are given as \"Amplitudes\" or \"Probabilities\". The
     default is \"Amplitudes\".
```

```
";
Options[ParseStatesByNumBasisVecs] = {
  "Coefficients" -> "Amplitudes",
  "Representation" -> "Ket",
  "ReturnAs" -> "Dot"
  };
ParseStatesByNumBasisVecs[eigensys_List, basis_List,
  numBasisVecs_Integer, roundTo_Real : 0.01, OptionsPattern[]] :=
  Module[
  {parsedStates, energy, eigenVec,
  probs, amplitudes, ordering,
  returnAs,
  chosenIndices, majorComponents,
  majorAmplitudes, majorRep},
  (
    returnAs     = OptionValue["ReturnAs"];
    parsedStates = Table[(
      {energy, eigenVec} = state;
      energy             = Chop[energy];
      probs              = Round[Abs[eigenVec^2], roundTo];
      amplitudes         = Round[eigenVec, roundTo];
      ordering           = Ordering[probs];
      chosenIndices      = ordering[[-numBasisVecs ;;]];
      majorComponents    = basis[[chosenIndices]];
      majorThings = If[OptionValue["Coefficients"] == "
  Probabilities",
        (
          probs[[chosenIndices]]
        ),
        (
          amplitudes[[chosenIndices]]
        )
      ];
      majorComponents    = PrettySaundersSLJmJ[#, "Representation"
  -> OptionValue["Representation"]] & /@ majorComponents;
      nonZ               = (# != 0.) & /@ majorThings;
      majorThings        = Pick[majorThings, nonZ];
      majorComponents    = Pick[majorComponents, nonZ];
      If[OptionValue["Coefficients"] == "Probabilities",
        (
          majorThings = majorThings * 100 * "%";
        )
      ];
      majorRep  = Which[
                  returnAs == "Dot",
                    majorThings . majorComponents,
                  returnAs == "List",
                    Transpose[{Reverse@majorThings,
  Reverse@majorComponents}]
                  ];
      {energy, majorRep}
      ),
      {state, eigensys}];
    Return[parsedStates]
  )
];

FindThresholdPosition::usage = "FindThresholdPosition[list,
  threshold] returns the position of the first element in list that
  is greater than or equal to threshold. If no such element exists,
  it returns the length of list. The elements of the given list must
   be in ascending order.";
FindThresholdPosition[list_, threshold_] := Module[
  {position},
  (
  position = Position[list, _?(# >= threshold &), 1, 1];
  thrPos = If[Length[position] > 0,
    position[[1, 1]],
    Length[list]];
  If[thrPos == 0,
    Return[1],
    Return[thrPos]]
  )
];

ParseStateByProbabilitySum[{energy_, eigenVec_}, probSum_, roundTo_
```

```
    :0.01, maxParts_:20] := Compile[
4330    {{energy, _Real, 0}, {eigenVec, _Complex, 1},
4331     {probSum, _Real, 0},{roundTo, _Real, 0},
4332     {maxParts, _Integer, 0}},
4333      Module[
4334        {numStates, state, amplitudes, probs, ordering,
4335        orderedProbs, truncationIndex, accProb, thresholdIndex,
    chosenIndices, majorComponents,
4336        majorAmplitudes, absMajorAmplitudes, notnullAmplitudes,
    majorRep},
4337        (
4338          numStates    = Length[eigenVec];
4339          (*Round them up*)
4340          amplitudes         = Round[eigenVec, roundTo];
4341          probs              = Round[Abs[eigenVec^2], roundTo];
4342          ordering           = Reverse[Ordering[probs]];
4343          (*Order the probabilities from high to low*)
4344          orderedProbs       = probs[[ordering]];
4345          (*To speed up Accumulate, assume that only as much as
    maxParts will be needed*)
4346          truncationIndex    = Min[maxParts, Length[orderedProbs]];
4347          orderedProbs       = orderedProbs[[;;truncationIndex]];
4348          (*Accumulate the probabilities*)
4349          accProb            = Accumulate[orderedProbs];
4350          (*Find the index of the first element in accProb that is
    greater than probSum*)
4351          thresholdIndex     = Min[Length[accProb],
    FindThresholdPosition[accProb, probSum]];
4352          (*Grab all the indicees up till that one*)
4353          chosenIndices      = ordering[[;; thresholdIndex]];
4354          (*Select the corresponding elements from the basis*)
4355          majorComponents    = basis[[chosenIndices]];
4356          (*Select the corresponding amplitudes*)
4357          majorAmplitudes    = amplitudes[[chosenIndices]];
4358          (*Take their absolute value*)
4359          absMajorAmplitudes = Abs[majorAmplitudes];
4360          (*Make sure that there are no effectively zero
    contributions*)
4361          notnullAmplitudes  = Flatten[Position[absMajorAmplitudes,
    x_ /; x != 0]];
4362          (* majorComponents    = PrettySaundersSLJmJ
    [{#[[1]],#[[2]],#[[3]]} & /@ majorComponents; *)
4363          majorComponents    = PrettySaundersSLJmJ /@ majorComponents
    ;
4364          majorAmplitudes    = majorAmplitudes[[notnullAmplitudes]];
4365          (*Make them into Kets*)
4366          majorComponents    = Ket /@ majorComponents[[
    notnullAmplitudes]];
4367          (*Multiply and add to build the final Ket*)
4368          majorRep           = majorAmplitudes . majorComponents;
4369          Return[{energy, majorRep}];
4370        )
4371      ],
4372    CompilationTarget -> "C",
4373    RuntimeAttributes -> {Listable},
4374    Parallelization -> True,
4375    RuntimeOptions -> "Speed"
4376  ];
4377
4378  ParseStatesByProbabilitySum::usage = "ParseStatesByProbabilitySum[
      eigensys, basis, probSum] takes a list of eigenstates in terms of
      their coefficients in the given basis and returns a list of the
      same states in terms of their energy and the coefficients of the
      basis vectors that sum to at least probSum.";
4379  ParseStatesByProbabilitySum[eigensys_, basis_, probSum_, roundTo_ :
      0.01, maxParts_: 20] := Module[
4380    {parsedByProb, numStates, state, energy,
4381    eigenVec, amplitudes, probs, ordering,
4382    orderedProbs, truncationIndex, accProb,
4383    thresholdIndex, chosenIndices, majorComponents,
4384    majorAmplitudes, absMajorAmplitudes, notnullAmplitudes, majorRep
    },
4385    (
4386      numStates    = Length[eigensys];
4387      parsedByProb = Table[(
4388        state              = eigensys[[idx]];
```

```
4389          {energy, eigenVec} = state;
4390          (*Round them up*)
4391          amplitudes         = Round[eigenVec, roundTo];
4392          probs              = Round[Abs[eigenVec^2], roundTo];
4393          ordering           = Reverse[Ordering[probs]];
4394          (*Order the probabilities from high to low*)
4395          orderedProbs       = probs[[ordering]];
4396          (*To speed up Accumulate, assume that only as much as
        maxParts will be needed*)
4397          truncationIndex    = Min[maxParts, Length[orderedProbs]];
4398          orderedProbs       = orderedProbs[[;;truncationIndex]];
4399          (*Accumulate the probabilities*)
4400          accProb            = Accumulate[orderedProbs];
4401          (*Find the index of the first element in accProb that is
        greater than probSum*)
4402          thresholdIndex     = Min[Length[accProb],
        FindThresholdPosition[accProb, probSum]];
4403          (*Grab all the indicees up till that one*)
4404          chosenIndices      = ordering[[;; thresholdIndex]];
4405          (*Select the corresponding elements from the basis*)
4406          majorComponents    = basis[[chosenIndices]];
4407          (*Select the corresponding amplitudes*)
4408          majorAmplitudes    = amplitudes[[chosenIndices]];
4409          (*Take their absolute value*)
4410          absMajorAmplitudes = Abs[majorAmplitudes];
4411          (*Make sure that there are no effectively zero contributions
        *)
4412          notnullAmplitudes  = Flatten[Position[absMajorAmplitudes, x_
        /; x != 0]];
4413          (* majorComponents    = PrettySaundersSLJmJ
        [{#[[1]],#[[2]],#[[3]]}] & /@ majorComponents; *)
4414          majorComponents    = PrettySaundersSLJmJ /@ majorComponents;
4415          majorAmplitudes    = majorAmplitudes[[notnullAmplitudes]];
4416          majorComponents    = majorComponents[[notnullAmplitudes]];
4417          (*Multiply and add to build the final Ket*)
4418          majorRep           = majorAmplitudes . majorComponents;
4419          {energy, majorRep}
4420          ), {idx, numStates}];
4421        Return[parsedByProb];
4422      )
4423    ];
4424
4425    (* ################## Eigensystem analysis ################### *)
4426    (* ############################################################ *)
4427
4428    (* ############################################################ *)
4429    (* ########################### Misc ########################### *)
4430
4431    SymbToNum::usage = "SymbToNum[expr, numAssociation] takes an
          expression expr and returns what results after making the
          replacements defined in the given replacementAssociation. If
          replacementAssociation doesn't define values for expected keys,
          they are taken to be zero.";
4432    SymbToNum[expr_, replacementAssociation_] := (
4433      includedKeys = Keys[replacementAssociation];
4434      (*If a key is not defined, make its value zero.*)
4435      fullAssociation = Table[(
4436        If[MemberQ[includedKeys, key],
4437          ToExpression[key]->replacementAssociation[key],
4438          ToExpression[key]->0
4439        ]
4440      ),
4441      {key, paramSymbols}];
4442      Return[expr/.fullAssociation];
4443    );
4444
4445    SimpleConjugate::usage = "SimpleConjugate[expr] takes an expression
          and applies a simplified version of the conjugate in that all it
          does is that it replaces the imaginary unit I with -I. It assumes
          that every other symbol is real so that it remains the same under
          complex conjugation. Among other expressions it is valid for any
          rational or polynomial expression with complex coefficients and
          real variables.";
4446    SimpleConjugate[expr_] := expr /. Complex[a_, b_] :> a - I b;
4447
4448    ExportMZip::usage = "ExportMZip[\"dest.[zip,m]\"] saves a
```

```
  compressed version of expr to the given destination.";
ExportMZip[filename_, expr_] := Module[
  {baseName, exportName, mImportName, zipImportName},
  (
    baseName    = FileBaseName[filename];
    exportName  = StringReplace[filename,".m"->".zip"];
    mImportName = StringReplace[exportName,".zip"->".m"];
    If[FileExistsQ[mImportName],
    (
      PrintTemporary[mImportName<>" exists already, deleting"];
      DeleteFile[mImportName];
      Pause[2];
    )
    ];
    Export[exportName, (baseName<>".m") -> expr];
  )
];

ImportMZip::usage = "ImportMZip[filename] imports a .m file inside
  a .zip file with corresponding filename. If the Option \"Leave
  Uncompressed\" is set to True (the default) then this function
  also leaves an umcompressed version of the object in the same
  folder of filename";
Options[ImportMZip]={"Leave Uncompressed" -> True};
ImportMZip[filename_String, OptionsPattern[]] := Module[
  {baseName, importKey, zipImportName, mImportName, imported},
  (
    baseName      = FileBaseName[filename];
    (*Function allows for the filename to be .m or .zip*)
    importKey     = baseName <> ".m";
    zipImportName = StringReplace[filename, ".m"->".zip"];
    mImportName   = StringReplace[zipImportName, ".zip"->".m"];
    mxImportName  = StringReplace[zipImportName, ".zip"->".mx"];
    Which[
      FileExistsQ[mxImportName],
    (
      PrintTemporary[".mx version exists already, importing that
  instead ..."];
      Return[Import[mxImportName]];
    ),
      FileExistsQ[mImportName],
    (
      PrintTemporary[".m version exists already, importing that
  instead ..."];
      Return[Import[mImportName]];
    )
    ];
    imported = Import[zipImportName, importKey];
    If[OptionValue["Leave Uncompressed"],
      (
        Export[mImportName, imported];
        Export[mxImportName, imported];
      )
    ];
    Return[imported];
  )
];

ReplaceInSparseArray::usage = "ReplaceInSparseArray[sparseArray,
  rules] takes a sparse array that may contain symbolic quantities
  and returns a sparse array in which the given rules have been used
   on every element.";
ReplaceInSparseArray[sparseA_SparseArray, rules_] := (
  SparseArray[Automatic,
    sparseA["Dimensions"],
    sparseA["Background"] /. rules,
    {
      1,
      {sparseA["RowPointers"], sparseA["ColumnIndices"]},
      sparseA["NonzeroValues"] /. rules
    }
    ]
);

MapToSparseArray::usage = "MapToSparseArray[sparseArray, function]
  takes a sparse array and returns a sparse array after the function
```

```
    has been applied to it.";
MapToSparseArray[sparseA_SparseArray, func_] := Module[
  {nonZ, backg, mapped},
  (
    nonZ   = func /@ sparseA["NonzeroValues"];
    backg  = func[sparseA["Background"]];
    mapped = SparseArray[Automatic,
      sparseA["Dimensions"],
      backg,
      {
        1,
        {sparseA["RowPointers"], sparseA["ColumnIndices"]},
        nonZ
      }
    ];
    Return[mapped];
  )
];

ParseTeXLikeSymbol::usage = "ParseTeXLikeSymbol[string] parses a
  string for a symbol given in LaTeX notation and returns a
  corresponding mathematica symbol. The string may have expressions
  for several symbols, they need to be separated by single spaces.
  In addition the _ and ^ symbols used in LaTeX notation need to
  have arguments that are enclosed in parenthesis, for example \"x_2
  \" is invalid, instead \"x_{2}\" should have been given.";
Options[ParseTeXLikeSymbol] = {"Form" -> "List"};
ParseTeXLikeSymbol[bigString_, OptionsPattern[]] := Module[
  {form, mainSymbol, symbols},
  (
    form = OptionValue["Form"];
    (* parse greek *)
    symbols = Table[(
        str = StringReplace[string, {"\\alpha" -> "α",
          "\\beta" -> "β",
          "\\gamma" -> "γ",
          "\\psi" -> "\[Psi]"}];
        symbol = Which[
          StringContainsQ[str, "_"] && Not[StringContainsQ[str, "^"
  ]],
          (
            (*yes sub no sup*)
            mainSymbol = StringSplit[str, "_"][[1]];
            mainSymbol = ToExpression[mainSymbol];

            subPart =
              StringCases[str,
                RegularExpression@"\\{(.*?)\\}" -> "$1"][[1]];
            Subscript[mainSymbol, subPart]
          ),
          Not[StringContainsQ[str, "_"]] && StringContainsQ[str, "^
  "],
          (
            (*no sub yes sup*)
            mainSymbol = StringSplit[str, "^"][[1]];
            mainSymbol = ToExpression[mainSymbol];

            supPart =
              StringCases[str,
                RegularExpression@"\\{(.*?)\\}" -> "$1"][[1]];
            Superscript[mainSymbol, supPart]
          ),
          StringContainsQ[str, "_"] && StringContainsQ[str, "^"],
          (
            (*yes sub yes sup*)
            mainSymbol = StringSplit[str, "_"][[1]];
            mainSymbol = ToExpression[mainSymbol];
            {subPart, supPart} =
              StringCases[str, RegularExpression@"\\{(.*?)\\}" -> "
  $1"];
            Subsuperscript[mainSymbol, subPart, supPart]
          ),
          True,
          (
            (*no sup or sub*)
            str
```

```
            )
            ];
          symbol
          ),
        {string, StringSplit[bigString, " "]}
      ];
      Which[
        form == "Row",
        Return[Row[symbols]],
        form == "List",
        Return[symbols]
      ]
    )
  ];

  FromArrayToTable::usage = "FromArrayToTable[array, labels, energies
    ] takes a square array of values and returns a table with the
    labels of the rows and columns, the energies of the initial and
    final levels, the level energies, the vacuum wavelength of the
    transition, and the value of the array. The array must be square
    and the labels and energies must be compatible with the order
    implied by the array. The array must be a square array of values.
    The function returns a list of lists with the following elements:
     - Initial level index
     - Final level index
     - Initial level label
     - Final level label
     - Initial level energy
     - Final level energy
     - Vacuum wavelength
     - Value of the array element.
  Elements in which the array is zero are not included in the return
    of this function.";
  FromArrayToTable[array_,labels_,energies_] := Module[
    {tableFun, atl},
    (
      tableFun = {
        #2[[1]],
        #2[[2]],
        labels[[#2[[1]]]],
        labels[[#2[[2]]]],
        energies[[#2[[1]]]],
        energies[[#2[[2]]]],
        If[#2[[1]]==#2[[2]],"--",10^7/(energies[[#2[[1]]]]-energies
    [[#2[[2]]]])],
        #1
      }&;
      atl = Select[Flatten[MapIndexed[tableFun,array
    ,{2}],1],#[[-1]]=!=0.&];
      atl = Append[#,1/#[[-1]]]&/@atl;
      Return[atl]
    )
  ]
  (* ########################## Misc ########################## *)
  (* ########################################################## *)

  (* ########################################################## *)
  (* ################## Some Plotting Routines ################## *)

  EnergyLevelDiagram::usage = "EnergyLevelDiagram[states] takes
    states and produces a visualization of its energy spectrum.
  The resultant visualization can be navigated by clicking and
    dragging to zoom in on a region, or by clicking and dragging
    horizontally while pressing Ctrl. Double-click to reset the view."
    ;
  Options[EnergyLevelDiagram] = {
    "Title"->"",
    "ImageSize"->1000,
    "AspectRatio" -> 1/8,
    "Background"->"Automatic",
    "Epilog"->{},
    "Explorer"->True
    };
  EnergyLevelDiagram[states_, OptionsPattern[]] := Module[
    {energies, epi, explora},
    (
```

```
4642      energies = First/@states;
4643      epi = OptionValue["Epilog"];
4644      explora = If[OptionValue["Explorer"],
4645        ExploreGraphics,
4646        Identity
4647      ];
4648      explora@ListPlot[Tooltip[{{#, 0}, {#, 1}}, {Quantity
        [#/8065.54429, "eV"], Quantity[#, 1/"Centimeters"]}] &/@ energies,
4649        Joined       -> True,
4650        PlotStyle    -> Black,
4651        AspectRatio  -> OptionValue["AspectRatio"],
4652        ImageSize    -> OptionValue["ImageSize"],
4653        Frame        -> True,
4654        PlotRange    -> {All, {0, 1}},
4655        FrameTicks   -> {{None, None}, {Automatic, Automatic}},
4656        FrameStyle   -> Directive[15, Dashed, Thin],
4657        PlotLabel    -> Style[OptionValue["Title"], 15, Bold],
4658        Background   -> OptionValue["Background"],
4659        FrameLabel   -> {"\!\(\*FractionBox[\(E\), SuperscriptBox[\(
        cm\), \(-1\)]]\)"},
4660        Epilog       -> epi]
4661    )
4662  ];
4663
4664  ExploreGraphics::usage = "Pass a Graphics object to explore it.
      Zoom by clicking and dragging a rectangle. Pan by clicking and
      dragging while pressing Ctrl. Click twice to reset view.
4665  Based on ZeitPolizei @ https://mathematica.stackexchange.com/
      questions/7142/how-to-manipulate-2d-plots.
4666  The option \"OptAxesRedraw\" can be used to specify whether the
      axes should be redrawn. The default is False.";
4667  Options[ExploreGraphics] = {OptAxesRedraw -> False};
4668  ExploreGraphics[graph_Graphics, opts : OptionsPattern[]] := With[
4669    {
4670      gr  = First[graph],
4671      opt = DeleteCases[Options[graph],
4672            PlotRange -> PlotRange | AspectRatio | AxesOrigin -> _],
4673      plr = PlotRange /. AbsoluteOptions[graph, PlotRange],
4674      ar  = AspectRatio /. AbsoluteOptions[graph, AspectRatio],
4675      ao  = AbsoluteOptions[graph, AxesOrigin],
4676      rectangle = {Dashing[Small],
4677        Line[{#1,
4678              {First[#2], Last[#1]},
4679              #2,
4680              {First[#1], Last[#2]},
4681              #1}]} &,
4682      optAxesRedraw = OptionValue[OptAxesRedraw]
4683    },
4684    DynamicModule[
4685        {dragging=False, first, second, rx1, rx2, ry1, ry2,
4686        range = plr},
4687        {{rx1, rx2}, {ry1, ry2}} = plr;
4688      Panel@
4689      EventHandler[
4690        Dynamic@Graphics[
4691          If[dragging, {gr, rectangle[first, second]}, gr],
4692          PlotRange -> Dynamic@range,
4693          AspectRatio -> ar,
4694          AxesOrigin -> If[optAxesRedraw,
4695            Dynamic@Mean[range\[Transpose]], ao],
4696          Sequence @@ opt],
4697        {{"MouseDown", 1} :> (
4698          first = MousePosition["Graphics"]
4699          ),
4700        {"MouseDragged", 1} :> (
4701          dragging = True;
4702          second = MousePosition["Graphics"]
4703          ),
4704        "MouseClicked" :> (
4705          If[CurrentValue@"MouseClickCount"==2,
4706            range = plr];
4707          ),
4708        {"MouseUp", 1} :> If[dragging,
4709          dragging = False;
4710
4711          range = {{rx1, rx2}, {ry1, ry2}} =
```

```
            Transpose@{first, second};
          range[[2]] = {0, 1}],
        {"MouseDown", 2} :> (
          first = {sx1, sy1} = MousePosition["Graphics"]
          ),
        {"MouseDragged", 2} :> (
          second = {sx2, sy2} = MousePosition["Graphics"];
          rx1 = rx1 - (sx2 - sx1);
          rx2 = rx2 - (sx2 - sx1);
          ry1 = ry1 - (sy2 - sy1);
          ry2 = ry2 - (sy2 - sy1);
          range = {{rx1, rx2}, {ry1, ry2}};
          range[[2]] = {0, 1};
          )}]]];

LabeledGrid::usage = "LabeledGrid[data, rowHeaders, columnHeaders]
  provides a grid of given data interpreted as a matrix of values
  whose rows are labeled by rowHeaders and whose columns are labeled
   by columnHeaders. When hovering with the mouse over the grid
  elements, the row and column labels are displayed with the given
  separator between them.";
Options[LabeledGrid]={
    ItemSize->Automatic,
    Alignment->Center,
    Frame->All,
    "Separator"->",",
    "Pivot"->""
};
LabeledGrid[data_,rowHeaders_,columnHeaders_,OptionsPattern[]] :=
  Module[
   {gridList=data, rowHeads=rowHeaders, colHeads=columnHeaders},
   (
     separator=OptionValue["Separator"];
     pivot=OptionValue["Pivot"];
     gridList=Table[
             Tooltip[
               data[[rowIdx,colIdx]],
               DisplayForm[
                 RowBox[{rowHeads[[rowIdx]],
                         separator,
                         colHeads[[colIdx]]}
                       ]
                       ]
                     ],
         {rowIdx,Dimensions[data][[1]]},
         {colIdx,Dimensions[data][[2]]}];
     gridList=Transpose[Prepend[gridList,colHeads]];
     rowHeads=Prepend[rowHeads,pivot];
     gridList=Prepend[gridList,rowHeads]//Transpose;
     Grid[gridList,
         Frame->OptionValue[Frame],
         Alignment->OptionValue[Alignment],
         Frame->OptionValue[Frame],
         ItemSize->OptionValue[ItemSize]
         ]
   )
];

HamiltonianForm::usage = "HamiltonianForm[hamMatrix, basisLabels]
  takes the matrix representation of a hamiltonian together with a
  set of symbols representing the ordered basis in which the
  operator is represented. With this it creates a displayed form
  that has adequately labeled row and columns together with
  informative values when hovering over the matrix elements using
  the mouse cursor.";
Options[HamiltonianForm]={"Separator"->","Pivot"->""};
HamiltonianForm[hamMatrix_, basisLabels_List, OptionsPattern[]] :=
  (
     braLabels=DisplayForm[RowBox[{"\[LeftAngleBracket]",#,"\[
  RightBracketingBar]"}]]& /@ basisLabels;
     ketLabels=DisplayForm[RowBox[{"\[LeftBracketingBar]",#,"\[
  RightAngleBracket]"}]]& /@ basisLabels;
     LabeledGrid[hamMatrix,braLabels,ketLabels,"Separator"->
  OptionValue["Separator"],"Pivot"->OptionValue["Pivot"]]
     )

```

```
4772  HamiltonianMatrixPlot::usage = "HamiltonianMatrixPlot[hamMatrix,
       basisLabels] creates a matrix plot of the given hamiltonian matrix
       with the given basis labels. The matrix elements can be hovered
       over to display the corresponding row and column labels together
       with the value of the matrix element. The option \"Overlay Values\
       " can be used to specify whether the matrix elements should be
       displayed on top of the matrix plot.";
4773  Options[HamiltonianMatrixPlot] = Join[Options[MatrixPlot], {"Hover"
       -> True, "Overlay Values" -> True}];
4774  HamiltonianMatrixPlot[hamMatrix_, basisLabels_, opts :
       OptionsPattern[]] := (
4775    braLabels = DisplayForm[RowBox[{"\[LeftAngleBracket]", #, "\[
       RightBracketingBar]"}]] & /@ basisLabels;
4776    ketLabels = DisplayForm[Rotate[RowBox[{"\[LeftBracketingBar]", #,
       "\[RightAngleBracket]"}],\[Pi]/2]] & /@ basisLabels;
4777    ketLabelsUpright = DisplayForm[RowBox[{"\[LeftBracketingBar]", #,
       "\[RightAngleBracket]"}]] & /@ basisLabels;
4778    numRows = Length[hamMatrix];
4779    numCols = Length[hamMatrix[[1]]];
4780    epiThings = Which[
4781      And[OptionValue["Hover"], Not[OptionValue["Overlay Values"]]],
4782      Flatten[
4783        Table[
4784          Tooltip[
4785            {
4786              Transparent,
4787              Rectangle[
4788              {j - 1, numRows - i},
4789              {j - 1, numRows - i} + {1, 1}
4790              ]
4791            },
4792          Row[{braLabels[[i]],ketLabelsUpright[[j]],"=",hamMatrix[[i,
       j]]}]
4793          ],
4794        {i, 1, numRows},
4795        {j, 1, numCols}
4796        ]
4797      ],
4798      And[OptionValue["Hover"], OptionValue["Overlay Values"]],
4799      Flatten[
4800        Table[
4801          Tooltip[
4802            {
4803              Transparent,
4804              Rectangle[
4805              {j - 1, numRows - i},
4806              {j - 1, numRows - i} + {1, 1}
4807              ]
4808            },
4809          DisplayForm[RowBox[{"\[LeftAngleBracket]", basisLabels[[i
       ]], "\[LeftBracketingBar]", basisLabels[[j]], "\[RightAngleBracket
       ]"}]]
4810          ],
4811        {i, numRows},
4812        {j, numCols}
4813        ]
4814      ],
4815      True,
4816      {}
4817      ];
4818    textOverlay = If[OptionValue["Overlay Values"],
4819      (
4820        Flatten[
4821        Table[
4822          Text[hamMatrix[[i, j]],
4823            {j - 1/2, numRows - i + 1/2}
4824          ],
4825        {i, 1, numRows},
4826        {j, 1, numCols}
4827        ]
4828        ]
4829      ),
4830      {}
4831      ];
4832    epiThings = Join[epiThings, textOverlay];
4833    MatrixPlot[hamMatrix,
```

```
4834        FrameTicks -> {
4835          {Transpose[{Range[Length[braLabels]], braLabels}], None},
4836          {None, Transpose[{Range[Length[ketLabels]], ketLabels}]}
4837          },
4838        Evaluate[FilterRules[{opts}, Options[MatrixPlot]]],
4839        Epilog -> epiThings
4840      ]
4841    );
4842
4843    (* ################## Some Plotting Routines ################## *)
4844    (* ############################################################ *)
4845
4846    (* ############################################################ *)
4847    (* ###################### Load Functions ###################### *)
4848
4849    LoadAll::usage = "LoadAll[] executes most Load* functions.";
4850    LoadAll[] := (
4851      LoadTermLabels[];
4852      LoadCFP[];
4853      LoadUk[];
4854      LoadV1k[];
4855      LoadT22[];
4856      LoadSOOandECSOLS[];
4857
4858      LoadElectrostatic[];
4859      LoadSpinOrbit[];
4860      LoadSOOandECSO[];
4861      LoadSpinSpin[];
4862      LoadThreeBody[];
4863      LoadChenDeltas[];
4864      LoadCarnall[];
4865    );
4866
4867    fnTermLabels::usage = "This list contains the labels of f^n
       configurations. Each element of the list has four elements {LS,
       seniority, W, U}. At first sight this seems to only include the
       labels for the f^6 and f^7 configuration, however, all is included
        in these two.";
4868
4869    LoadTermLabels::usage = "LoadTermLabels[] loads into the session
       the labels for the terms in the f^n configurations.";
4870    LoadTermLabels[] := (
4871      If[ValueQ[fnTermLabels], Return[]];
4872      PrintTemporary["Loading data for state labels in the f^n
       configurations..."];
4873      fnTermsFname = FileNameJoin[{moduleDir, "data", "fnTerms.m"}];
4874
4875      If[!FileExistsQ[fnTermsFname],
4876        (PrintTemporary[">> fnTerms.m not found, generating ..."];
4877          fnTermLabels = ParseTermLabels["Export"->True];
4878        ),
4879        fnTermLabels = Import[fnTermsFname];
4880      ];
4881    );
4882
4883    Carnall::usage = "Association of data from Carnall et al (1989)
       with the following keys: {data, annotations, paramSymbols,
       elementNames, rawData, rawAnnotations, annnotatedData, appendix:Pr
       :Association, appendix:Pr:Calculated, appendix:Pr:RawTable,
       appendix:Headings}";
4884
4885    LoadCarnall::usage = "LoadCarnall[] loads data for trivalent
       lanthanides in LaF3 using the data from Bill Carnall's 1989 paper.
       ";
4886    LoadCarnall[] := (
4887      If[ValueQ[Carnall], Return[]];
4888      carnallFname = FileNameJoin[{moduleDir, "data", "Carnall.m"}];
4889      If[!FileExistsQ[carnallFname],
4890        (PrintTemporary[">> Carnall.m not found, generating ..."];
4891          Carnall = ParseCarnall[];
4892        ),
4893        Carnall = Import[carnallFname];
4894      ];
4895    );
4896
4897    LoadChenDeltas::usage = "LoadChenDeltas[] loads the differences
```

```
  noted by Chen.";
LoadChenDeltas[] := (
  If[ValueQ[chenDeltas], Return[]];
  PrintTemporary["Loading the association of discrepancies found by
   Chen ..."];
  chenDeltasFname = FileNameJoin[{moduleDir, "data", "chenDeltas.m"
  }];
  If[!FileExistsQ[chenDeltasFname],
    (PrintTemporary[">> chenDeltas.m not found, generating ..."];
      chenDeltas = ParseChenDeltas[];
    ),
    chenDeltas = Import[chenDeltasFname];
  ];
);

ParseChenDeltas::usage = "ParseChenDeltas[] parses the data found
  in ./data/the-chen-deltas-A.csv and ./data/the-chen-deltas-B.csv.
  If the option \"Export\" is set to True (True is the default),
  then the parsed data is saved to ./data/chenDeltas.m";
Options[ParseChenDeltas] = {"Export" -> True};
ParseChenDeltas[OptionsPattern[]] := (
  chenDeltasRaw = Import[FileNameJoin[{moduleDir, "data", "the-chen
  -deltas-A.csv"}]];
  chenDeltasRaw = chenDeltasRaw[[2 ;;]];
  chenDeltas = <||>;
  chenDeltasA = <||>;
  Off[Power::infy];
  Do[
    ({right, wrong} = {chenDeltasRaw[[row]][[4 ;;]],
      chenDeltasRaw[[row + 1]][[4 ;;]]};
    key = chenDeltasRaw[[row]][[1 ;; 3]];
    repRule = (#[[1]] -> #[[2]]*#[[1]]) & /@
      Transpose[{{M0, M2, M4, P2, P4, P6}, right/wrong}];
    chenDeltasA[key] = <|"right" -> right, "wrong" -> wrong,
      "repRule" -> repRule|>;
    chenDeltasA[{key[[1]], key[[3]], key[[2]]}] = <|"right" ->
  right,
      "wrong" -> wrong, "repRule" -> repRule|>;
    ),
    {row, 1, Length[chenDeltasRaw], 2}];
  chenDeltas["A"] = chenDeltasA;

  chenDeltasRawB = Import[FileNameJoin[{moduleDir, "data", "the-
  chen-deltas-B.csv"}], "Text"];
  chenDeltasB = StringSplit[chenDeltasRawB, "\n"];
  chenDeltasB = StringSplit[#, ","] & /@ chenDeltasB;
  chenDeltasB = {ToExpression[StringTake[#[[1]], {2}]], #[[2]],
  #[[3]]} & /@ chenDeltasB;
  chenDeltas["B"] = chenDeltasB;
  On[Power::infy];
  If[OptionValue["Export"],
    (chenDeltasFname = FileNameJoin[{moduleDir, "data", "chenDeltas
  .m"}];
    Export[chenDeltasFname, chenDeltas];
    )
    ];
  Return[chenDeltas];
);

ParseCarnall::usage = "ParseCarnall[] parses the data found in ./
  data/Carnall.xls. If the option \"Export\" is set to True (True is
   the default), then the parsed data is saved to ./data/Carnall.
  This data is from the tables and appendices of Carnall et al
  (1989).";
Options[ParseCarnall] = {"Export" -> True};
ParseCarnall[OptionsPattern[]] := (
  ions          = {"Ce","Pr","Nd","Pm","Sm","Eu","Gd","Tb","Dy","Ho"
  ,"Er","Tm","Yb"};
  templates     = StringTemplate/@StringSplit["appendix:`ion`:
  Association appendix:`ion`:Calculated appendix:`ion`:RawTable
  appendix:`ion`:Headings","  "];

  (* How many unique eigenvalues, after removing Kramer's
  degeneracy *)
  fullSizes     = AssociationThread[ions, {7, 91, 182, 1001, 1001,
  3003, 1716, 3003, 1001, 1001, 182, 91, 7}];
```

```
    carnall      = Import[FileNameJoin[{moduleDir,"data","Carnall.xls"}]][[2]];
    carnallErr   = Import[FileNameJoin[{moduleDir,"data","Carnall.xls"}]][[3]];

    elementNames = carnall[[1]][[2;;]];
    carnall      = carnall[[2;;]];
    carnallErr   = carnallErr[[2;;]];
    carnall      = Transpose[carnall];
    carnallErr   = Transpose[carnallErr];
    paramNames   = ToExpression/@carnall[[1]][[1;;]];
    carnall      = carnall[[2;;]];
    carnallErr   = carnallErr[[2;;]];
    carnallData  = Table[(
                    data         = carnall[[i]];
                    data         = (#[[1]]->#[[2]])&/@Select[Transpose[{paramNames,data}],#[[2]]!=""&];
                    elementNames[[i]]->data
                    ),
                    {i,1,13}
                    ];
    carnallData  = Association[carnallData];
    carnallNotes = Table[(
                    data        = carnallErr[[i]];
                    elementName = elementNames[[i]];
                    dataFun     = (
                        #[[1]] -> If[#[[2]]=="[]",
                        "Not allowed to vary in fitting.",
                        If[#[[2]]=="[R]",
                            "Ratio constrained by: " <> <|"Eu"->"F4/F2=0.713; F6/F2=0.512",
                                "Gd"->"F4/F2=0.710]",
                                "Tb"->"F4/F2=0.707"|>[elementName],
                            If[#[[2]]=="i",
                                "Interpolated",
                                #[[2]]
                            ]
                        ]
                        ]) &;
                    data = dataFun /@ Select[Transpose[{paramNames,data}],#[[2]]!=""&];
                    elementName->data
                      ),
                {i,1,13}
                ];
    carnallNotes  = Association[carnallNotes];

    annotatedData = Table[
                    If[NumberQ[#[[1]]],Tooltip[#[[1]],#[[2]]],""] & /@ Transpose[{paramNames/.carnallData[element],
                      paramNames/.carnallNotes[element]
                        }],
                    {element,elementNames}
                    ];
    annotatedData = Transpose[annotatedData];

    Carnall = <|"data"      -> carnallData,
        "annotations"       -> carnallNotes,
        "paramSymbols"      -> paramNames,
        "elementNames"      -> elementNames,
        "rawData"           -> carnall,
        "rawAnnotations"    -> carnallErr,
        "includedTableIons" -> ions,
        "annnotatedData"    -> annotatedData
    |>;

    Do[(
        carnallData   = Import[FileNameJoin[{moduleDir,"data","Carnall.xls"}]][[sheetIdx]];
        headers       = carnallData[[1]];
        calcIndex     = Position[headers,"Calc (1/cm)"][[1,1]];
        headers       = headers[[2;;]];
        carnallLabels = carnallData[[1]];
        carnallData   = carnallData[[2;;]];
        carnallTerms  = DeleteDuplicates[First/@carnallData];
        parsedData    = Table[(
```

```
                        rows = Select[carnallData,#[[1]]==term&];
                        rows = #[[2;;]]&/@rows;
                        rows = Transpose[rows];
                        rows = Transpose[{headers,rows}];
                        rows = Association[(#[[1]]->#[[2]])&/@rows
];
                        term->rows
                        ),
                    {term,carnallTerms}
                    ];
      carnallAssoc        = Association[parsedData];
      carnallCalcEnergies = #[[calcIndex]]&/@carnallData;
      carnallCalcEnergies = If[NumberQ[#],#,Missing[]]&/
@carnallCalcEnergies;
      ion                 = ions[[sheetIdx-3]];
      carnallCalcEnergies = PadRight[carnallCalcEnergies, fullSizes
[ion], Missing[]];
      keys                = #[<|"ion"->ion|>]&/@templates;
      Carnall[keys[[1]]]  = carnallAssoc;
      Carnall[keys[[2]]]  = carnallCalcEnergies;
      Carnall[keys[[3]]]  = carnallData;
      Carnall[keys[[4]]]  = headers;
      ),
    {sheetIdx,4,16}
    ];

    goodions = Select[ions,#!="Pm"&];
    expData  = Select[Transpose[Carnall["appendix:"<>#<>":RawTable"
    ]][[1+Position[Carnall["appendix:"<>#<>":Headings"],"Exp (1/cm)"
    ]][[1,1]]]],NumberQ]&/@goodions;
    Carnall["All Experimental Data"]=AssociationThread[goodions,
    expData];
    If[OptionValue["Export"],
      (
        carnallFname = FileNameJoin[{moduleDir, "data", "Carnall.m"
    }];
        Print["Exporting to "<>carnallFname];
        Export[carnallFname, Carnall];
      )
      ];
    Return[Carnall];
  );

  CFP::usage = "CFP[{n, NKSL}] provides a list whose first element
    echoes NKSL and whose other elements are lists with two elements
    the first one being the symbol of a parent term and the second
    being the corresponding coefficient of fractional parentage. n
    must satisfy 1 <= n <= 7.
  These are according to the tables from Nielson & Koster.";

  CFPAssoc::usage = " CFPAssoc is an association where keys are of
    lists of the form {num_electrons, daugherTerm, parentTerm} and
    values are the corresponding coefficients of fractional parentage.
     The terms given in string-spectroscopic notation. If a certain
    daughter term does not have a parent term, the value is 0. Loaded
    using LoadCFP[].
  These are according to the tables from Nielson & Koster.";

  LoadCFP::usage = "LoadCFP[] loads CFP, CFPAssoc, and CFPTable into
    the session.";
  LoadCFP[] := (
    If[And[ValueQ[CFP], ValueQ[CFPTable], ValueQ[CFPAssoc]],Return
    []];

    PrintTemporary["Loading CFPtable ..."];
    CFPTablefname = FileNameJoin[{moduleDir, "data", "CFPTable.m"}];
    If[!FileExistsQ[CFPTablefname],
      (PrintTemporary[">> CFPTable.m not found, generating ..."];
        CFPTable = GenerateCFPTable["Export"->True];
      ),
      CFPTable = Import[CFPTablefname];
    ];

    PrintTemporary["Loading CFPs.m ..."];
    CFPfname = FileNameJoin[{moduleDir, "data", "CFPs.m"}];
    If[!FileExistsQ[CFPfname],
```

```
5081        (PrintTemporary[">> CFPs.m not found, generating ..."];
5082          CFP = GenerateCFP["Export"->True];
5083        ),
5084        CFP = Import[CFPfname];
5085      ];
5086
5087      PrintTemporary["Loading CFPAssoc.m ..."];
5088      CFPAfname = FileNameJoin[{moduleDir, "data", "CFPAssoc.m"}];
5089      If[!FileExistsQ[CFPAfname],
5090        (PrintTemporary[">> CFPAssoc.m not found, generating ..."];
5091          CFPAssoc = GenerateCFPAssoc["Export"->True];
5092        ),
5093        CFPAssoc = Import[CFPAfname];
5094      ];
5095    );
5096
5097    ReducedUkTable::usage = "ReducedUkTable[{n, l = 3, SL, SpLp, k}]
       provides reduced matrix elements of the unit spherical tensor
       operator Uk. See TASS section 11-9 \"Unit Tensor Operators\".
       Loaded using LoadUk[].";
5098
5099    LoadUk::usage = "LoadUk[] loads into session the reduced matrix
       elements for unit tensor operators.";
5100    LoadUk[] := (
5101      If[ValueQ[ReducedUkTable], Return[]];
5102      PrintTemporary["Loading the association of reduced matrix
       elements for unit tensor operators ..."];
5103      ReducedUkTableFname = FileNameJoin[{moduleDir, "data", "
       ReducedUkTable.m"}];
5104      If[!FileExistsQ[ReducedUkTableFname],
5105        (PrintTemporary[">> ReducedUkTable.m not found, generating ..."
       ];
5106          ReducedUkTable = GenerateReducedUkTable[7];
5107        ),
5108        ReducedUkTable = Import[ReducedUkTableFname];
5109      ];
5110    );
5111
5112    ElectrostaticTable::usage = "ElectrostaticTable[{n, SL, SpLp}]
       provides the calculated result of Electrostatic[{n, SL, SpLp}].
       Load using LoadElectrostatic[].";
5113
5114    LoadElectrostatic::usage = "LoadElectrostatic[] loads the reduced
       matrix elements for the electrostatic interaction.";
5115    LoadElectrostatic[] := (
5116      If[ValueQ[ElectrostaticTable], Return[]];
5117      PrintTemporary["Loading the association of matrix elements for
       the electrostatic interaction ..."];
5118      ElectrostaticTablefname = FileNameJoin[{moduleDir, "data", "
       ElectrostaticTable.m"}];
5119      If[!FileExistsQ[ElectrostaticTablefname],
5120        (PrintTemporary[">> ElectrostaticTable.m not found, generating
       ..."];
5121          ElectrostaticTable = GenerateElectrostaticTable[7];
5122        ),
5123        ElectrostaticTable = Import[ElectrostaticTablefname];
5124      ];
5125    );
5126
5127    LoadV1k::usage = "LoadV1k[] loads into session the matrix elements
       of V1k.";
5128    LoadV1k[] := (
5129      If[ValueQ[ReducedV1kTable], Return[]];
5130      PrintTemporary["Loading the association of matrix elements for
       V1k ..."];
5131      ReducedV1kTableFname = FileNameJoin[{moduleDir, "data", "
       ReducedV1kTable.m"}];
5132      If[!FileExistsQ[ReducedV1kTableFname],
5133        (PrintTemporary[">> ReducedV1kTable.m not found, generating ...
       "];
5134          ReducedV1kTable = GenerateReducedV1kTable[7];
5135        ),
5136        ReducedV1kTable = Import[ReducedV1kTableFname];
5137      ]
5138    );
5139
```

```
LoadSpinOrbit::usage = "LoadSpinOrbit[] loads into session the matrix elements of the spin-orbit interaction.";
LoadSpinOrbit[] := (
  If[ValueQ[SpinOrbitTable], Return[]];
  PrintTemporary["Loading the association of matrix elements for spin-orbit ..."];
  SpinOrbitTableFname = FileNameJoin[{moduleDir, "data", "SpinOrbitTable.m"}];
  If[!FileExistsQ[SpinOrbitTableFname],
    (
      PrintTemporary[">> SpinOrbitTable.m not found, generating ..."];
      SpinOrbitTable = GenerateSpinOrbitTable[7, "Export" -> True];
    ),
    SpinOrbitTable = Import[SpinOrbitTableFname];
  ]
);

LoadSOOandECSOLS::usage = "LoadSOOandECSOLS[] loads into session the LS reduced matrix elements of the SOO-ECSO interaction.";
LoadSOOandECSOLS[] := (
  If[ValueQ[SOOandECSOLSTable], Return[]];
  PrintTemporary["Loading the association of LS reduced matrix elements for SOO-ECSO ..."];
  SOOandECSOLSTableFname = FileNameJoin[{moduleDir, "data", "ReducedSOOandECSOLSTable.m"}];
  If[!FileExistsQ[SOOandECSOLSTableFname],
    (PrintTemporary[">> ReducedSOOandECSOLSTable.m not found, generating ..."];
      SOOandECSOLSTable = GenerateSOOandECSOLSTable[7];
    ),
    SOOandECSOLSTable = Import[SOOandECSOLSTableFname];
  ];
);

LoadSOOandECSO::usage = "LoadSOOandECSO[] loads into session the LSJ reduced matrix elements of spin-other-orbit and electrostatically-correlated-spin-orbit.";
LoadSOOandECSO[] := (
  If[ValueQ[SOOandECSOTableFname], Return[]];
  PrintTemporary["Loading the association of matrix elements for spin-other-orbit and electrostatically-correlated-spin-orbit ..."];
  SOOandECSOTableFname = FileNameJoin[{moduleDir, "data", "SOOandECSOTable.m"}];
  If[!FileExistsQ[SOOandECSOTableFname],
    (PrintTemporary[">> SOOandECSOTable.m not found, generating ..."];
      SOOandECSOTable = GenerateSOOandECSOTable[7, "Export"->True];
    ),
    SOOandECSOTable = Import[SOOandECSOTableFname];
  ];
);

LoadT22::usage = "LoadT22[] loads into session the matrix elements of T22.";
LoadT22[] := (
  If[ValueQ[T22Table], Return[]];
  PrintTemporary["Loading the association of reduced T22 matrix elements ..."];
  T22TableFname = FileNameJoin[{moduleDir, "data", "ReducedT22Table.m"}];
  If[!FileExistsQ[T22TableFname],
    (PrintTemporary[">> ReducedT22Table.m not found, generating ..."];
      T22Table = GenerateT22Table[7];
    ),
    T22Table = Import[T22TableFname];
  ];
);

LoadSpinSpin::usage = "LoadSpinSpin[] loads into session the matrix elements of spin-spin.";
LoadSpinSpin[] := (
  If[ValueQ[SpinSpinTable], Return[]];
  PrintTemporary["Loading the association of matrix elements for
```

```
    spin-spin ..."];
5197     SpinSpinTableFname = FileNameJoin[{moduleDir, "data", "
    SpinSpinTable.m"}];
5198     If[!FileExistsQ[SpinSpinTableFname],
5199       (PrintTemporary[">> SpinSpinTable.m not found, generating ..."
    ];
5200         SpinSpinTable = GenerateSpinSpinTable[7, "Export" -> True];
5201       ),
5202       SpinSpinTable = Import[SpinSpinTableFname];
5203     ];
5204   );
5205
5206   LoadThreeBody::usage = "LoadThreeBody[] loads into session the
      matrix elements of three-body configuration-interaction effects.";
5207   LoadThreeBody[] := (
5208     If[ValueQ[ThreeBodyTable], Return[]];
5209     PrintTemporary["Loading the association of matrix elements for
    three-body configuration-interaction effects ..."];
5210     ThreeBodyFname   = FileNameJoin[{moduleDir, "data", "
    ThreeBodyTable.m"}];
5211     ThreeBodiesFname = FileNameJoin[{moduleDir, "data", "
    ThreeBodyTables.m"}];
5212     If[!FileExistsQ[ThreeBodyFname],
5213       (PrintTemporary[">> ThreeBodyTable.m not found, generating ..."
    ];
5214         {ThreeBodyTable, ThreeBodyTables} = GenerateThreeBodyTables
    [14, "Export" -> True];
5215       ),
5216       ThreeBodyTable  = Import[ThreeBodyFname];
5217       ThreeBodyTables = Import[ThreeBodiesFname];
5218     ];
5219   );
5220
5221   (* ###################### Load Functions ###################### *)
5222   (* ########################################################### *)
5223
5224 End[];
5225
5226 LoadTermLabels[];
5227 LoadCFP[];
5228
5229 EndPackage[];
```

17.2 fittings.m

This file has code useful for fitting the Hamiltonian.

```
1  (*-------------------------------------------------------------------+
2  |~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~|
3  |~~~+---------------------------------------------------------+~~~|
4  |~~~|                 _____ __  __  _                         |~~~|
5  |~~~|               / __(_) /_/ /_(_)___  ____ ______          |~~~|
6  |~~~|              / /_/ / __/ __/ / __ \/ __ `/ ___/          |~~~|
7  |~~~|             / __/ / /_/ /_/ / / / / /_/ (__  )           |~~~|
8  |~~~|            /_/ /_/\__/\__/_/_/ /_/\__, /____/            |~~~|
9  |~~~|                                  /____/                 |~~~|
10 |~~~|                                                         |~~~|
11 |~~~+---------------------------------------------------------+~~~|
12 |~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~|
13 |~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~|
14 |~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~|
15 |~~~~+-----------------------------------------------------+~~~~|
16 |~~~~|                                                     |~~~~|
17 |~~~~|                                                     |~~~~|
18 |~~~~| This script puts together some code useful for fitting |~~~~|
19 |~~~~|             the model Hamiltonian to data.          |~~~~|
20 |~~~~|                                                     |~~~~|
21 |~~~~|                                                     |~~~~|
22 |~~~~+-----------------------------------------------------+~~~~|
23 |~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~|
24 |~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~|
25 +-------------------------------------------------------------------*)
26
27 Get["qlanth.m"]
28 Get["qonstants.m"];
29 Get["misc.m"];
```

```
LoadCarnall[];
LoadFreeIon[];

Jiggle::usage = "Jiggle[num, wiggleRoom] takes a number and randomizes it a little by adding or subtracting a random fraction of itself. The fraction is controlled by wiggleRoom.";
Jiggle[num_, wiggleRoom_ : 0.1] := RandomReal[{1 - wiggleRoom, 1 + wiggleRoom}] * num;

AddToList::usage = "AddToList[list, element, maxSize, addOnlyNew] prepends the element to list and returns the list. If maxSize is reached, the last element is dropped. If addOnlyNew is True (the default), the element is only added if it is different from the last element.";
AddToList[list_, element_, maxSize_, addOnlyNew_ : True] := Module[{
    tempList = If[
      addOnlyNew,
      If[
       Length[list] == 0,
       {element},
       If[
        element != list[[-1]],
        Append[list, element],
        list
        ]
       ],
      Append[list, element]
      ]},
   If[Length[tempList] > maxSize,
    Drop[tempList, Length[tempList] - maxSize],
    tempList]
   ];

ProgressNotebook::usage="ProgressNotebook[] creates a progress notebook for the solver. This notebook includes a plot of the RMS history and the current parameter values. The notebook is returned. The RMS history and the parameter values are updated by setting the variables rmsHistory and paramSols. The variables stringPartialVars and paramSols are used to display the parameter values in the notebook.";
Options[ProgressNotebook] = {
    "Basic" -> True,
    "UpdateInterval" -> 0.5};
ProgressNotebook[OptionsPattern[]] := (
   nb = Which[
   OptionValue["Basic"],
   CreateDocument[(
      {
       Dynamic[
        TextCell[
         If[
          Length[paramSols] > 0,
          TableForm[
            Prepend[
              Transpose[{stringPartialVars,
              paramSols[[-1]]}],
              {"RMS",rmsHistory[[-1]]}]
              ],
          ""
          ],
         "Output"
         ],
        TrackedSymbols :> {paramSols, stringPartialVars},
        UpdateInterval -> OptionValue["UpdateInterval"]
        ]
       }
      ),
     WindowSize -> {600, 1000},
     WindowSelected -> True,
     TextAlignment -> Center,
     WindowTitle -> "Solver Progress"
     ],
   True,
   CreateDocument[(
      {
        "",
```

```
93          Dynamic[Framed[progressMessage],
94            UpdateInterval -> OptionValue["UpdateInterval"]],
95          Dynamic[
96          GraphicsColumn[
97           {ListPlot[rmsHistory,
98             PlotMarkers -> "OpenMarkers",
99             Frame -> True,
100            FrameLabel -> {"Iteration", "RMS"},
101            ImageSize -> 800,
102            AspectRatio -> 1/3,
103            FrameStyle -> Directive[Thick, 15],
104            PlotLabel -> If[Length[rmsHistory] != 0, rmsHistory[[-1]],
     ""]
105           ],
106          ListPlot[(#/#[[1]]) & /@ Transpose[paramSols],
107           Joined -> True,
108           PlotRange -> {All, {-5, 5}},
109           Frame -> True,
110           ImageSize -> 800,
111           AspectRatio -> 1,
112           FrameStyle -> Directive[Thick, 15],
113           FrameLabel -> {"Iteration", "Params"}
114           ]
115          }
116         ],
117         TrackedSymbols :> {rmsHistory, paramSols},
118         UpdateInterval -> OptionValue["UpdateInterval"]
119         ],
120        Dynamic[
121         TextCell[
122          If[
123           Length[paramSols] > 0,
124           TableForm[Transpose[{stringPartialVars, paramSols[[-1]]}]],
125           ""
126           ],
127          "Output"
128          ],
129         TrackedSymbols :> {paramSols, stringPartialVars}
130         ]
131        }
132       ),
133      WindowSize -> {600, 1000},
134      WindowSelected -> True,
135      TextAlignment -> Center,
136      WindowTitle -> "Solver Progress"
137      ]
138    ];
139    Return[nb];
140    );
141
142 energyCostFunTemplate::usage="energyCostFunTemplate is template used
       to define the cost function for the energy matching. The template
       is used to define a function TheRightEnergyPath that takes a list
       of variables and returns the RMS of the energy differences between
        the computed and the experimental energies. The template requires
        the values to the following keys to be provided: `vars` and `
       varPatterns`";
143 energyCostFunTemplate = StringTemplate["
144 TheRightEnergyPath[`varPatterns`]:= (
145   {eigenEnergies, eigenVecs} = Eigensystem[compHam[`vars`]];
146   ordering      = Ordering[eigenEnergies];
147   eigenEnergies = eigenEnergies - Min[eigenEnergies];
148   states        = Transpose[{eigenEnergies, eigenVecs}];
149   states        = states[[ordering]];
150   coarseStates  = ParseStates[states, basis];
151   coarseStates  = {#[[1]],#[[-1]]}& /@ coarseStates;
152   (* The eigenvectors need to be simplified in order to compare
        labels to labels *)
153   missingLevels = Length[coarseStates]-Length[expData];
154   (* The energies are in the first element of the tuples. *)
155   energyDiffFun = (Abs[#1[[1]]-#2[[1]]])&;
156   (* match disregarding labels *)
157   energyFlow    = FlowMatching[coarseStates,
158                   expData,
159                   \"notMatched\" -> missingLevels,
160                   \"CostFun\"    -> energyDiffFun
```

```
                    ];
  energyPairs   = {#[[1]][[1]], #[[2]][[1]]} & /@ energyFlow[[1]];
  energyRms     = Sqrt[Total[(Abs[#[[2]]-#[[1]]])^2 & /@ energyPairs]
    / Length[energyPairs]];
  Return[energyRms];
)"];

AppendToLog[message_, file_String] := Module[
  {timestamp = DateString["ISODateTime"], msgString},
  (
    msgString = ToString[message, InputForm]; (* Convert any
    expression to a string *)
    OpenAppend[file];
    WriteString[file, timestamp, " - ", msgString, "\n"];
    Close[file];
  )
  ];

energyAndLabelCostFunTemplate::usage="energyAndLabelCostFunTemplate
    is a template used to define the cost function that includes both
    the differences between energies and the differences between
    labels. The template is used to define a function
    TheRightSignedPath that takes a list of variables and returns the
    RMS of the energy differences between the computed and the
    experimental energies together with a term that depends on the
    differences between the labels. The template requires the values
    to the following keys to be provided: `vars` and `varPatterns`";
energyAndLabelCostFunTemplate = StringTemplate["
TheRightSignedPath[`varPatterns`] := Module[
  {energyRms, eigenEnergies, eigenVecs, ordering, states,
    coarseStates, missingLevels, energyDiffFun, energyFlow,
    energyPairs, energyAndLabelFun, energyAndLabelFlow, totalAvgCost},
  (
    {eigenEnergies, eigenVecs} = Eigensystem[compHam[`vars`]];
    ordering      = Ordering[eigenEnergies];
    eigenEnergies = eigenEnergies - Min[eigenEnergies];
    states        = Transpose[{eigenEnergies, eigenVecs}];
    states        = states[[ordering]];
    coarseStates  = ParseStates[states, basis];

    (* The eigenvectors need to be simplified in order to compare
    labels to labels *)
    coarseStates  = {#[[1]],#[[-1]]}& /@ coarseStates;
    missingLevels = Length[coarseStates]-Length[expData];

    (* The energies are in the first element of the tuples. *)
    energyDiffFun = ( Abs[#1[[1]]-#2[[1]]] ) &;

    (* matching disregarding labels to get overall scale for scaling
    differences in labels *)
    energyFlow    = FlowMatching[coarseStates,
                    expData,
                    \"notMatched\" -> missingLevels,
                    \"CostFun\"    -> energyDiffFun
                    ];
    energyPairs   = {#[[1]][[1]], #[[2]][[1]]}&/@energyFlow[[1]];
    energyRms     = Sqrt[Total[(Abs[#[[2]]-#[[1]]])^2 & /@
    energyPairs]/Length[energyPairs]];

    (* matching using both labels and energies  *)
    energyAndLabelFun  = With[{del=energyRms},
                (Abs[#1[[1]]-#2[[1]]] +
                If[#1[[2]]==#2[[2]],
                  0.,
                  del])&];

    (* energyAndLabelFun  = With[{del=energyRms},
                (Abs[#1[[1]]-#2[[1]]] +
                del*EditDistance[#1[[2]],#2[[2]]])&]; *)
    energyAndLabelFun  = ( Abs[#1[[1]] - #2[[1]]] + EditDistance
    [#1[[2]],#2[[2]]] )&;
    energyAndLabelFlow = FlowMatching[coarseStates,
                         expData,
                         \"notMatched\" -> missingLevels,
                         \"CostFun\"    -> energyAndLabelFun
                         ];
```

```
    totalAvgCost       = Total[energyAndLabelFun@@# & /@
    energyAndLabelFlow[[1]]]/Length[energyAndLabelFlow[[1]]];
    Return[totalAvgCost];
  )
]"];

truncatedEnergyCostTemplate = StringTemplate["
TheTruncatedAndSignedPath[`varsWithNumericQ`] :=
(
  (* Calculate the truncated Hamiltonian *)
  numericalFreeIonHam = compileIntermediateTruncatedHam[`
    varsMixedWithFixedVals`];

  (* Diagonalize it *)
  {truncatedEigenvalues, truncatedEigenVectors} = Eigensystem[
    numericalFreeIonHam];

  (* Using the truncated eigenvectors push them up to the full state
    space *)
  pulledTruncatedEigenVectors = truncatedEigenVectors.Transpose[
    truncatedIntermediateBasis];
  states = Transpose[{truncatedEigenvalues,
    pulledTruncatedEigenVectors}];
  states = SortBy[states, First];
  states = ShiftedLevels[states];

  (* Coarsen the resulting eigenstates *)
  coarseStates = ParseStates[states, basis];

  (* Grab the parts that are needed for fitting *)
  coarseStates = {#[[1]], #[[-1]]} & /@ coarseStates;

  (* This cost function takes into account both labels and energies a
     random factor is added for the sake of stability of the solver*)
  energyAndLabelFun = (
        (
          Abs[#1[[1]] - #2[[1]]] +
          EditDistance[#1[[2]], #2[[2]]]
        ) *
        (1 + RandomReal[{-10^-6, 10^-6}])) &;

  (* This one only takes into account the energies *)
  energyFun = (Abs[#1[[1]] - #2[[1]]]*(1 + RandomReal[{0, 10^-6}]) &)
    ;

  (* Choose which cost function to use *)
  costFun = energyAndLabelFun;

  (* Not all states are to be matched to the experimental data *)
  missingLevels = Length[coarseStates] - Length[expData];

  (* If there are more experimental data than calculated ones, don't
    leave any state unmatched to those*)
  missingLevels = If[missingLevels < 0, 0, missingLevels];

  (* Apply the Hungarian algorithm to match the two sets of data *)
  energyAndLabelFlow = FlowMatching[coarseStates,
    expData,
    \"notMatched\" -> missingLevels,
    \"CostFun\" -> costFun];
  totalCosts   = (costFun @@ #)& /@ energyAndLabelFlow[[1]];
  totalAvgCost = Total[totalCosts] / Length[energyAndLabelFlow[[1]]];
  Return[totalAvgCost]
)"
];

Constrainer::usage = "Constrainer[problemVars, ln] returns a list of
    constraints for the variables in problemVars for trivalent
    lanthanide ion ln. problemVars are standard model symbols (F2, F4,
     ...). The ranges returned are based in the fitted parameters for
    LaF3 as found in Carnall et al. They could probably be more fine
    grained, but these ranges are seen to describe all the ions in
    that case.";
Constrainer[problemVars_, ln_] := (
  slater = Which[
    MemberQ[{"Ce", "Yb"}, ln],
```

```
    {},
    True,
    {#, (20000. < # < 120000.)} & /@ {F2, F4, F6}
    ];
  alpha = Which[
    MemberQ[{"Ce", "Yb"}, ln],
    {},
    True,
    {{α, 14. < α < 22.}}
    ];
  zeta = {{ζ, 500. < ζ < 3200.}};
  beta = Which[
    MemberQ[{"Ce", "Yb"}, ln],
    {},
    True,
    {{β, -1000. < β < -400.}}
    ];
  gamma = Which[
    MemberQ[{"Ce", "Yb"}, ln],
    {},
    True,
    {{γ, 1000. < γ < 2000.}}
    ];
  tees = Which[
    ln == "Tm",
    {100. < T2 < 500.},
    MemberQ[{"Ce", "Pr", "Yb"}, ln],
    {},
    True,
    {#, -500. < # < 500.} & /@ {T2, T3, T4, T6, T7, T8}];
  marvins = Which[
    MemberQ[{"Ce", "Yb"}, ln],
    {},
    True,
    {{M0, 1.0 < M0 < 5.0}}
    ];
  peas = Which[
    MemberQ[{"Ce", "Yb"}, ln],
    {},
    True,
    {{P2, -200. < P2 < 1200.}}
    ];
  crystalRanges = {#, (-2000. < # < 2000.)} & /@ (Intersection[
      cfSymbols, problemVars]);
  allCons =
   Join[slater, zeta, alpha, beta, gamma, tees, marvins, peas,
    crystalRanges];
  allCons = Select[allCons, MemberQ[problemVars, #[[1]]] &];
  Return[Flatten[Rest /@ allCons]]
  )

Options[LogSol] = {"PrintFun" -> PrintTemporary};
LogSol::usage = "LogSol[expr, solHistory, prefix] saves the given
    expression to a file. The file is named with the given prefix and
    a created UUID. The file is saved in the \"log\" directory under
    the current directory. The file is saved in the format of a .m
    file. The function returns the name of the file.";
LogSol[theSolution_, prefix_, OptionsPattern[]] := (
  PrintFun = OptionValue["PrintFun"];
  fname = prefix <> "-sols-" <> CreateUUID[] <> ".m";
  fname = FileNameJoin[{".", "log", fname}];
  PrintFun["Saving solution to: ", fname];
  Export[fname, theSolution];
  Return[fname];
  );


FitToHam::usage = "FitToHam[numE, expData, fitToSymbols, simplifier,
    OptionsPattern[]] fits the model Hamiltonian to the experimental
    data for the trivalent lanthanide ion with number numE. The
    experimental data is given in the form of a list of tuples. The
    first element of the tuple is the energy and the second element is
     the label. The function saves the results to a file, with the
    string filePrefix prepended to it, by default this is an empty
    string, in which case the filePrefix is modified to be the name of
     the lanthanide.
```

```
346 The fitToSymbols is a list of the symbols to be fit. The simplifier
        is a list of rules that simplify the Hamiltonian.
347 The options and their defaults are:
348 \"PrintFun\"->PrintTemporary,
349 \"FilePrefix\"->\"\",
350 \"SlackChannel\"->None,
351 \"MaxHistory\"->100,
352 \"MaxIters\"->100,
353 \"NumCycles\"->10,
354 \"ProgressWindow\"->True
355 The PrintFun option is the function used to print progress messages.
356 The FilePrefix option is the prefix to use for the file name, by
        default this is the symbol for the lanthanide.
357 The SlackChannel option is the channel to post progress messages to.
358 The MaxHistory option is the maximum number of iterations to keep in
        the history.
359 The MaxIters option is the maximum number of iterations for the
        solver.
360 The NumCycles option is the number of cycles to run the solver for.
361 The function returns a list of solutions. The solutions are the
        results of the NMinimize function. The solutions are a list of
        tuples. The first element of the tuple is the RMS error and the
        second element is the parameter values
362 The function also saves the solutions to a file. The file is named
        with a prefix and a UUID. The file is saved in the current
        directory. The file is saved in the format of a .m file.";
363 Options[FitToHam] = {
364       "PrintFun"-> PrintTemporary,
365       "FilePrefix" -> "",
366       "SlackChannel" -> None,
367       "MaxHistory" -> 100,
368       "ProgressWindow"-> True,
369       "MaxIters" -> 100,
370       "NumCycles" -> 10};
371 FitToHam[numE_Integer, expData_List, fitToSymbols_List,
        simplifier_List, OptionsPattern[]] :=
372   (
373     PrintFun         = OptionValue["PrintFun"];
374     fitToVars        = ToExpression[ToString[#] <> "v"] & /@
        fitToSymbols;
375     stringfitToVars  = ToString /@ fitToVars;
376     slackChan        = OptionValue["SlackChannel"];
377     maxHistory       = OptionValue["MaxHistory"];
378     maxIters         = OptionValue["MaxIters"];
379     numCycles        = OptionValue["NumCycles"];
380     ln               = theLanthanides[[numE]];
381     logFilePrefix    = If[OptionValue["FilePrefix"] == "",
382                           ToString[theLanthanides[[numE]]],
383                           OptionValue["FilePrefix"]];
384     PrintFun["Assembling the Hamiltonian for f^", numE, " ..."];
385     ham = HamMatrixAssembly[numE];
386     PrintFun["Simplifying the symbolic expression for the Hamiltonian
         in terms of the given simplifier..."];
387     ham = ReplaceInSparseArray[ham, simplifier];
388     PrintFun["Determining the variables to be fit for ..."];
389     (* as they remain after simplifying *)
390     fitVars = Variables[Normal[ham]];
391     (* append v to symbols *)
392     varVars = ToExpression[ToString[#] <> "v"] & /@ fitVars;
393
394     PrintFun[
395     "Compiling a function for efficient evaluation of the Hamiltonian
         matrix ..."];
396     compHam = Compile[Evaluate[fitVars], Evaluate[N[Normal[ham]]]];
397
398     PrintFun[
399     "Defining the cost function according to given energies and state
         labels ..."];
400
401     varPatterns = StringJoin[{ToString[#], "_?NumericQ"}] & /@
        fitVars;
402     varPatterns = Riffle[varPatterns, ", "];
403     varPatterns = StringJoin[varPatterns];
404     vars = ToString[#] & /@ fitVars;
405     vars = Riffle[vars, ", "];
406     vars = StringJoin[vars];
```

```
407
408     basis = BasisLSJMJ[numE];
409
410     (* define the cost functions given the problem variables *)
411     energyCostFunString =
412     energyCostFunTemplate[<|
413       "varPatterns" -> varPatterns,
414       "vars" -> vars|>];
415     ToExpression[energyCostFunString];
416     energyAndLabelCostFunString = energyAndLabelCostFunTemplate[<|"
      varPatterns" -> varPatterns, "vars" -> vars|>];
417     ToExpression[energyAndLabelCostFunString];
418
419     PrintFun["getting starting values from LaF3..."];
420     lnParams = LoadLaF3Parameters[ln];
421     bills = Table[lnParams[ToExpression[StringTake[ToString[varvar],
      {1, -2}]]], {varvar, varVars}];
422
423     (* define the function arguments with the frozen args in place *)
424     activeArgs = Table[
425       If[MemberQ[fitToVars, varvar],
426       varvar,
427       lnParams[ToExpression[StringTake[ToString[varvar], {1, -2}]]]],
428     {varvar, varVars}
429     ];
430     activeArgs = StringJoin[Riffle[ToString /@ activeArgs, ", "]];
431     (* the constraints, very important *)
432     constraints = N[Constrainer[fitToVars, ln]];
433     complementaryArgs = Table[
434       If[MemberQ[fitToVars, varvar],
435       varvar,
436       lnParams[ToExpression[StringTake[ToString[varvar], {1, -2}]]]],
437     {varvar, varVars}
438     ];
439
440     fromBill = {B02v -> B02, B04v -> B04, B06v -> B06, B22v -> B22,
441     B24v -> B24, B26v -> B26, B44v -> B44, B46v -> B46, B66v -> B66,
442     M0v -> M0, P2v -> P2} /. lnParams;
443
444     If[Not[ValueQ[noteboo]] && OptionValue["ProgressWindow"],
445     noteboo = ProgressNotebook["Basic" -> False];
446     ];
447
448     threadHeaderTemplate = StringTemplate[
449     "(`idx`/`reps`) Fitting data for `ln` using `freeVars`."
450     ];
451     solutions = {};
452     Do[
453     (
454       (* Remove the downvalues of the cost function *)
455       (* DownValues[TheRightSignedPath] = {DownValues[
      TheRightSignedPath][[-1]]}; *)
456       (* start history anew *)
457       rmsHistory = {};
458       paramSols  = {};
459       startTime  = Now;
460       threadMessage = threadHeaderTemplate[
461           <|"reps" -> numCycles,
462           "idx" -> rep,
463           "ln" -> ln,
464           "freeVars" -> ToString[fitToVars]|>];
465       If[slackChan =!= None,
466         threadTS = PostMessageToSlack[threadMessage, slackChan]["ts"]
467       ];
468       solverTemplateNMini = StringTemplate["
469         numIter = 0;
470         sol = NMinimize[
471           Evaluate[
472             Join[{TheRightSignedPath[`activeArgs`]},
473               constraints
474             ]
475           ],
476           fitToVars,
477           MaxIterations->`maxIterations`,
478           Method->`Method`,
479           `Monitor`:>(
```

```
480             currentErr = TheRightSignedPath[‘activeArgs‘];
481             numIter   += 1;
482             rmsHistory = AddToList[rmsHistory, currentErr, maxHistory
    , False];
483             paramSols  = AddToList[paramSols, fitToVars, maxHistory,
    False];
484           )
485         ]
486       "];
487       solverCode = solverTemplateNMini[<|
488         "maxIterations" -> maxIters,
489         "Method" -> "{\"DifferentialEvolution\",
490             \"PostProcess\"   -> False,
491             \"ScalingFactor\" -> 0.9,
492             \"RandomSeed\"    -> RandomInteger[{0,1000000}],
493             \"SearchPoints\"  -> 10}",
494         "Monitor" -> "StepMonitor",
495         "activeArgs" -> activeArgs|>];
496       ToExpression[solverCode];
497       timeTaken = QuantityMagnitude[Now - startTime, "Seconds"];
498       Print["Took " <> ToString[timeTaken] <> "s"];
499       Print[sol];
500       {bestError, bestParams} = sol;
501       resultMessage = "sigma=" <> ToString[Round[bestError, 0.1]];
502       logFname = LogSol[sol, logFilePrefix];
503       If[slackChan =!= None,
504       (
505         PostMessageToSlack[resultMessage,   slackChan, "threadTS" ->
    threadTS];
506         PostFileToSlack[logFname, logFname, slackChan, "threadTS" ->
    threadTS];
507       )
508       ];
509
510       vsBill = TableForm[
511       Transpose[{
512         First /@ fromBill,
513         Last /@ fromBill,
514         Round[Last /@ bestParams, 1.]}],
515       TableHeadings -> {None, {"Param", "Bill Bkq", "ql Bkq"}}
516       ];
517       If[slackChan =!= None,
518         PostPdfToSlack[logFname, vsBill, slackChan, "threadTS" ->
    threadTS]
519       ];
520
521       (* analysis code *)
522
523       finalHam = compHam @@ (complementaryArgs /. bestParams);
524       {eigenEnergies, eigenVecs} = Eigensystem[finalHam];
525       ordering = Ordering[eigenEnergies];
526       eigenEnergies = eigenEnergies - Min[eigenEnergies];
527       states = Transpose[{eigenEnergies, eigenVecs}];
528       states = states[[ordering]];
529       coarseStates = ParseStates[states, basis];
530
531       (* The eigenvectors need to be simplified in order to compare
    labels to labels *)
532       coarseStates = {#[[1]], #[[-1]]} & /@ coarseStates;
533       missingLevels = Length[coarseStates] - Length[expData];
534       (* The energies are in the first element of the tuples. *)
535       energyDiffFun = (Abs[#1[[1]] - #2[[1]]]) &;
536       (* matching disregarding labels to get overall scale for
    scaling differences in labels *)
537       energyFlow = FlowMatching[coarseStates,
538               expData,
539               "notMatched" -> missingLevels,
540               "CostFun" -> energyDiffFun];
541       energyPairs = {#[[1]][[1]], #[[2]][[1]]} & /@ energyFlow[[1]];
542       energyRms = Sqrt[Total[(Abs[#[[2]] - #[[1]]])^2 & /@
    energyPairs] / Length[energyPairs]];
543       (* matching using both labels and energies *)
544       energyAndLabelFun = (Abs[#1[[1]] - #2[[1]]] + EditDistance
    [#1[[2]],#2[[2]]]) &;
545       energyAndLabelFlow = FlowMatching[coarseStates,
546             expData,
```

```
            "notMatched" -> (Length[coarseStates] - Length[expData]),
            "CostFun" -> energyAndLabelFun];
      totalAvgCost = Total[energyAndLabelFun @@ # & /@
    energyAndLabelFlow[[1]]] / Length[energyAndLabelFlow[[1]]];

      compa = (Flatten /@ energyAndLabelFlow[[1]]);
      compa = Join[
          #,
          {
            #[[2]] == #[[4]],
            If[NumberQ[#[[1]]],
              Round[#[[1]] - #[[3]],1],
              ""
            ],
          #[[5]] - #[[3]],
          Which[
            Round[Abs[#[[1]] - #[[3]]]] < Round[Abs[#[[5]] -
    #[[3]]]],
              "Better",
            Round[Abs[#[[1]] - #[[3]]]] == Round[Abs[#[[5]] -
    #[[3]]]],
              "Equal",
            True,
              "Worse"
          ]
          }
      ] & /@ compa;
      atable = TableForm[compa,
        TableHeadings -> {None,
          {"ql", "ql", "Bill (exp)", "Bill (exp)",
          "Bill (calc)", "labels=", "ql - exp", "bill - exp"}}
      ];
      atable = Framed[atable, FrameMargins -> 20];
      upsAndDowns = {
        {"Better", Length[Select[compa, #[[-1]] == "Better" &]]},
        {"Equal",  Length[Select[compa, #[[-1]] == "Equal" &]]},
        {"Worse",  Length[Select[compa, #[[-1]] == "Worse" &]]}
      };
      upsAndDowns = TableForm[upsAndDowns];
      If[slackChan =!= None,
        PostPdfToSlack["table", atable, slackChan, "threadTS" ->
    threadTS];
      ];
      solutions = Append[solutions, sol];
    ),
    {rep, 1, numCycles}
    ];
  )

TruncationFit::usage="TruncationFit[numE, expData, numReps,
    activeVars, startingValues, Options] fits the given expData in an
    f^numE configuration, generating numReps different solutions, and
    varying the symbols in activeVars. The list startingValues is a
    list with all of the parameters needed to define the Hamiltonian (
    including values for activeVars, which will be disregarded but are
     required as position placeholders). The function returns a list
    of solutions. The solutions are the results of the NMinimize
    function using the Differential Evolution method. The solutions
    are a list of tuples. The first element of the tuple is the RMS
    error and the second element is a list of replacement rules for
    the fitted parameters. Once each NMinimize is done, the function
    saves the solutions to a file. The file is named with a prefix and
     a UUID. The file is saved in the log sub-directory as a .m file.
    The solver is always constrained by the relevant subsets of
    constraints for the parameters as provided by the Constrainer
    function. By default the Differential Evolution method starts with
     a generation of points within the given constraints, however it
    is also possible here to have a different region from which the
    initial points are chosen with the option \"StartingForVars\".

The following options can be used:
  \"SignatureCheck\" : if True then then the function ends
    prematurely, printing a list with the symbols that would have
    defined the Hamiltonian after all simplifications have been
    applied. Useful to check the entire parameter set that the
    Hamiltonian has, which has to match one-to-one what is provided by
```

```
    startingValues.
596 \"FilePrefix\" : the prefix to use for the file name, by default
    this is the symbol for the lanthanide.
597 \"AccuracyGoal\": sets the accuracy goal for NMinimize, the default
    is 3.
598 \"MaxHistory\" : determines how long the logs for the solver can be
    .
599 \"MaxIterations\": determines the maximum number of iterations used
    by NMinimize.
600
601 \"AccuracyGoal\": the accuracy goal used by NMinimize, default of
    3.
602 \"TrucationEnergy\": if Automatic then the maximum energy in
    expData is taken, else it takes the value set by this option. In
    all cases the energies in expData are truncated to this value.
603 \"PrintFun\": the function used to print progress messages, the
    default is PrintTemporary.
604 \"SlackChannel\": name of the Slack channel to which to dump
    progress messaages, the default is None which disables this option
    entirely.
605 \"ProgressView\": whether or not a progress window will be opened
    to show the progress of the solver, the default is True.
606
607 \"ReturnHashFileNameAndExit\": if True then the function returns
    the name of the file with the solutions and exits, the default is
    False.
608 \"StartingForVars\": if different from {} then it has to be a list
    with two elements. The first element being a number that
    determines the fraction half-width of the interval used for
    choosing the initial generation of points. The second element
    being a list with as many elements as activeVars corresponding to
    the midpoints from which the intial generation points are chosen.
    The default is {}.
609 \"DE:CrossProbability\": the cross probability used by the
    Differential Evolution method, the default is 0.5.
610 \"DE:ScalingFactor\": the scaling factor used by the Differential
    Evolution method, the default is 0.6.
611 \"DE:SearchPoints\": the number of search points used by the
    Differential Evolution method, the default is Automatic.
612
613 \"MagneticSimplifier\": a list of replacement rules to simplify the
    Marvin and pesudo-magnetic paramters.
614 \"MagFieldSimplifier\": a list of replacement rules to specify a
    magnetic field (in T), if set to {}, then {Bx, By, Bz} can also
    then be used as variables to be fitted for.
615 \"SymmetrySimplifier\": a list of replacements rules to simplify
    the crystal field.
616 \"OtherSimplifier\": an additiona list of replacement rules that
    are applied to the Hamiltonian before computing with it.
617 \"ThreeBodySimplifier\": the default is an Association that simply
    states which three body parameters Tk are zero in different
    configurations, if a list of replacement rules is used then that
    is used instead for the given problem.
618
619 \"FreeIonSymbols\": a list with the symbols to be included in the
    intermediate coupling basis.
620 \"AppendToLogFile\": an association appended to the log file under
    the key \"Appendix\".
621 ";
622 Options[TruncationFit]={
623   "MaxHistory"       -> 200,
624   "MaxIterations"    -> 100,
625   "FilePrefix"       -> "",
626   "AccuracyGoal"     -> 3,
627   "TruncationEnergy" -> Automatic,
628   "PrintFun"         -> PrintTemporary,
629   "SlackChannel"     -> None,
630   "ProgressView"     -> True,
631   "SignatureCheck"   -> False,
632   "AppendToLogFile"  -> <||>,
633   "StartingForVars"  -> {},
634   "ReturnHashFileNameAndExit" -> False,
635   "DE:CrossProbability" -> 0.5,
636   "DE:ScalingFactor"    -> 0.6,
637   "DE:SearchPoints"     -> Automatic,
638   "MagneticSimplifier"  -> {
```

```
    M2 -> 56/100 M0,
    M4 -> 31/100 M0,
    P4 -> 1/2 P2,
    P6 -> 1/10 P2},
  "MagFieldSimplifier" -> {
    Bx->0,By->0,Bz->0
    },
  "SymmetrySimplifier" -> {
    B12->0,B14->0,B16->0,B34->0,B36->0,B56->0,
    S12->0,S14->0,S16->0,S22->0,S24->0,S26->0,S34->0,S36->0,
    S44->0,S46->0,S56->0,S66->0
    },
"OtherSimplifier" -> {
    F0->0,
    P0->0,
    \[Sigma]SS->0,
    T11p->0,T12->0,T14->0,T15->0,
    T16->0,T18->0,T17->0,T19->0,T2p->0
    },
"ThreeBodySimplifier" -> <|
    1  -> {
      T2->0,T3->0,T4->0,T6->0,T7->0,T8->0,T11p->0,T12->0,T14->0,T15
    ->0,T16->0,T18->0,T17->0,T19->0,T2p->0},2->{T2->0,T3->0,T4->0,T6
    ->0,T7->0,T8->0,T11p->0,T12->0,T14->0,T15->0,T16->0,T18->0,T17->0,
    T19->0,T2p->0
      },
    3  -> {},
    4  -> {},
    5  -> {},
    6  -> {},
    7  -> {},
    8  -> {},
    9  -> {},
    10 -> {},
    11 -> {},
    12 -> {
      T3->0,T4->0,T6->0,T7->0,T8->0,T11p->0,T12->0,T14->0,T15->0,T16
    ->0,T18->0,T17->0,T19->0,T2p->0
      },
    13->{
      T2->0,T3->0,T4->0,T6->0,T7->0,T8->0,T11p->0,T12->0,T14->0,T15
    ->0,T16->0,T18->0,T17->0,T19->0,T2p->0
      }
    |>,
"FreeIonSymbols" -> {F0, F2, F4, F6, M0, P2, α, β, γ, ζ, T2, T3, T4,
    T6, T7, T8}
};
TruncationFit[numE_Integer, expData0_List, numReps_Integer,
    activeVars_List, startingValues_List, OptionsPattern[]]:=(
  ln               = theLanthanides[[numE]];
  expData          = expData0;
  PrintFun         = OptionValue["PrintFun"];
  truncationEnergy = If[OptionValue["TruncationEnergy"]===Automatic,
    Max[First/@expData],
    OptionValue["TruncationEnergy"]
  ];
  oddsAndEnds   = <||>;
  expData       = Select[expData, #[[1]] <= truncationEnergy &];
  maxIterations = OptionValue["MaxIterations"];
  maxHistory    = OptionValue["MaxHistory"];
  slackChan     = OptionValue["SlackChannel"];
  accuracyGoal  = OptionValue["AccuracyGoal"];
  logFilePrefix = If[OptionValue["FilePrefix"] == "",
                     ToString[theLanthanides[[numE]]],
                     OptionValue["FilePrefix"]];

  usingInitialRange  = Not[OptionValue["StartingForVars"] === {}];
  If[usingInitialRange,
    (
      PrintFun["Using the solver for initial values in range ..."];
      {fractionalWidth, startVarValues} = OptionValue["
    StartingForVars"];
    )
  ];

  magneticSimplifier  = OptionValue["MagneticSimplifier"];
```

```
magFieldSimplifier  = OptionValue["MagFieldSimplifier"];
symmetrySimplifier  = OptionValue["SymmetrySimplifier"];
otherSimplifier     = OptionValue["OtherSimplifier"];
threeBodySimplifier = If[Head[OptionValue["ThreeBodySimplifier"]]
  == Association,
   OptionValue["ThreeBodySimplifier"][numE],
   OptionValue["ThreeBodySimplifier"]
];
simplifier          = Join[magneticSimplifier,
                      magFieldSimplifier,
                      symmetrySimplifier,
                      threeBodySimplifier,
                      otherSimplifier];
freeIonSymbols = OptionValue["FreeIonSymbols"];
runningInteractive = (Head[$ParentLink] === LinkObject);

oddsAndEnds["simplifier"] = simplifier;
oddsAndEnds["freeIonSymbols"] = freeIonSymbols;
oddsAndEnds["truncationEnergy"] = truncationEnergy;
oddsAndEnds["numE"] = numE;
oddsAndEnds["expData"] = expData;
oddsAndEnds["numReps"] = numReps;
oddsAndEnds["activeVars"] = activeVars;
oddsAndEnds["startingValues"] = startingValues;
oddsAndEnds["maxIterations"] = maxIterations;
oddsAndEnds["PrintFun"] = PrintFun;
oddsAndEnds["ln"] = ln;
oddsAndEnds["numE"] = numE;
oddsAndEnds["accuracyGoal"] = accuracyGoal;
oddsAndEnds["Appendix"] = OptionValue["AppendToLogFile"];

hamDim         = Binomial[14, numE];
modelSymbols   = Sort[Select[paramSymbols, Not[MemberQ[Join[
  racahSymbols, chenSymbols,{t2Switch, \[Epsilon],gs},#]]&]];
(* Remove the symbols that will be removed by the simplifier, no
  symbol should remain here that is not in the symbolic hamiltonian
  *)
reducedModelSymbols = Select[modelSymbols, Not[MemberQ[Keys[
  simplifier],#]]&];
If[OptionValue["SignatureCheck"],
   (
   PrintFun["Given the model parameters and the simplifying
  assumptions, the resultant model parameters are:"];
   PrintFun[{reducedModelSymbols}];
   PrintFun["The ordering in these needs to be respected in the
  startValues parameter ..."];
   PrintFun["Exiting ..."];
   Return[""];
   )
];

(*calculate the basis*)
basis          = BasisLSJMJ[numE];
(* grab the Hamiltonian preserving its block structure *)
PrintFun["Assembling the Hamiltonian for f^",numE," keeping the
  block structure ..."];
ham            = HamMatrixAssembly[numE, "ReturnInBlocks"->True];
(* apply the simplifier *)
PrintFun["Simplifying using the given aggregate set of
  simplification rules ..."];
ham            = Map[ReplaceInSparseArray[#, simplifier]&, ham, {2}];

(* Get the reference parameters from LaF3 *)
PrintFun["Getting reference parameters for ",ln," using LaF3 ..."];
lnParams = LoadLaF3Parameters[ln];
freeBies = Prepend[Values[(#->(#/.lnParams))&/@freeIonSymbols],numE
  ];
(* a more explicit alias *)
allVars            = reducedModelSymbols;

oddsAndEnds["allVars"]  = allVars;
oddsAndEnds["freeBies"] = freeBies;

(* reload compiled version if found *)
varHash                  = Hash[{numE, allVars, freeBies,
  truncationEnergy}];
```

```
772  compileIntermediateFname = "compileIntermediateTruncatedHam-"<>
       ToString[varHash]<>".mx";
773  truncadedFname           = "TheTruncatedAndSignedPath-"<>ToString[
       varHash]<>".mx";
774  If[OptionValue["ReturnHashFileNameAndExit"],
775    (
776      Print[varHash];
777      Return[truncadedFname];
778    )
779  ];
780  If[FileExistsQ[compileIntermediateFname],
781    PrintFun["This ion and free-ion params have been compiled before
       (as determined by {numE, allVars, freeBies, truncationEnergy}).
       Loading the previously saved function and intermediate coupling
       basis ..."];
782    {compileIntermediateTruncatedHam, truncatedIntermediateBasis} =
       Import[compileIntermediateFname];,
783  (
784    PrintFun["Zeroing out every symbol in the Hamiltonian that is not
       a free-ion parameter ..."];
785    (* Get the free ion symbols *)
786    freeIonSimplifier = (#->0) & /@ Complement[reducedModelSymbols,
       freeIonSymbols];
787    (* Take the diagonal blocks for the intermediate analysis *)
788    PrintFun["Grabbing the diagonal blocks of the Hamiltonian ..."];
789    diagonalBlocks       = Diagonal[ham];
790    (* simplify them to only keep the free ion symbols *)
791    PrintFun["Simplifying the diagonal blocks to only keep the free
       ion symbols ..."];
792    diagonalScalarBlocks = ReplaceInSparseArray[#,freeIonSimplifier
       ]&/@diagonalBlocks;
793    (* these include the MJ quantum numbers, remove that *)
794    PrintFun["Contracting the basis vectors by removing the MJ
       quantum numbers from the diagonal blocks ..."];
795    diagonalScalarBlocks = FreeHam[diagonalScalarBlocks, numE];
796
797    argsOfTheIntermediateEigensystems         = StringJoin[Riffle[
       Prepend[(ToString[#]<>"v_") & /@ freeIonSymbols,"numE_"],", "]];
798    argsForEvalInsideOfTheIntermediateSystems = StringJoin[Riffle[(
       ToString[#]<>"v") & /@ freeIonSymbols,", "]];
799    PrintFun["argsOfTheIntermediateEigensystems = ",
       argsOfTheIntermediateEigensystems];
800    PrintFun["argsForEvalInsideOfTheIntermediateSystems = ",
       argsForEvalInsideOfTheIntermediateSystems];
801    PrintFun["If the following fails, make sure to modify the
       arguments of TheIntermediateEigensystems to match the ones above
       ..."];
802
803    (* Compile a function that will effectively calculate the
       spectrum of all of the scalar blocks given the parameters of the
       free-ion part of the Hamiltonian *)
804    (* Compile one function for each of the blocks *)
805    PrintFun["Compiling functions for the diagonal blocks of the
       Hamiltonian ..."];
806    compiledDiagonal = Compile[Evaluate[freeIonSymbols], Evaluate[N[
       Normal[#]]]]&/@diagonalScalarBlocks;
807    (* Use that to create a function that will calculate the free-ion
       eigensystem *)
808    TheIntermediateEigensystems[numEv_, F0v_, F2v_, F4v_, F6v_, M0v_,
       P2v_, αv_, βv_, γv_, ζv_, T2v_, T3v_, T4v_, T6v_, T7v_, T8v_] :=
       (
809      theNumericBlocks = (#[F0v, F2v, F4v, F6v, M0v, P2v, αv, βv, γv,
       ζv, T2v, T3v, T4v, T6v, T7v, T8v]&)/@compiledDiagonal;
810      theIntermediateEigensystems = Eigensystem/@theNumericBlocks;
811      Js      = AllowedJ[numEv];
812      basisJ = BasisLSJMJ[numEv,"AsAssociation"->True];
813      (* Having calculated the eigensystems with the removed
       degeneracies, put the degeneracies back in explicitly *)
814      elevatedIntermediateEigensystems = MapIndexed[EigenLever[#1,2Js
       [[#2[[1]]]]+1]&, theIntermediateEigensystems];
815      pivot = If[EvenQ[numEv],0,-1/2];
816      LSJmultiplets = (#[[1]]<>ToString[InputForm[#[[2]]]])&/@Select[
       BasisLSJMJ[numEv],#[[-1]]== pivot &];
817      (* Calculate the multiplet assignments that the intermediate
       basis eigenvectors have *)
818      multipletAssignments = Table[
```

```
819         (
820           J         = Js[[idx]];
821           eigenVecs = theIntermediateEigensystems[[idx]][[2]];
822           majorComponentIndices        = Ordering[Abs[#]][[-1]]&/
    @eigenVecs;
823           majorComponentAssignments    = LSJmultiplets[[#]]&/
    @majorComponentIndices;
824           (* All of the degenerate eigenvectors belong to the same
    multiplet*)
825           elevatedMultipletAssignments = ListRepeater[
    majorComponentAssignments,2J+1];
826           elevatedMultipletAssignments
827         ),
828       {idx, 1, Length[Js]}
829       ];
830       (* Put together the multiplet assignments and the energies *)
831       freeIenergiesAndMultiplets = Transpose/@Transpose[{First/
    @elevatedIntermediateEigensystems, multipletAssignments}];
832       freeIenergiesAndMultiplets = Flatten[freeIenergiesAndMultiplets
    , 1];
833       (* Calculate the change of basis matrix using the intermediate
    coupling eigenvectors *)
834       basisChanger = BlockDiagonalMatrix[Transpose/@Last/
    @elevatedIntermediateEigensystems];
835       basisChanger = SparseArray[basisChanger];
836       Return[{theIntermediateEigensystems, multipletAssignments,
    elevatedIntermediateEigensystems, freeIenergiesAndMultiplets,
    basisChanger}]
837     );
838
839     PrintFun["Calculating the intermediate eigensystems for ",ln,"
    using free-ion params from LaF3 ..."];
840     (* Calculate intermediate coupling basis using the free-ion
    params for LaF3 *)
841     {theIntermediateEigensystems, multipletAssignments,
    elevatedIntermediateEigensystems, freeIenergiesAndMultiplets,
    basisChanger} = TheIntermediateEigensystems@@freeBies;
842
843     (* Use that intermediate coupling basis to compile a function for
     the full Hamiltonian *)
844     allFreeEnergies = Flatten[First/@elevatedIntermediateEigensystems
    ];
845     (* Important that the intermediate coupling basis have attached
    energies, which make possible the truncation *)
846     ordering = Ordering[allFreeEnergies];
847     (* Sort the free ion energies and determine which indices should
    be included in the truncation *)
848     allFreeEnergiesSorted          = Sort[allFreeEnergies];
849     {minFreeEnergy, maxFreeEnergy} = MinMax[allFreeEnergies];
850     (* Determine the index at which the energy is equal or larger
    than the truncation energy *)
851     sortedTruncationIndex = Which[
852       truncationEnergy > (maxFreeEnergy-minFreeEnergy),
853       hamDim,
854       True,
855       FirstPosition[allFreeEnergiesSorted-Min[allFreeEnergiesSorted],
    x_/;x>truncationEnergy,{0},1][[1]]
856     ];
857     (* The actual energy at which the truncation is made *)
858     roundedTruncationEnergy = allFreeEnergiesSorted[[
    sortedTruncationIndex]];
859
860     (* The indices that enact the truncation *)
861     truncationIndices = ordering[[;;sortedTruncationIndex]];
862
863     (* Using the ham (with all the symbols) change the basis to the
    computed one *)
864     PrintFun["Changing the basis of the Hamiltonian to the
    intermediate coupling basis ..."];
865     intermediateHam            = Transpose[basisChanger].ArrayFlatten
    [ham].basisChanger;
866     (* Using the truncation indices truncate that one *)
867     PrintFun["Truncating the Hamiltonian ..."];
868     truncatedIntermediateHam   = intermediateHam[[truncationIndices,
    truncationIndices]];
869     (* These are the basis vectors for the truncated hamiltonian *)
```

```
870     PrintFun["Saving the truncated intermediate basis ..."];
871     truncatedIntermediateBasis = basisChanger[[All,truncationIndices
        ]];
872
873     PrintFun["Compiling a function for the truncated Hamiltonian ..."
        ];
874     (* Compile a function that will calculate the truncated
        Hamiltonian given the parameters in allVars, this is the function
        to be use in fitting *)
875     compileIntermediateTruncatedHam = Compile[Evaluate[allVars],
876                                       Evaluate[N[Normal[
        truncatedIntermediateHam]]]];
877     (* Save the compiled function *)
878     PrintFun["Saving the compiled function for the truncated
        Hamiltonian and the truncatedIntermediateBasis..."];
879     Export[compileIntermediateFname, {compileIntermediateTruncatedHam
        , truncatedIntermediateBasis}];
880   )
881   ];
882
883   TheTruncatedAndSignedPathGenerator::usage = "This function puts
        together the necessary expression for defining a function which
        has as arguments all the symbolic values in varsMixedWithVals and
        which feeds to compileIntermediateTruncatedHam the arguments as
        given in varsMixedWithVals. varsMixedWithVals needs to respect the
         order of aruments expected by compileIntermediateTruncatedHam.
        Once the necessary template has been used this function then
        results in the definition of the function
        TheTruncatedAndSignedPath.";
884   TheTruncatedAndSignedPathGenerator[varsMixedWithVals_List]:=(
885     variableVars   = Select[varsMixedWithVals,Not[NumericQ[#]]&];
886     numQSignature = StringJoin[Riffle[(ToString[#]<>"_?NumericQ")&/
        @variableVars,", "]];
887     varWithValsSignature = StringJoin[Riffle[(ToString[#]<>"")&/
        @varsMixedWithVals,", "]];
888     funcString     = truncatedEnergyCostTemplate[<|"varsWithNumericQ"
        ->numQSignature,"varsMixedWithFixedVals" -> varWithValsSignature
        |>];
889     ClearAll[TheTruncatedAndSignedPath];
890     ToExpression[funcString]
891   );
892
893   (* We need to create a function call that has all the frozen
        parameters in place and all the active symbols unevaluated *)
894   (* find the indices of the activeVars to create the function
        signature *)
895   activeVarIndices = Flatten[Position[allVars,#]&/@activeVars];
896   (* we start from the numerical values in the current best*)
897   jobVars          = startingValues;
898   (* we then put back the symbols that should be unevaluated *)
899   jobVars[[activeVarIndices]] = activeVars;
900
901   oddsAndEnds["jobVars"] = jobVars;
902   (* calculate the constraints *)
903   constraints                = N[Constrainer[activeVars,ln]];
904   oddsAndEnds["constraints"] = constraints;
905   (* This is useful for the progress window *)
906   activeVarsString  = StringJoin[Riffle[ToString/@activeVars,", "]];
907   TheTruncatedAndSignedPathGenerator[jobVars];
908   stringPartialVars = ToString/@activeVars;
909
910   activeVarsWithRange = If[usingInitialRange,
911     MapIndexed[Flatten[{#1,
912         (1-Sign[startVarValues[[#2]]]*fractionalWidth) *
        startVarValues[[#2]],
913         (1+Sign[startVarValues[[#2]]]*fractionalWidth) *
        startVarValues[[#2]]
914         }]&, activeVars],
915     activeVars
916   ];
917
918   (* this is the template for the minimizer *)
919   solverTemplateNMini = StringTemplate["
920   numIter = 0;
921   sol = NMinimize[
922     Evaluate[
```

```
      Join[{TheTruncatedAndSignedPath[`activeVarsString`]},
        constraints
      ]
    ],
    activeVarsWithRange,
    AccuracyGoal   -> `accuracyGoal`,
    MaxIterations  -> `maxIterations`,
    Method->`Method`,
    `Monitor`:>(
      currentErr = TheTruncatedAndSignedPath[`activeVarsString`];
      currentParams = activeVars;
      numIter   += 1;
      rmsHistory = AddToList[rmsHistory, currentErr, maxHistory, False];
      paramSols  = AddToList[paramSols, activeVars, maxHistory, False];
      If[Not[runningInteractive],(
        Print[numIter,\"/\",`maxIterations`];
        Print[\"err    = \", ToString[NumberForm[Round[currentErr,0.001],{Infinity,3}]]];
        Print[\"params = \", ToString[NumberForm[Round[#,0.0001],{Infinity,4}]] &/@ currentParams];
        )
      ];
    )
  ]
  "];
  methodStringTemplate = StringTemplate["
      {\"DifferentialEvolution\",
        \"PostProcess\"   -> False,
        \"ScalingFactor\" -> `DE:ScalingFactor`,
        \"CrossProbability\" -> `DE:CrossProbability`,
        \"RandomSeed\"    -> RandomInteger[{0,1000000}],
        \"SearchPoints\"  -> `DE:SearchPoints`}"];
  methodString = methodStringTemplate[<|
    "DE:ScalingFactor" -> OptionValue["DE:ScalingFactor"],
    "DE:CrossProbability" -> OptionValue["DE:CrossProbability"],
    "DE:SearchPoints" -> OptionValue["DE:SearchPoints"]|>];
  (* Evaluate the template *)
  solverCode = solverTemplateNMini[<|
    "accuracyGoal"  -> accuracyGoal,
    "maxIterations" -> maxIterations,
    "Method"->"{\"DifferentialEvolution\",
                \"PostProcess\"   -> False,
                \"ScalingFactor\" -> 0.6,
                \"CrossProbability\" -> 0.25,
                \"RandomSeed\"    -> RandomInteger[{0,1000000}],
                \"SearchPoints\"  -> Automatic}",
    "Monitor"->"StepMonitor",
    "activeVarsString"->activeVarsString|>
  ];
  threadHeaderTemplate = StringTemplate[ "(`idx`/`reps`) Fitting data for `ln` using `freeVars`."];
  (* Find as many solutions as numReps *)
  sols = Table[(
    rmsHistory        = {};
    paramSols         = {};
    openNotebooks     = If[runningInteractive,
              ("WindowTitle"/.NotebookInformation[#]) & /@ Notebooks[],
              {}];
    If[Not[MemberQ[openNotebooks,"Solver Progress"]] && OptionValue["ProgressView"],
      ProgressNotebook["Basic"->False]
    ];
    If[Not[slackChan === None],
    (
      threadMessage = threadHeaderTemplate[<|"reps" -> numReps, "idx" -> rep, "ln" -> ln,
        "freeVars" -> ToString[activeVars]|>];
      threadTS = PostMessageToSlack[threadMessage, slackChan]["ts"];
    )
    ];
    startTime = Now;
    ToExpression[solverCode];

```

```
    timeTaken = QuantityMagnitude[Now - startTime, "Seconds"];
    Print["Took " <> ToString[timeTaken] <> "s"];
    Print[sol];
    bestError     = sol[[1]];
    bestParams    = sol[[2]];
    resultMessage = "sigma=" <> ToString[Round[bestError, 0.1]];
    solAssoc = <|
      "bestRMS"      -> bestError,
      "solHistory"   -> rmsHistory,
      "bestParams"   -> bestParams,
      "paramHistory" -> paramSols,
      "timeTaken/s"    -> timeTaken
      |>;
    solAssoc = Join[solAssoc,   oddsAndEnds];
    logFname = LogSol[solAssoc, logFilePrefix];

    If[Not[slackChan===None],(
      PostMessageToSlack[resultMessage,   slackChan, "threadTS" ->
    threadTS];
      PostFileToSlack[StringSplit[logFname,"/"][[-1]], logFname,
    slackChan, "threadTS" -> threadTS]
    )
    ];
    solAssoc
    ),
    {rep,1,numReps}
  ];
  Return[sols];
);

ClassicalFit::usage="ClassicalFit[numE, expData, excludeDataIndices,
    problemVars, startValues, \[Sigma]exp, constraints_List, Options]
    fits the given expData in an f^numE configuration, by using the
    symbols in problemVars. The symbols given in problemVars may be
    constrained or held constant, this being controlled by constraints
     list which is a list of replacement rules expressing desired
    constraints. The constraints list additional constraints imposed
    upon the model parameters that remain once other simplifications
    have been \"baked\" into the compiled Hamiltonians that are used
    to increase the speed of the calculation.

Important, note that in the case of odd number of electrons the given
     data must explicitly include the Kramers degeneracy;
    excludeDataIndices must be compatible with this.

The list expData needs to be a list of lists with the only
    restriction that the first element of them corresponds to energies
     of levels. In this list, an empty value can be used to indicate
    known gaps in the data. Even if the energy value for a level is
    known (and given in expData) certain values can be omitted from
    the fitting procedure through the list excludeDataIndices, which
    correspond to indices in expData that should be skipped over.

The Hamiltonian used for fitting is version that has been truncated
    either by using the maximum energy given in expData or by manually
     setting a truncation energy using the option \"TruncationEnergy\"
    .

The argument \[Sigma]exp is the estimated uncertainty in the
    differences between the calculated and the experimental energy
    levels. This is used to estimate the uncertainty in the fitted
    parameters. Admittedly this will be a rough estimate (at least on
    the contribution of the calculated uncertainty), but it is better
    than nothing and may at least provide a lower bound to the
    uncertainty in the fitted parameters. It is assumed that the
    uncertainty in the differences between the calculated and the
    experimental energy levels is the same for all of them.

The list startValues is a list with all of the parameters needed to
    define the Hamiltonian (including the initial values for
    problemVars).

The function saves the solution to a file. The file is named with a
    prefix (controlled by the option \"FilePrefix\") and a UUID. The
    file is saved in the log sub-directory as a .m file.

```

```
Here's a description of the different parts of this function: first the Hamiltonian is assembled and simplified using the given simplifications. Then the intermediate coupling basis is calculated using the free-ion parameters for the given lanthanide. The Hamiltonian is then changed to the intermediate coupling basis and truncated. The truncated Hamiltonian is then compiled into a function that can be used to calculate the energy levels of the truncated Hamiltonian. The function that calculates the energy levels is then used to fit the experimental data. The fitting is done using FindMinimum with the Levenberg-Marquardt method.

The function returns an association with the following keys:

- \"bestRMS\" which is the best \[Sigma] value found.
- \"bestParams\" which is the best set of parameters found for the variables that were not constrained.
- \"bestParamsWithConstraints\" which has the best set of parameters (from - \"bestParams\") together with the used constraints. These include all the parameters in the model, even those that were not fitted for.
- \"paramSols\" which is a list of the parameters trajectories during the stepping of the fitting algorithm.
- \"timeTaken/s\" which is the time taken to find the best fit.
- \"simplifier\" which is the simplifier used to simplify the Hamiltonian.
- \"excludeDataIndices\" as given in the input.
- \"startValues\" as given in the input.

- \"freeIonSymbols\" which are the symbols used in the intermediate coupling basis.
- \"truncationEnergy\" which is the energy used to truncate the Hamiltonian, if it was set to Automatic, the value here is the actual energy used.
- \"numE\" which is the number of electrons in the f^numE configuration.
- \"expData\" which is the experimental data used for fitting.
- \"problemVars\" which are the symbols considered for fitting

- \"maxIterations\" which is the maximum number of iterations used by NMinimize.
- \"hamDim\" which is the dimension of the full Hamiltonian.
- \"allVars\" which are all the symbols defining the Hamiltonian under the aggregate simplifications.
- \"freeBies\" which are the free-ion parameters used to define the intermediate coupling basis.
- \"truncatedDim\" which is the dimension of the truncated Hamiltonian.
- \"compiledIntermediateFname\" the file name of the compiled function used for the truncated Hamiltonian.

- \"fittedLevels\" which is the number of levels fitted for.
- \"actualSteps\" the number of steps that FindMiniminum actually took.
- \"solWithUncertainty\" which is a list of replacement rules of the form (paramSymbol -> {bestEstimate, uncertainty}).
- \"rmsHistory\" which is a list of the \[Sigma] values found during the fitting.
- \"Appendix\" which is an association appended to the log file under the key \"Appendix\".
- \"presentDataIndices\" which is the list of indices in expData that were used for fitting, this takes into account both the empty indices in expData and also the indices in excludeDataIndices.

- \"states\" which contains a list of eigenvalues and eigenvectors for the fitted model, this is only available if the option \"SaveEigenvectors\" is set to True; if a general shift of energy was allowed for in the fitting, then the energies are shifted accordingly.
- \"energies\" which is a list of the energies of the fitted levels, this is only available if the option \"SaveEigenvectors\" is set to False. If a general shift of energy was allowed for in the fitting, then the energies are shifted accordingly.
- \"degreesOfFreedom\" which is equal to the nunber of fitted state energies minus the number of parameters used in fitting.
```

```
The function admits the following options with corresponding default values:
- \"MaxHistory\" : determines how long the logs for the solver can be.
- \"MaxIterations\": determines the maximum number of iterations used by NMinimize.
- \"FilePrefix\" : the prefix to use for the subfolder in the log filder, in which the solution files are saved, by default this is \"calcs\" so that the calculation files are saved under the directory \"log/calcs\".
- \"AddConstantShift\" : if True then a constant shift is allowed in the fitting, default is False. If this is the case the variable \"\[Epsilon]\" is added to the list of variables to be fitted for, it must not be included in problemVars.

- \"AccuracyGoal\": the accuracy goal used by NMinimize, default of 5.
- \"TrucationEnergy\": if Automatic then the maximum energy in expData is taken, else it takes the value set by this option. In all cases the energies in expData are only considered up to this value.
- \"PrintFun\": the function used to print progress messages, the default is PrintTemporary.
- \"RefParamsVintage\": the vintage of the reference parameters to use. The reference parameters are both used to determine the truncated Hamiltonian, and also as starting values for the solver. It may be \"LaF3\", in which case reference parameters from Carnall are used. It may also be \"LiYF4\", in which case the reference parameters from the LiYF4 paper are used. It may also be Automatic, in which case the given experimental data is used to determine starting values for F^k and ζ. It may also be a list or association that provides values for the Slater integrals and spin-orbit coupling, the remaining necessary parameters complemented by using \"LaF3\".

- \"SlackChannel\": name of the Slack channel to which to dump progress messaages, the default is None which disables this option.
- \"ProgressView\": whether or not a progress window will be opened to show the progress of the solver, the default is True.
- \"SignatureCheck\": if True then then the function returns prematurely, returning a list with the symbols that would have defined the Hamiltonian after all simplifications have been applied. Useful to check the entire parameter set that the Hamiltonian has, which has to match one-to-one what is provided by startingValues.
- \"SaveEigenvectors\": if True then the both the eigenvectors and eigenvalues are saved under the \"states\" key of the returned association. If False then only the energies are saved, the default is False.

- \"AppendToLogFile\": an association appended to the log file under the key \"Appendix\".
- \"MagneticSimplifier\": a list of replacement rules to simplify the Marvin and pesudo-magnetic paramters. Here the ratios of the Marvin parameters and the pseudo-magnetic parameters are defined to simplify the magnetic part of the Hamiltonian.
- \"MagFieldSimplifier\": a list of replacement rules to specify a magnetic field (in T), if set to {}, then {Bx, By, Bz} can also be used as variables to be fitted for.

- \"SymmetrySimplifier\": a list of replacements rules to simplify the crystal field.
- \"OtherSimplifier\": an additional list of replacement rules that are applied to the Hamiltonian before computing with it. Here the spin-spin contribution can be turned off by setting \[Sigma]SS->0, which is the default.
";
Options[ClassicalFit] = {
  "MaxHistory"       -> 200,
  "MaxIterations"    -> 100,
  "FilePrefix"       -> "calcs",
  "ProgressView"     -> True,
  "TruncationEnergy" -> Automatic,
  "AccuracyGoal"     -> 5,
  "PrintFun"         -> PrintTemporary,
```

```
  "SlackChannel"     -> None,
  "RefParamsVintage" -> "LaF3",
  "SignatureCheck"   -> False,
  "AddConstantShift" -> False,
  "SaveEigenvectors" -> False,
  "AppendToLogFile"  -> <||>,
  "SaveToLog"        -> False,
  "Energy Uncertainty in K" -> Automatic,
  "MagneticSimplifier" -> {
    M2 -> 56/100 M0,
    M4 -> 31/100 M0,
    P4 -> 1/2 P2,
    P6 -> 1/10 P2
    },
  "MagFieldSimplifier" -> {
    Bx -> 0,
    By -> 0,
    Bz -> 0
    },
  "SymmetrySimplifier" -> {
    B12->0, B14->0, B16->0, B34->0, B36->0, B56->0,
    S12->0 ,S14->0, S16->0, S22->0, S24->0, S26->0,
    S34->0 ,S36->0, S44->0, S46->0, S56->0, S66->0
    },
  "OtherSimplifier" -> {
    F0->0,
    P0->0,
    \[Sigma]SS->0,
    T11p->0, T12->0, T14->0, T15->0,
    T16->0, T18->0, T17->0, T19->0, T2p->0,
    wChErrA ->0, wChErrB->0
    },
  "ThreeBodySimplifier" -> <|
    1  -> {
      T2->0, T3->0, T4->0, T6->0, T7->0, T8->0,
      T11p->0, T12->0, T14->0, T15->0, T16->0, T18->0, T17->0, T19
    ->0,
      T2p->0},
    2 -> {
      T2->0, T3->0, T4->0, T6->0, T7->0, T8->0,
      T11p->0, T12->0, T14->0, T15->0, T16->0, T18->0, T17->0, T19
    ->0,
      T2p->0
      },
    3  -> {},
    4  -> {},
    5  -> {},
    6  -> {},
    7  -> {},
    8  -> {},
    9  -> {},
    10 -> {},
    11 -> {},
    12 -> {
      T3->0, T4->0, T6->0, T7->0, T8->0,
      T11p->0, T12->0, T14->0, T15->0, T16->0, T18->0, T17->0, T19
    ->0,
      T2p->0
      },
    13->{
      T2->0, T3->0, T4->0, T6->0, T7->0, T8->0,
      T11p->0, T12->0, T14->0, T15->0, T16->0, T18->0, T17->0, T19
    ->0,
      T2p->0
      }
    |>,
    "FreeIonSymbols" -> {F0, F2, F4, F6, ζ}
};
ClassicalFit[numE_Integer, expData_List, excludeDataIndices_List,
    problemVars_List, startValues_Association, constraints_List,
    OptionsPattern[]]:=Module[
  {accuracyGoal, activeVarIndices, activeVars, activeVarsString,
    activeVarsWithRange, allFreeEnergies, allFreeEnergiesSorted,
    allVars, allVarsVec, argsForEvalInsideOfTheIntermediateSystems,
    argsOfTheIntermediateEigensystems, aVar, aVarPosition, basis,
    basisChanger, basisChangerBlocks, bestError, bestParams, bestRMS,
```

```
    blockShifts, blockSizes, colIdx, compiledDiagonal,
    compiledIntermediateFname, constrainedProblemVars,
    constrainedProblemVarsList, currentRMS, degressOfFreedom,
    dependentVars, diagonalBlocks, diagonalScalarBlocks, diff,
    eigenEnergies, eigenvalueDispenserTemplate, eigenVectors,
    elevatedIntermediateEigensystems, endTime, fmSol, fmSolAssoc,
    fractionalWidth, freeBies, freeIenergiesAndMultiplets, fullHam,
    fullSolVec, funcString, ham, hamDim, hamEigenvaluesTemplate,
    hamString, indepSolVecVec, indepVars, intermediateHam,
    isolationValues, jobVars, lin, linMat, ln, lnParams, logFilePrefix
    , logFname, magneticSimplifier, maxFreeEnergy, maxHistory,
    maxIterations, methodString, methodStringTemplate, minFreeEnergy,
    minpoly, modelSymbols, multipletAssignments, needlePosition,
    numBlocks, numQSignature, numReps, solCompendium, openNotebooks,
    ordering, otherSimplifier, p0, paramBest, paramSigma, perHam,
    polySols, presentDataIndices, PrintFun, problemVarsPositions,
    problemVarsQ, problemVarsQString, problemVarsVec,
    problemVarsWithStartValues, reducedModelSymbols, resultMessage,
    roundedTruncationEnergy, rowIdx, runningInteractive, shiftToggle,
    simplifier, slackChan, sol, solAssoc, sols, solWithUncertainty,
    sortedTruncationIndex, sqdiff, standardValues, starTime,
    startingValues, startTime, startVarValues, states, steps,
    symmetrySimplifier, theIntermediateEigensystems,
    TheIntermediateEigensystems, TheTruncatedAndSignedPathGenerator,
    thisPoly, threadHeaderTemplate, threadMessage, threadTS, timeTaken
    , totalVariance, truncadedFname, truncatedIntermediateBasis,
    truncatedIntermediateHam, truncationEnergy, truncationIndices,
    RefParams, truncationUmbral, usingInitialRange, varHash, varIdx,
    varsWithConstants, varWithValsSignature, \[Lambda]0Vec, \[Lambda]
    exp},
1167 (
1168   \[Sigma]exp   = OptionValue["Energy Uncertainty in K"];
1169   solCompendium = <||>;
1170   refParamsVintage = OptionValue["RefParamsVintage"];
1171   RefParams = Which[
1172     refParamsVintage === "LaF3",
1173     LoadLaF3Parameters,
1174     refParamsVintage === "LiYF4",
1175     LoadLiYF4Parameters,
1176     True,
1177     refParamsVintage
1178   ];
1179   hamDim         = Binomial[14, numE];
1180   addShift       = OptionValue["AddConstantShift"];
1181   ln             = theLanthanides[[numE]];
1182   maxHistory     = OptionValue["MaxHistory"];
1183   maxIterations  = OptionValue["MaxIterations"];
1184   logFilePrefix  = If[OptionValue["FilePrefix"] == "",
1185                     ToString[theLanthanides[[numE]]],
1186                     OptionValue["FilePrefix"]
1187                   ];
1188   accuracyGoal  = OptionValue["AccuracyGoal"];
1189   slackChan     = OptionValue["SlackChannel"];
1190   PrintFun      = OptionValue["PrintFun"];
1191   freeIonSymbols       = OptionValue["FreeIonSymbols"];
1192   runningInteractive   = (Head[$ParentLink] === LinkObject);
1193   magneticSimplifier   = OptionValue["MagneticSimplifier"];
1194   magFieldSimplifier   = OptionValue["MagFieldSimplifier"];
1195   symmetrySimplifier   = OptionValue["SymmetrySimplifier"];
1196   otherSimplifier      = OptionValue["OtherSimplifier"];
1197   threeBodySimplifier = If[Head[OptionValue["ThreeBodySimplifier"]]
      == Association,
1198                            OptionValue["ThreeBodySimplifier"][numE],
1199                            OptionValue["ThreeBodySimplifier"]
1200                          ];
1201
1202   truncationEnergy = If[OptionValue["TruncationEnergy"] ===
      Automatic,
1203     (
1204       PrintFun["Truncation energy set to Automatic, using the
      maximum energy (+20%) in the data ..."];
1205       Round[1.2 * Max[Select[First /@ expData, NumericQ[#] &]]]
1206     ),
1207     OptionValue["TruncationEnergy"]
1208     ];
1209   truncationEnergy = Max[50000, truncationEnergy];
```

```
1210    PrintFun["Using a truncation energy of ", truncationEnergy, " K"
        ];
1211
1212    simplifier = Join[magneticSimplifier,
1213                      magFieldSimplifier,
1214                      symmetrySimplifier,
1215                      threeBodySimplifier,
1216                      otherSimplifier];
1217
1218    PrintFun["Determining gaps in the data ..."];
1219    (* whatever is non-numeric is assumed as a known gap *)
1220    presentDataIndices = Flatten[Position[expData, {_?(NumericQ[#] &)
        , ___}]];
1221    (* some indices omitted here based on the excludeDataIndices
        argument *)
1222    presentDataIndices = Complement[presentDataIndices,
        excludeDataIndices];
1223
1224    solCompendium["simplifier"]         = simplifier;
1225    solCompendium["excludeDataIndices"] = excludeDataIndices;
1226    solCompendium["startValues"]        = startValues;
1227    solCompendium["freeIonSymbols"]     = freeIonSymbols;
1228    solCompendium["truncationEnergy"]   = truncationEnergy;
1229    solCompendium["numE"]               = numE;
1230    solCompendium["expData"]            = expData;
1231    solCompendium["problemVars"]        = problemVars;
1232    solCompendium["maxIterations"]      = maxIterations;
1233    solCompendium["hamDim"]             = hamDim;
1234    solCompendium["constraints"]        = constraints;
1235
1236    modelSymbols  = Sort[Select[paramSymbols, Not[MemberQ[Join[
        racahSymbols, juddOfeltIntensitySymbols, chenSymbols,{t2Switch, \[
        Epsilon], gs, nE}],#]]&]];
1237    (* remove the symbols that will be removed by the simplifier, no
        symbol should remain here that is not in the symbolic Hamiltonian
        *)
1238    reducedModelSymbols = Select[modelSymbols, Not[MemberQ[Keys[
        simplifier],#]]&];
1239
1240    (* this is useful to understand what are the arguments of the
        truncated compiled Hamiltonian *)
1241    If[OptionValue["SignatureCheck"],
1242      (
1243        PrintFun["Given the model parameters and the simplifying
        assumptions, the resultant model parameters are:"];
1244        PrintFun[{reducedModelSymbols}];
1245        PrintFun["Exiting ..."];
1246        Return[""];
1247      )
1248    ];
1249
1250    (* calculate the basis *)
1251    PrintFun["Retrieving the LSJMJ basis for f^", numE, " ..."];
1252    basis    = BasisLSJMJ[numE];
1253
1254    Which[refParamsVintage === Automatic,
1255      (
1256        PrintFun["Using the automatic vintage with freshly fitted
        free-ion parameters and others as in LaF3 ..."];
1257        lnParams      = LoadLaF3Parameters[ln];
1258        freeIonSol    = FreeIonSolver[expData, numE];
1259        freeIonParams = freeIonSol["bestParams"];
1260        lnParams      = Join[lnParams, freeIonParams];
1261      ),
1262      MemberQ[{List, Association}, Head[RefParams]],
1263      (
1264        RefParams = Association[RefParams];
1265        PrintFun["Using the given parameters as a starting point ..."
        ];
1266        lnParams    = RefParams;
1267        extraParams = LoadLaF3Parameters[ln];
1268        lnParams    = Join[extraParams, lnParams];
1269      ),
1270      True,
1271      (
1272        (* get the reference parameters from the given vintage *)
```

```
1273          PrintFun["Getting reference free-ion parameters for ", ln, "
      using ", refParamsVintage, " ..."];
1274          lnParams = ParamPad[RefParams[ln], "PrintFun" -> PrintFun];
1275        )
1276      ];
1277      freeBies = Prepend[Values[(#->(#/.lnParams)) &/@ freeIonSymbols],
        numE];
1278      (* a more explicit alias *)
1279      allVars            = reducedModelSymbols;
1280      numericConstraints = Association@Select[constraints, NumericQ
      [#[[2]]] &];
1281      standardValues     = allVars /. Join[lnParams, numericConstraints
      ];
1282      solCompendium["allVars"]  = allVars;
1283      solCompendium["freeBies"] = freeBies;
1284
1285      (* reload compiled version if found *)
1286      varHash                   = Hash[{numE, allVars, freeBies,
      truncationEnergy, simplifier}];
1287      compiledIntermediateFname = ln <> "-compiled-intermediate-
      truncated-ham-" <> ToString[varHash] <> ".mx";
1288      compiledIntermediateFname = FileNameJoin[{moduleDir, "compiled",
      compiledIntermediateFname}];
1289      solCompendium["compiledIntermediateFname"] =
      compiledIntermediateFname;
1290
1291      If[FileExistsQ[compiledIntermediateFname],
1292        PrintFun["This ion, free-ion params, and full set of variables
      have been used before (as determined by {numE, allVars, freeBies,
      truncationEnergy, simplifier}). Loading the previously saved
      compiled function and intermediate coupling basis ..."];
1293        PrintFun["Using : ", compiledIntermediateFname];
1294        {compileIntermediateTruncatedHam, truncatedIntermediateBasis} =
       Import[compiledIntermediateFname];,
1295      (
1296        If[truncationEnergy == Infinity,
1297        (
1298            ham = HamMatrixAssembly[numE, "ReturnInBlocks" -> False];
1299            theSimplifier = simplifier;
1300            ham = Normal@ReplaceInSparseArray[ham, simplifier];
1301            PrintFun["Compiling a function for the Hamiltonian with no
      truncation ..."];
1302            (* compile a function that will calculate the truncated
      Hamiltonian given the parameters in allVars, this is the function
      to be use in fitting *)
1303            compileIntermediateTruncatedHam = Compile[Evaluate[allVars
      ], Evaluate[ham]];
1304            truncatedIntermediateBasis = SparseArray@IdentityMatrix[
      Binomial[14, numE]];
1305            (* save the compiled function *)
1306            PrintFun["Saving the compiled function for the Hamiltonian
      with no truncation and a placeholder intermediate basis ..."];
1307            Export[compiledIntermediateFname, {
      compileIntermediateTruncatedHam, truncatedIntermediateBasis}];
1308        ),
1309        (
1310          (* grab the Hamiltonian preserving the block structure *)
1311          PrintFun["Assembling the Hamiltonian for f^",numE," keeping
      the block structure ..."];
1312          ham           = HamMatrixAssembly[numE, "ReturnInBlocks"->True
      ];
1313          (* apply the simplifier *)
1314          PrintFun["Simplifying using the aggregate set of
      simplification rules ..."];
1315          ham           = Map[ReplaceInSparseArray[#, simplifier]&, ham,
       {2}];
1316          PrintFun["Zeroing out every symbol in the Hamiltonian that is
       not a free-ion parameter ..."];
1317          (* Get the free ion symbols *)
1318          freeIonSimplifier = (#->0) & /@ Complement[
      reducedModelSymbols, freeIonSymbols];
1319          (* Take the diagonal blocks for the intermediate analysis *)
1320          PrintFun["Grabbing the diagonal blocks of the Hamiltonian ...
      "];
1321          diagonalBlocks       = Diagonal[ham];
1322          (* simplify them to only keep the free ion symbols *)
```

```
1323        PrintFun["Simplifying the diagonal blocks to only keep the
    free ion symbols ..."];
1324        diagonalScalarBlocks = ReplaceInSparseArray[#,
    freeIonSimplifier]&/@diagonalBlocks;
1325        (* these include the MJ quantum numbers, remove that *)
1326        PrintFun["Contracting the basis vectors by removing the MJ
    quantum numbers from the diagonal blocks ..."];
1327        diagonalScalarBlocks = FreeHam[diagonalScalarBlocks, numE];
1328
1329        argsOfTheIntermediateEigensystems         = StringJoin[Riffle
    [Prepend[(ToString[#]<>"v_") & /@ freeIonSymbols,"numE_"],", "]];
1330        argsForEvalInsideOfTheIntermediateSystems = StringJoin[Riffle
    [(ToString[#]<>"v") & /@ freeIonSymbols,", "]];
1331        PrintFun["argsOfTheIntermediateEigensystems = ",
    argsOfTheIntermediateEigensystems];
1332        PrintFun["argsForEvalInsideOfTheIntermediateSystems = ",
    argsForEvalInsideOfTheIntermediateSystems];
1333        PrintFun["(if the following fails, it might help to see if
    the arguments of TheIntermediateEigensystems match the ones shown
    above)"];
1334
1335        (* compile a function that will effectively calculate the
    spectrum of all of the scalar blocks given the parameters of the
    free-ion part of the Hamiltonian *)
1336        (* compile one function for each of the blocks *)
1337        PrintFun["Compiling functions for the diagonal blocks of the
    Hamiltonian ..."];
1338        compiledDiagonal = Compile[Evaluate[freeIonSymbols], Evaluate
    [N[Normal[#]]]]&/@diagonalScalarBlocks;
1339        (* use that to create a function that will calculate the free
    -ion eigensystem *)
1340        TheIntermediateEigensystems[numEv_, F0v_, F2v_, F4v_, F6v_, ζ
    v_] := (
1341          theNumericBlocks = (#[F0v, F2v, F4v, F6v, ζv]&) /@
    compiledDiagonal;
1342          theIntermediateEigensystems = Eigensystem /@
    theNumericBlocks;
1343          Js     = AllowedJ[numEv];
1344          basisJ = BasisLSJMJ[numEv,"AsAssociation"->True];
1345          (* having calculated the eigensystems with the removed
    degeneracies, put the degeneracies back in explicitly *)
1346          elevatedIntermediateEigensystems = MapIndexed[EigenLever
    [#1, 2Js[[#2[[1]]]]+1 ]&, theIntermediateEigensystems];
1347          (* Identify a single MJ to keep *)
1348          pivot          = If[EvenQ[numEv],0,-1/2];
1349          LSJmultiplets = (#[[1]]<>ToString[InputForm[#[[2]]]])&/
    @Select[BasisLSJMJ[numEv],#[[-1]]== pivot &];
1350          (* calculate the multiplet assignments that the
    intermediate basis eigenvectors have *)
1351          needlePosition = 0;
1352          multipletAssignments = Table[
1353            (
1354              J          = Js[[idx]];
1355              eigenVecs = theIntermediateEigensystems[[idx]][[2]];
1356              majorComponentIndices        = Ordering[Abs[#]][[-1]]&/
    @eigenVecs;
1357              majorComponentIndices       += needlePosition;
1358              needlePosition              += Length[
    majorComponentIndices];
1359              majorComponentAssignments    = LSJmultiplets[[#]]&/
    @majorComponentIndices;
1360              (* All of the degenerate eigenvectors belong to the
    same multiplet*)
1361              elevatedMultipletAssignments = ListRepeater[
    majorComponentAssignments, 2J+1];
1362              elevatedMultipletAssignments
1363            ),
1364          {idx, 1, Length[Js]}
1365          ];
1366          (* put together the multiplet assignments and the energies
    *)
1367          freeIenergiesAndMultiplets = Transpose/@Transpose[{First/
    @elevatedIntermediateEigensystems, multipletAssignments}];
1368          freeIenergiesAndMultiplets = Flatten[
    freeIenergiesAndMultiplets, 1];
1369          (* calculate the change of basis matrix using the
```

```
    intermediate coupling eigenvectors *)
1370        basisChanger = BlockDiagonalMatrix[Transpose/@Last/
    @elevatedIntermediateEigensystems];
1371        basisChanger = SparseArray[basisChanger];
1372        Return[{theIntermediateEigensystems, multipletAssignments,
    elevatedIntermediateEigensystems, freeIenergiesAndMultiplets,
    basisChanger}]
1373      );
1374
1375      PrintFun["Calculating the intermediate eigensystems for ",ln,
    " using free-ion params from LaF3 ..."];
1376      (* calculate intermediate coupling basis using the free-ion
    params for LaF3 *)
1377      {theIntermediateEigensystems, multipletAssignments,
    elevatedIntermediateEigensystems, freeIenergiesAndMultiplets,
    basisChanger} = TheIntermediateEigensystems@@freeBies;
1378
1379      (* use that intermediate coupling basis to compile a function
     for the full Hamiltonian *)
1380      allFreeEnergies = Flatten[First/
    @elevatedIntermediateEigensystems];
1381      (* important that the intermediate coupling basis have
    attached energies, which make possible the truncation *)
1382      ordering = Ordering[allFreeEnergies];
1383      (* sort the free ion energies and determine which indices
    should be included in the truncation *)
1384      allFreeEnergiesSorted          = Sort[allFreeEnergies];
1385      {minFreeEnergy, maxFreeEnergy} = MinMax[allFreeEnergies];
1386      (* determine the index at which the energy is equal or larger
     than the truncation energy *)
1387      sortedTruncationIndex = Which[
1388        truncationEnergy > (maxFreeEnergy-minFreeEnergy),
1389        hamDim,
1390        True,
1391        FirstPosition[allFreeEnergiesSorted - Min[
    allFreeEnergiesSorted],x_/;x>truncationEnergy,{0},1][[1]]
1392      ];
1393      (* the actual energy at which the truncation is made *)
1394      roundedTruncationEnergy = allFreeEnergiesSorted[[
    sortedTruncationIndex]];
1395
1396      (* the indices that participate in the truncation *)
1397      truncationIndices = ordering[[;;sortedTruncationIndex]];
1398      PrintFun["Computing the block structure of the change of
    basis array ..."];
1399      blockSizes         = BlockArrayDimensionsArray[ham];
1400      basisChangerBlocks = ArrayBlocker[basisChanger, blockSizes];
1401      blockShifts        = First /@ Diagonal[blockSizes];
1402      numBlocks          = Length[blockSizes];
1403      (* using the ham (with all the symbols) change the basis to
    the computed one *)
1404      PrintFun["Changing the basis of the Hamiltonian to the
    intermediate coupling basis ..."];
1405      intermediateHam = BlockMatrixMultiply[ham, basisChangerBlocks
    ];
1406      PrintFun["Distributing products inside of symbolic matrix
    elements to keep complexity in check ..."];
1407      Do[
1408        intermediateHam[[rowIdx, colIdx]] = MapToSparseArray[
    intermediateHam[[rowIdx, colIdx]], Distribute /@ # &];,
1409        {rowIdx, 1, numBlocks},
1410        {colIdx, 1, numBlocks}
1411      ];
1412      intermediateHam = BlockMatrixMultiply[BlockTranspose[
    basisChangerBlocks],intermediateHam];
1413      PrintFun["Distributing products inside of symbolic matrix
    elements to keep complexity in check ..."];
1414      Do[
1415        intermediateHam[[rowIdx, colIdx]] = MapToSparseArray[
    intermediateHam[[rowIdx, colIdx]], Distribute /@ # &];,
1416        {rowIdx, 1, numBlocks},
1417        {colIdx, 1, numBlocks}
1418      ];
1419      (* using the truncation indices truncate that one *)
1420      PrintFun["Truncating the Hamiltonian ..."];
1421      truncatedIntermediateHam = TruncateBlockArray[intermediateHam
```

```
, truncationIndices, blockShifts];
    (* these are the basis vectors for the truncated hamiltonian *)
    PrintFun["Saving the truncated intermediate basis ..."];
    truncatedIntermediateBasis = basisChanger[[All, truncationIndices]];

    PrintFun["Compiling a function for the truncated Hamiltonian ..."];
    (* compile a function that will calculate the truncated Hamiltonian given the parameters in allVars, this is the function to be use in fitting *)
    compileIntermediateTruncatedHam = Compile[Evaluate[allVars], Evaluate[truncatedIntermediateHam]];
    (* save the compiled function *)
    PrintFun["Saving the compiled function for the truncated Hamiltonian and the truncated intermediate basis ..."];
    Export[compiledIntermediateFname, {compileIntermediateTruncatedHam, truncatedIntermediateBasis}];
  )
];
)
];

truncationUmbral = Dimensions[truncatedIntermediateBasis][[2]];
PrintFun["The truncated Hamiltonian has a dimension of ", truncationUmbral, "x", truncationUmbral, " ..."];
presentDataIndices = Select[presentDataIndices, # <= truncationUmbral &];
solCompendium["presentDataIndices"] = presentDataIndices;

(* the problemVars are the symbols that will be fitted for *)

PrintFun["Starting up the fitting process using the Levenberg-Marquardt method ..."];
(* using the problemVars I need to create the argument list including _?NumericQ *)
problemVarsQ       = (ToString[#] <> "_?NumericQ") & /@ problemVars;
problemVarsQString = StringJoin[Riffle[problemVarsQ, ", "]];
(* we also need to have the padded arguments with the variables in the right position and the fixed values in the remaining ones *)
problemVarsPositions = Position[allVars, #][[1, 1]] & /@ problemVars;
problemVarsString    = StringJoin[Riffle[ToString /@ problemVars, ", "]];
(* to feed parameters to the Hamiltonian, which includes all parameters, we need to form the set of arguments, with fixed values where needed, and the variables in the right position *)
varsWithConstants                          = standardValues;
varsWithConstants[[problemVarsPositions]] = problemVars;
varsWithConstantsString                    = ToString[varsWithConstants];

(* this following function serves eigenvalues from the Hamiltonian, has memoization so it might grow to use a lot of RAM *)
Clear[HamSortedEigenvalues];
hamEigenvaluesTemplate = StringTemplate["
HamSortedEigenvalues[`problemVarsQ`]:=(
  ham         = compileIntermediateTruncatedHam@@`varsWithConstants`;
  eigenValues = Chop[Sort@Eigenvalues@ham];
  eigenValues = eigenValues - Min[eigenValues];
  HamSortedEigenvalues[`problemVarsString`] = eigenValues;
  Return[eigenValues]
)"];
hamString = hamEigenvaluesTemplate[<|
    "problemVarsQ"      -> problemVarsQString,
    "varsWithConstants" -> varsWithConstantsString,
    "problemVarsString" -> problemVarsString
    |>];
ToExpression[hamString];

(* we also need a function that will pick the i-th eigenvalue,
```

```
    this seems unnecessary but it's needed to form the right
    functional form expected by the Levenberg-Marquardt method *)
1474  eigenvalueDispenserTemplate = StringTemplate["
1475  PartialHamEigenvalues[`problemVarsQ`][i_]:=(
1476    eigenVals = HamSortedEigenvalues[`problemVarsString`];
1477    eigenVals[[i]]
1478  )
1479  "];
1480  eigenValueDispenserString = eigenvalueDispenserTemplate[<|
1481      "problemVarsQ"      -> problemVarsQString,
1482      "problemVarsString" -> problemVarsString
1483      |>];
1484  ToExpression[eigenValueDispenserString];
1485
1486  PrintFun["Determining the free variables after constraints ..."];
1487  constrainedProblemVars     = (problemVars /. constraints);
1488  constrainedProblemVarsList = Variables[constrainedProblemVars];
1489  If[addShift,
1490    PrintFun["Adding a constant shift to the fitting parameters ...
    "];
1491    constrainedProblemVarsList = Append[constrainedProblemVarsList,
     \[Epsilon]]
1492  ];
1493
1494  indepVars = Complement[problemVars, #[[1]] & /@ constraints];
1495  stringPartialVars = ToString/@constrainedProblemVarsList;
1496
1497  paramSols  = {};
1498  rmsHistory = {};
1499  steps      = 0;
1500  problemVarsWithStartValues = KeyValueMap[{#1,#2} &, startValues];
1501  If[addShift,
1502    problemVarsWithStartValues = Append[problemVarsWithStartValues,
     {\[Epsilon] ,0}];
1503  ];
1504  openNotebooks     = If[runningInteractive,
1505            ("WindowTitle"/.NotebookInformation[#]) & /@ Notebooks
    [],
1506            {}];
1507  If[Not[MemberQ[openNotebooks,"Solver Progress"]] && OptionValue["
    ProgressView"],
1508    ProgressNotebook["Basic"->False]
1509  ];
1510  degressOfFreedom = Length[presentDataIndices] - Length[
    problemVars] - 1;
1511  PrintFun["Fitting for ", Length[presentDataIndices], " data
    points with ", Length[problemVars], " free parameters.", " The
    effective degrees of freedom are ", degressOfFreedom, " ..."];
1512
1513  PrintFun["Fitting model to data ..."];
1514  startTime   = Now;
1515  shiftToggle = If[addShift, 1, 0];
1516  sol = FindMinimum[
1517    Sum[(expData[[j]][[1]] - (PartialHamEigenvalues @@
    constrainedProblemVars)[j] - shiftToggle * \[Epsilon])^2,
1518    {j, presentDataIndices}],
1519    problemVarsWithStartValues,
1520    Method        -> "LevenbergMarquardt",
1521    MaxIterations -> OptionValue["MaxIterations"],
1522    AccuracyGoal  -> OptionValue["AccuracyGoal"],
1523    StepMonitor :> (
1524      steps      += 1;
1525      currentSqSum = Sum[(expData[[j]][[1]] - (
    PartialHamEigenvalues @@ constrainedProblemVars)[j] - shiftToggle
    * \[Epsilon])^2, {j, presentDataIndices}];
1526      currentRMS = Sqrt[currentSqSum / degressOfFreedom];
1527      paramSols  = AddToList[paramSols, constrainedProblemVarsList,
     maxHistory];
1528      rmsHistory = AddToList[rmsHistory, currentRMS, maxHistory];
1529      )
1530    ];
1531  endTime = Now;
1532  timeTaken = QuantityMagnitude[endTime - startTime, "Seconds"];
1533  PrintFun["Solution found in ", timeTaken, "s"];
1534
1535  solVec = constrainedProblemVars /. sol[[-1]];
```

```
1536    indepSolVec = indepVars /. sol[[-1]];
1537    If[addShift,
1538      \[Epsilon]Best = \[Epsilon]/. sol[[-1]],
1539      \[Epsilon]Best = 0
1540    ];
1541    fullSolVec = standardValues;
1542    fullSolVec[[problemVarsPositions]] = solVec;
1543    PrintFun["Calculating the truncated numerical Hamiltonian
        corresponding to the solution ..."];
1544    fullHam = compileIntermediateTruncatedHam @@ fullSolVec;
1545    PrintFun["Calculating energies and eigenvectors ..."];
1546    {eigenEnergies, eigenVectors} = Eigensystem[fullHam];
1547    states = Transpose[{eigenEnergies, eigenVectors}];
1548    states = SortBy[states, First];
1549    eigenEnergies = First /@ states;
1550    PrintFun["Shifting energies to make ground state zero of energy
        ..."];
1551    eigenEnergies = eigenEnergies - eigenEnergies[[1]];
1552    PrintFun["Calculating the linear approximant to each eigenvalue
        ..."];
1553    allVarsVec = Transpose[{allVars}];
1554    p0 = Transpose[{fullSolVec}];
1555    linMat = {};
1556    If[addShift,
1557      tail = -2,
1558      tail = -1];
1559    Do[
1560      (
1561        aVarPosition = Position[allVars, aVar][[1, 1]];
1562        isolationValues = ConstantArray[0, Length[allVars]];
1563        isolationValues[[aVarPosition]] = 1;
1564        dependentVars = KeyValueMap[{#1, D[#2, aVar]} &, Association[
        constraints]];
1565        Do[
1566          isolationValues[[Position[allVars, dVar[[1]]][[1, 1]]]] =
        dVar[[2]],
1567          {dVar, dependentVars}
1568        ];
1569        perHam  = compileIntermediateTruncatedHam @@ isolationValues;
1570        lin     = FirstOrderPerturbation[Last /@ states, perHam];
1571        linMat  = Append[linMat, lin];
1572      ),
1573      {aVar, constrainedProblemVarsList[[;;tail]]}
1574    ];
1575    PrintFun["Removing the gradient of the ground state ..."];
1576    linMat = (# - #[[1]] & /@ linMat);
1577    PrintFun["Transposing derivative matrices into columns ..."];
1578    linMat  = Transpose[linMat];
1579
1580    PrintFun["Calculating the eigenvalue vector at solution ..."];
1581    \[Lambda]0Vec = Transpose[{eigenEnergies[[presentDataIndices]]}];
1582    PrintFun["Putting together the experimental vector ..."];
1583    \[Lambda]exp = Transpose[{First /@ expData[[presentDataIndices
        ]]}];
1584    problemVarsVec = If[addShift,
1585      Transpose[{constrainedProblemVarsList[[;;-2]]}],
1586      Transpose[{constrainedProblemVarsList}]
1587    ];
1588    indepSolVecVec = Transpose[{indepSolVec}];
1589    PrintFun["Calculating the difference between eigenvalues at
        solution ..."];
1590    diff = If[linMat == {},
1591      (\[Lambda]0Vec - \[Lambda]exp)  + \[Epsilon]Best,
1592      (\[Lambda]0Vec - \[Lambda]exp)  + \[Epsilon]Best + linMat[[
        presentDataIndices]].(problemVarsVec - indepSolVecVec)
1593    ];
1594    PrintFun["Calculating the sum of squares of differences around
        solution ... "];
1595    sqdiff        = Expand[(Transpose[diff] . diff)[[1, 1]]];
1596    PrintFun["Calculating the minimum (which should coincide with sol
        ) ..."];
1597    minpoly       = sqdiff /. AssociationThread[problemVars -> solVec
        ];
1598    fmSolAssoc    = Association[sol[[2]]];
1599    If[\[Sigma]exp == Automatic,
1600      \[Sigma]exp = Sqrt[minpoly / degressOfFreedom];
```

```
    ];
    \[CapitalDelta]\[Chi]2 = Sqrt[degressOfFreedom];
    Amat = (1/\[Sigma]exp^2) * Transpose[linMat[[presentDataIndices
    ]]].linMat[[presentDataIndices]];
    paramIntervals = EllipsoidBoundingBox[Amat, \[CapitalDelta]\[Chi
    ]2];
    PrintFun["Calculating the uncertainty in the parameters ..."];
    solWithUncertainty = Table[
      (
        aVar        = constrainedProblemVarsList[[varIdx]];
        paramBest   = aVar /. fmSolAssoc;
        (aVar -> {paramBest, paramIntervals[[varIdx, 2]]})
      ),
    {varIdx, 1, Length[constrainedProblemVarsList]-shiftToggle}
    ];

    bestRMS    = Sqrt[minpoly / degressOfFreedom];
    bestParams = sol[[2]];
    bestWithConstraints = Association@Join[constraints, bestParams];
    bestWithConstraints = bestWithConstraints /. bestWithConstraints;
    bestWithConstraints = (# + 0.) & /@ bestWithConstraints;

    solCompendium["degreesOfFreedom"]    = degressOfFreedom;
    solCompendium["solWithUncertainty"]  = solWithUncertainty;
    solCompendium["truncatedDim"]        = truncationUmbral;
    solCompendium["fittedLevels"]        = Length[presentDataIndices
    ];
    solCompendium["actualSteps"]         = steps;
    solCompendium["bestRMS"]             = bestRMS;
    solCompendium["problemVars"]         = problemVars;
    solCompendium["paramSols"]           = paramSols;
    solCompendium["rmsHistory"]          = rmsHistory;
    solCompendium["Appendix"]            = OptionValue["
    AppendToLogFile"];
    solCompendium["timeTaken/s"]         = timeTaken;
    solCompendium["bestParams"]          = bestParams;
    solCompendium["bestParamsWithConstraints"] = bestWithConstraints;

    If[OptionValue["SaveEigenvectors"],
        solCompendium["states"] = {#[[1]] + \[Epsilon]Best, #[[2]]}
    &/@ (Chop /@ ShiftedLevels[states]),
        (
          finalEnergies = Sort[First /@ states];
          finalEnergies = finalEnergies - finalEnergies[[1]];
          finalEnergies = finalEnergies + \[Epsilon]Best;
          finalEnergies  = Chop /@ finalEnergies;
          solCompendium["energies"] = finalEnergies;
      )
    ];
    If[OptionValue["SaveToLog"],
      PrintFun["Saving the solution to the log file ..."];
      LogSol[solCompendium, logFilePrefix];
    ];
    PrintFun["Finished ..."];
    Return[solCompendium];
  )
];


MostlyOrthogonalFit::usage="MostlyOrthogonalFit[numE, expData,
    excludeDataIndices, problemVars, startValues, \[Sigma]exp,
    constraints_List, Options] fits the given expData in an f^numE
    configuration, by using the symbols in problemVars. The symbols
    given in problemVars may be constrained or held constant, this
    being controlled by constraints list which is a list of
    replacement rules expressing desired constraints. The constraints
    list additional constraints imposed upon the model parameters that
     remain once other simplifications have been \"baked\" into the
    compiled Hamiltonians that are used to increase the speed of the
    calculation.

Important, note that in the case of odd number of electrons the given
     data must explicitly include the Kramers degeneracy;
    excludeDataIndices must be compatible with this.

The list expData needs to be a list of lists with the only
```

```
    restriction that the first element of them corresponds to energies
     of levels. In this list, an empty value can be used to indicate
    known gaps in the data. Even if the energy value for a level is
    known (and given in expData) certain values can be omitted from
    the fitting procedure through the list excludeDataIndices, which
    correspond to indices in expData that should be skipped over.
1660
1661 The Hamiltonian used for fitting is version that has been truncated
    either by using the maximum energy given in expData or by manually
     setting a truncation energy using the option \"TruncationEnergy\"
    .
1662
1663 The argument \[Sigma]exp is the estimated uncertainty in the
    differences between the calculated and the experimental energy
    levels. This is used to estimate the uncertainty in the fitted
    parameters. Admittedly this will be a rough estimate (at least on
    the contribution of the calculated uncertainty), but it is better
    than nothing and may at least provide a lower bound to the
    uncertainty in the fitted parameters. It is assumed that the
    uncertainty in the differences between the calculated and the
    experimental energy levels is the same for all of them.
1664
1665 The list startValues is a list with all of the parameters needed to
    define the Hamiltonian (including the initial values for
    problemVars).
1666
1667 The function saves the solution to a file. The file is named with a
    prefix (controlled by the option \"FilePrefix\") and a UUID. The
    file is saved in the log sub-directory as a .m file.
1668
1669 Here's a description of the different parts of this function: first
    the Hamiltonian is assembled and simplified using the given
    simplifications. Then the intermediate coupling basis is
    calculated using the free-ion parameters for the given lanthanide.
     The Hamiltonian is then changed to the intermediate coupling
    basis and truncated. The truncated Hamiltonian is then compiled
    into a function that can be used to calculate the energy levels of
     the truncated Hamiltonian. The function that calculates the
    energy levels is then used to fit the experimental data. The
    fitting is done using FindMinimum with the Levenberg-Marquardt
    method.
1670
1671 The function returns an association with the following keys:
1672
1673 - \"bestRMS\" which is the best \[Sigma] value found.
1674 - \"bestParams\" which is the best set of parameters found for the
    variables that were not constrained.
1675 - \"bestParamsWithConstraints\" which has the best set of parameters
    (from - \"bestParams\") together with the used constraints. These
    include all the parameters in the model, even those that were not
    fitted for.
1676 - \"paramSols\" which is a list of the parameters trajectories during
     the stepping of the fitting algorithm.
1677 - \"timeTaken/s\" which is the time taken to find the best fit.
1678 - \"simplifier\" which is the simplifier used to simplify the
    Hamiltonian.
1679 - \"excludeDataIndices\" as given in the input.
1680 - \"startValues\" as given in the input.
1681
1682 - \"freeIonSymbols\" which are the symbols used in the intermediate
    coupling basis.
1683 - \"truncationEnergy\" which is the energy used to truncate the
    Hamiltonian, if it was set to Automatic, the value here is the
    actual energy used.
1684 - \"numE\" which is the number of electrons in the f^numE
    configuration.
1685 - \"expData\" which is the experimental data used for fitting.
1686 - \"problemVars\" which are the symbols considered for fitting
1687
1688 - \"maxIterations\" which is the maximum number of iterations used by
     NMinimize.
1689 - \"hamDim\" which is the dimension of the full Hamiltonian.
1690 - \"allVars\" which are all the symbols defining the Hamiltonian
    under the aggregate simplifications.
1691 - \"freeBies\" which are the free-ion parameters used to define the
    intermediate coupling basis.
```

```
- \"truncatedDim\" which is the dimension of the truncated Hamiltonian.
- \"compiledIntermediateFname\" the file name of the compiled function used for the truncated Hamiltonian.

- \"fittedLevels\" which is the number of levels fitted for.
- \"actualSteps\" the number of steps that FindMiniminum actually took.
- \"solWithUncertainty\" which is a list of replacement rules of the form (paramSymbol -> {bestEstimate, uncertainty}).
- \"rmsHistory\" which is a list of the \[Sigma] values found during the fitting.
- \"Appendix\" which is an association appended to the log file under the key \"Appendix\".
- \"presentDataIndices\" which is the list of indices in expData that were used for fitting, this takes into account both the empty indices in expData and also the indices in excludeDataIndices.

- \"states\" which contains a list of eigenvalues and eigenvectors for the fitted model, this is only available if the option \"SaveEigenvectors\" is set to True; if a general shift of energy was allowed for in the fitting, then the energies are shifted accordingly.
- \"energies\" which is a list of the energies of the fitted levels, this is only available if the option \"SaveEigenvectors\" is set to False. If a general shift of energy was allowed for in the fitting, then the energies are shifted accordingly.
- \"degreesOfFreedom\" which is equal to the nunber of fitted state energies minus the number of parameters used in fitting.

The function admits the following options with corresponding default values:
- \"MaxHistory\" : determines how long the logs for the solver can be.
- \"MaxIterations\": determines the maximum number of iterations used by NMinimize.
- \"FilePrefix\" : the prefix to use for the subfolder in the log filder, in which the solution files are saved, by default this is \"calcs\" so that the calculation files are saved under the directory \"log/calcs\".
- \"AddConstantShift\" : if True then a constant shift is allowed in the fitting, default is False. If this is the case the variable \"\[Epsilon]\" is added to the list of variables to be fitted for, it must not be included in problemVars.

- \"AccuracyGoal\": the accuracy goal used by NMinimize, default of 5.
- \"TrucationEnergy\": if Automatic then the maximum energy in expData is taken, else it takes the value set by this option. In all cases the energies in expData are only considered up to this value.
- \"PrintFun\": the function used to print progress messages, the default is PrintTemporary.
- \"RefParamsVintage\": the vintage of the reference parameters to use. The reference parameters are both used to determine the truncated Hamiltonian, and also as starting values for the solver. It may be \"LaF3\", in which case reference parameters from Carnall are used. It may also be \"LiYF4\", in which case the reference parameters from the LiYF4 paper are used. It may also be Automatic, in which case the given experimental data is used to determine starting values for F^k and ζ. It may also be a list or association that provides values for the Slater integrals and spin-orbit coupling, the remaining necessary parameters complemented by using \"LaF3\".

- \"ProgressView\": whether or not a progress window will be opened to show the progress of the solver, the default is True.
- \"SignatureCheck\": if True then then the function returns prematurely, returning a list with the symbols that would have defined the Hamiltonian after all simplifications have been applied. Useful to check the entire parameter set that the Hamiltonian has.
- \"SaveEigenvectors\": if True then the both the eigenvectors and eigenvalues are saved under the \"states\" key of the returned association. If False then only the energies are saved, the default is False.
```

```
1720
1721 - \"AppendToLogFile\": an association appended to the log file under
       the key \"Appendix\".
1722 - \"MagneticSimplifier\": a list of replacement rules to simplify the
       Marvin and pesudo-magnetic paramters. Here the ratios of the
       Marvin parameters and the pseudo-magnetic parameters are defined
       to simplify the magnetic part of the Hamiltonian.
1723 - \"MagFieldSimplifier\": a list of replacement rules to specify a
       magnetic field (in T), if set to {}, then {Bx, By, Bz} can also be
       used as variables to be fitted for.
1724
1725 - \"SymmetrySimplifier\": a list of replacements rules to simplify
       the crystal field.
1726 - \"OtherSimplifier\": an additional list of replacement rules that
       are applied to the Hamiltonian before computing with it. Here the
       spin-spin contribution can be turned off by setting \[Sigma]SS->0,
       which is the default.
1727 ";
1728 Options[MostlyOrthogonalFit] = {
1729   "MaxHistory"       -> 200,
1730   "MaxIterations"    -> 100,
1731   "FilePrefix"       -> "calcs",
1732   "ProgressView"     -> True,
1733   "TruncationEnergy" -> Automatic,
1734   "AccuracyGoal"     -> 5,
1735   "PrintFun"         -> PrintTemporary,
1736   "RefParamsVintage" -> "LaF3",
1737   "SignatureCheck"   -> False,
1738   "AddConstantShift" -> False,
1739   "SaveEigenvectors" -> False,
1740   "AppendToLogFile"  -> <||>,
1741   "SaveToLog"        -> False,
1742   "Energy Uncertainty in K" -> Automatic,
1743   "MagneticSimplifier" -> {
1744     M2 -> 56/100 M0,
1745     M4 -> 31/100 M0,
1746     P4 -> 1/2 P2,
1747     P6 -> 1/10 P2
1748     },
1749   "MagFieldSimplifier" -> {
1750     Bx -> 0,
1751     By -> 0,
1752     Bz -> 0
1753     },
1754   "SymmetrySimplifier" -> {
1755     B12->0, B14->0, B16->0, B34->0, B36->0, B56->0,
1756     S12->0 ,S14->0, S16->0, S22->0, S24->0, S26->0,
1757     S34->0 ,S36->0, S44->0, S46->0, S56->0, S66->0
1758     },
1759   "OtherSimplifier" -> {
1760     E0p->0,
1761     P0->0,
1762     \[Sigma]SS->0,
1763     T11p->0,
1764     T12->0,
1765     T14->0,
1766     T15->0,
1767     T16->0,
1768     T18->0,
1769     T17->0,
1770     T19->0,
1771     wChErrA ->0,
1772     wChErrB->0
1773     },
1774   "ThreeBodySimplifier" -> <|
1775     1  -> {
1776       T2->0,
1777       T3->0,
1778       T4->0,
1779       T6->0,
1780       T7->0,
1781       T8->0,
1782       T11p->0,
1783       T12->0,
1784       T14->0,
1785       T15->0,
```

```
      T16->0,
      T18->0,
      T17->0,
      T19->0,
      T2p->0},
    2 -> {
      T2->0,
      T3->0,
      T4->0,
      T6->0,
      T7->0,
      T8->0,
      T11p->0,
      T12->0,
      T14->0,
      T15->0,
      T16->0,
      T18->0,
      T17->0,
      T19->0,
      T2p->0
      },
    3  -> {t2Switch -> 1},
    4  -> {t2Switch -> 1},
    5  -> {t2Switch -> 1},
    6  -> {t2Switch -> 1},
    7  -> {t2Switch -> 1},
    8  -> {t2Switch -> 0},
    9  -> {t2Switch -> 0},
    10 -> {t2Switch -> 0},
    11 -> {t2Switch -> 0},
    12 -> {
      t2Switch -> 0,
      T2->0,
      T2p->0,
      T3->0,
      T4->0,
      T6->0,
      T7->0,
      T8->0,
      T11p->0,
      T12->0,
      T14->0,
      T15->0,
      T16->0,
      T18->0,
      T17->0,
      T19->0
      },
    13->{
      t2Switch -> 0,
      T2->0,
      T2p->0,
      T3->0,
      T4->0,
      T6->0,
      T7->0,
      T8->0,
      T11p->0,
      T12->0,
      T14->0,
      T15->0,
      T16->0,
      T18->0,
      T17->0,
      T19->0
      }
    |>,
    "FreeIonSymbols" -> {E0p, E1p, E2p, E3p, ζ}
};
MostlyOrthogonalFit[numE_Integer, expData_List,
    excludeDataIndices_List, problemVars_List, startValues_Association
    , constraints_List, OptionsPattern[]]:=Module[
  {accuracyGoal,
  allFreeEnergies,
  allFreeEnergiesSorted,
```

```
  allVars,
  allVarsVec,
  argsForEvalInsideOfTheIntermediateSystems,
  argsOfTheIntermediateEigensystems,
  aVar,
  aVarPosition,
  basis,

  basisChanger,
  basisChangerBlocks,
  bestParams,
  bestRMS,
  blockShifts,
  blockSizes,
  compiledDiagonal,
  compiledIntermediateFname,
  constrainedProblemVars,
  constrainedProblemVarsList,

  currentRMS,
  degressOfFreedom,
  dependentVars,
  diagonalBlocks,
  diagonalScalarBlocks,
  diff,
  eigenEnergies,
  eigenvalueDispenserTemplate,
  eigenVectors,
  elevatedIntermediateEigensystems,

  endTime,
  fmSolAssoc,
  freeBies,
  freeIenergiesAndMultiplets,
  fullHam,
  fullSolVec,
  ham,
  hamDim,
  hamEigenvaluesTemplate,
  hamString,

  indepSolVecVec,
  indepVars,
  intermediateHam,
  isolationValues,
  lin,
  linMat,
  ln,
  lnParams,
  logFilePrefix,
  magneticSimplifier,

  maxFreeEnergy,
  maxHistory,
  maxIterations,
  minFreeEnergy,
  minpoly,
  modelSymbols,
  multipletAssignments,
  needlePosition,
  numBlocks,
  openNotebooks,

  ordering,
  otherSimplifier,
  p0,
  paramBest,
  perHam,
  presentDataIndices,
  PrintFun,
  problemVarsPositions,
  problemVarsQ,
  problemVarsQString,

  problemVarsVec,
  problemVarsWithStartValues,
```

```
  reducedModelSymbols,
  RefParams,
  roundedTruncationEnergy,
  runningInteractive,
  shiftToggle,
  simplifier,
  sol,
  solCompendium,

  solWithUncertainty,
  sortedTruncationIndex,
  sqdiff,
  standardValues,
  startTime,
  states,
  steps,
  symmetrySimplifier,
  theIntermediateEigensystems,
  TheIntermediateEigensystems,

  timeTaken,
  truncatedIntermediateBasis,
  truncatedIntermediateHam,
  truncationEnergy,
  truncationIndices,
  truncationUmbral,
  varHash,
  varsWithConstants,
  \[Lambda]0Vec,
  \[Lambda]exp
},
  (
    \[Sigma]exp   = OptionValue["Energy Uncertainty in K"];
    refParamsVintage = OptionValue["RefParamsVintage"];
    RefParams = Which[
      refParamsVintage === "LaF3",
      LoadLaF3Parameters,
      refParamsVintage === "LiYF4",
      LoadLiYF4Parameters,
      True,
      refParamsVintage
    ];
    solCompendium = <||>;
    hamDim        = Binomial[14, numE];
    addShift      = OptionValue["AddConstantShift"];
    ln            = theLanthanides[[numE]];
    maxHistory    = OptionValue["MaxHistory"];
    maxIterations = OptionValue["MaxIterations"];
    logFilePrefix = If[OptionValue["FilePrefix"] == "",
                       ToString[theLanthanides[[numE]]],
                       OptionValue["FilePrefix"]
                     ];
    accuracyGoal = OptionValue["AccuracyGoal"];
    PrintFun     = OptionValue["PrintFun"];
    freeIonSymbols      = OptionValue["FreeIonSymbols"];
    runningInteractive  = (Head[$ParentLink] === LinkObject);
    magneticSimplifier  = OptionValue["MagneticSimplifier"];
    magFieldSimplifier  = OptionValue["MagFieldSimplifier"];
    symmetrySimplifier  = OptionValue["SymmetrySimplifier"];
    otherSimplifier     = OptionValue["OtherSimplifier"];
    threeBodySimplifier = If[Head[OptionValue["ThreeBodySimplifier"]]
     == Association,
                             OptionValue["ThreeBodySimplifier"][numE],
                             OptionValue["ThreeBodySimplifier"]
                           ];

    truncationEnergy = If[OptionValue["TruncationEnergy"] ===
    Automatic,
      (
        PrintFun["Truncation energy set to Automatic, using the
    maximum energy (+20%) in the data ..."];
        Round[1.2 * Max[Select[First /@ expData, NumericQ[#] &]]]
      ),
      OptionValue["TruncationEnergy"]
      ];
    truncationEnergy = Max[50000, truncationEnergy];
```

```
    PrintFun["Using a truncation energy of ", truncationEnergy, " K"
];

    simplifier = Join[magneticSimplifier,
                      magFieldSimplifier,
                      symmetrySimplifier,
                      threeBodySimplifier,
                      otherSimplifier];

    PrintFun["Determining gaps in the data ..."];
    (* whatever is non-numeric is assumed as a known gap *)
    presentDataIndices = Flatten[Position[expData, {_?(NumericQ[#] &)
, ___}]];
    (* some indices omitted here based on the excludeDataIndices
    argument, note that presentDataIndices is somewhat a misnomer*)
    presentDataIndices = Complement[presentDataIndices,
    excludeDataIndices];

    solCompendium["simplifier"]         = simplifier;
    solCompendium["excludeDataIndices"] = excludeDataIndices;
    solCompendium["startValues"]        = startValues;
    solCompendium["freeIonSymbols"]     = freeIonSymbols;
    solCompendium["truncationEnergy"]   = truncationEnergy;
    solCompendium["numE"]               = numE;
    solCompendium["expData"]            = expData;
    solCompendium["problemVars"]        = problemVars;
    solCompendium["maxIterations"]      = maxIterations;
    solCompendium["hamDim"]             = hamDim;
    solCompendium["constraints"]        = constraints;

    modelSymbols  = Select[paramSymbols,
      Not[MemberQ[Join[racahSymbols,
      juddOfeltIntensitySymbols,
      slaterSymbols,
      {α, β, γ},
      {T2},
      chenSymbols,
      {t2Switch, \[Epsilon], gs, nE}],#]]&
    ];
    modelSymbols = Sort[modelSymbols];
    (* remove the symbols that will be removed by the simplifier, no
    symbol should remain here that is not in the symbolic Hamiltonian
    *)
    reducedModelSymbols = Select[modelSymbols, Not[MemberQ[Keys[
    simplifier],#]]&];

    (* this is useful to understand what are the arguments of the
    truncated compiled Hamiltonian *)
    If[OptionValue["SignatureCheck"],
      (
        PrintFun["Given the model parameters and the simplifying
    assumptions, the resultant model parameters are:"];
        PrintFun[{reducedModelSymbols}];
        PrintFun["Exiting ..."];
        Return[""];
      )
    ];

    (* calculate the basis *)
    PrintFun["Retrieving the LSJMJ basis for f^", numE, " ..."];
    basis    = BasisLSJMJ[numE];

    Which[refParamsVintage === Automatic,
      (
        PrintFun["This is not currently supported, please provide a
    vintage for the reference parameters."];
        Return[$Failed];
      ),
      MemberQ[{List, Association}, Head[RefParams]],
      (
        RefParams = Association[RefParams];
        PrintFun["Using the given parameters as a starting point ..."
];
        lnParams    = RefParams;
        extraParams = FromNonOrthogonalToMostlyOrthogonal[
    LoadLaF3Parameters[ln], numE];
```

```
2073          lnParams    = Join[extraParams, lnParams];
2074        ),
2075        True,
2076        (
2077          (* get the reference parameters from the given vintage *)
2078          PrintFun["Getting reference free-ion parameters for ", ln, "
    using ", refParamsVintage, " ..."];
2079          lnParams = ParamPad[FromNonOrthogonalToMostlyOrthogonal[
    RefParams[ln], numE],
2080            "PrintFun" -> PrintFun];
2081        )
2082      ];
2083
2084      freeBies = Prepend[Values[(# -> (# /. lnParams)) &/@
    freeIonSymbols],  numE];
2085
2086      (* a more explicit alias *)
2087      allVars            = reducedModelSymbols;
2088      numericConstraints = Association@Select[constraints, NumericQ
    [#[[2]]] &];
2089      standardValues     = allVars /. Join[lnParams, numericConstraints
    ];
2090      solCompendium["allVars"]  = allVars;
2091      solCompendium["freeBies"] = freeBies;
2092
2093      (* reload compiled version if found *)
2094      varHash                   = Hash[{numE, allVars, freeBies,
    truncationEnergy, simplifier}];
2095      compiledIntermediateFname = ln <> "-compiled-intermediate-
    truncated-ham-" <> ToString[varHash] <> ".mx";
2096      compiledIntermediateFname = FileNameJoin[{moduleDir, "compiled",
    compiledIntermediateFname}];
2097      solCompendium["compiledIntermediateFname"] =
    compiledIntermediateFname;
2098
2099      If[FileExistsQ[compiledIntermediateFname],
2100      (
2101        PrintFun["This ion, free-ion params, and full set of variables
    have been used before (as determined by {numE, allVars, freeBies,
    truncationEnergy, simplifier}). Loading the previously saved
    compiled function and intermediate coupling basis ..."];
2102        PrintFun["Using : ", compiledIntermediateFname];
2103        {compileIntermediateTruncatedHam, truncatedIntermediateBasis} =
     Import[compiledIntermediateFname];
2104      ),
2105      (
2106        If[truncationEnergy == Infinity,
2107        (
2108            ham = HamMatrixAssembly[numE,
2109                  "ReturnInBlocks" -> False,
2110                  "OperatorBasis" -> "MostlyOrthogonal"];
2111            theSimplifier = simplifier;
2112            ham = Normal @ ReplaceInSparseArray[ham, simplifier];
2113            PrintFun["Compiling a function for the Hamiltonian with no
    truncation ..."];
2114            (* compile a function that will calculate the truncated
    Hamiltonian given the parameters in allVars, this is the function
    to be use in fitting *)
2115            compileIntermediateTruncatedHam = Compile[Evaluate[allVars
    ], Evaluate[ham]];
2116            truncatedIntermediateBasis      =
    SparseArray@IdentityMatrix[Binomial[14, numE]];
2117            (* save the compiled function *)
2118            PrintFun["Saving the compiled function for the Hamiltonian
    with no truncation and a placeholder intermediate basis ..."];
2119            Export[compiledIntermediateFname, {
    compileIntermediateTruncatedHam, truncatedIntermediateBasis}];
2120        ),
2121        (
2122          (* grab the Hamiltonian preserving the block structure *)
2123          PrintFun["Assembling the Hamiltonian for f^", numE, " keeping
     the block structure ..."];
2124          ham           = HamMatrixAssembly[numE,
2125                            "ReturnInBlocks"->True,
2126                            "OperatorBasis"->"MostlyOrthogonal"];
2127          (* apply the simplifier *)
```

```
2128        PrintFun["Simplifying using the aggregate set of
    simplification rules ..."];
2129        ham           = Map[ReplaceInSparseArray[#, simplifier]&, ham,
     {2}];
2130        PrintFun["Zeroing out every symbol in the Hamiltonian that is
     not a free-ion parameter ..."];
2131
2132        (* Get the free ion symbols *)
2133        freeIonSimplifier = (#->0) & /@ Complement[
    reducedModelSymbols, freeIonSymbols];
2134
2135        (* Take the diagonal blocks for the intermediate analysis *)
2136        PrintFun["Grabbing the diagonal blocks of the Hamiltonian ...
    "];
2137        diagonalBlocks        = Diagonal[ham];
2138
2139        (* simplify them to only keep the free ion symbols *)
2140        PrintFun["Simplifying the diagonal blocks to only keep the
    free ion symbols ..."];
2141        diagonalScalarBlocks = ReplaceInSparseArray[#,
    freeIonSimplifier] &/@ diagonalBlocks;
2142
2143        (* these include the MJ quantum numbers, remove that *)
2144        PrintFun["Contracting the basis vectors by removing the MJ
    quantum numbers from the diagonal blocks ..."];
2145        diagonalScalarBlocks = FreeHam[diagonalScalarBlocks, numE];
2146
2147        argsOfTheIntermediateEigensystems          = StringJoin[Riffle
    [Prepend[(ToString[#]<>"v_") & /@ freeIonSymbols,"numE_"],", "]];
2148        argsForEvalInsideOfTheIntermediateSystems = StringJoin[Riffle
    [(ToString[#]<>"v") & /@ freeIonSymbols,", "]];
2149        PrintFun["argsOfTheIntermediateEigensystems = ",
    argsOfTheIntermediateEigensystems];
2150        PrintFun["argsForEvalInsideOfTheIntermediateSystems = ",
    argsForEvalInsideOfTheIntermediateSystems];
2151        PrintFun["(if the following fails, it might help to see if
    the arguments of TheIntermediateEigensystems match the ones shown
    above)"];
2152
2153        (* compile a function that will effectively calculate the
    spectrum of all of the scalar blocks given the parameters of the
    free-ion part of the Hamiltonian *)
2154        (* compile one function for each of the blocks *)
2155        PrintFun["Compiling functions for the diagonal blocks of the
    Hamiltonian ..."];
2156        compiledDiagonal = Compile[Evaluate[freeIonSymbols], Evaluate
    [N[Normal[#]]]]& /@ diagonalScalarBlocks;
2157        (* use that to create a function that will calculate the free
    -ion eigensystem *)
2158        TheIntermediateEigensystems[numEv_, E0pv_, E1pv_, E2pv_,
    E3pv_, ζv_] := (
2159          theNumericBlocks = (#[E0pv, E1pv, E2pv, E3pv, ζv]&) /@
    compiledDiagonal;
2160          theIntermediateEigensystems = Eigensystem /@
    theNumericBlocks;
2161          Js     = AllowedJ[numEv];
2162          basisJ = BasisLSJMJ[numEv, "AsAssociation"->True];
2163          (* having calculated the eigensystems with the removed
    degeneracies, put the degeneracies back in explicitly *)
2164          elevatedIntermediateEigensystems = MapIndexed[EigenLever
    [#1, 2Js[[#2[[1]]]]+1 ]&, theIntermediateEigensystems];
2165          (* Identify a single MJ to keep *)
2166          pivot          = If[EvenQ[numEv],0,-1/2];
2167          LSJmultiplets = (#[[1]]<>ToString[InputForm[#[[2]]]])&/
    @Select[BasisLSJMJ[numEv],#[[-1]]== pivot &];
2168          (* calculate the multiplet assignments that the
    intermediate basis eigenvectors have *)
2169          needlePosition = 0;
2170          multipletAssignments = Table[
2171            (
2172              J          = Js[[idx]];
2173              eigenVecs = theIntermediateEigensystems[[idx]][[2]];
2174              majorComponentIndices         = Ordering[Abs[#]][[-1]]&/
    @eigenVecs;
2175              majorComponentIndices       += needlePosition;
2176              needlePosition              += Length[
```

```
    majorComponentIndices];
2177            majorComponentAssignments    = LSJmultiplets[[#]]&/
    @majorComponentIndices;
2178            (* All of the degenerate eigenvectors belong to the
    same multiplet*)
2179            elevatedMultipletAssignments = ListRepeater[
    majorComponentAssignments, 2J+1];
2180            elevatedMultipletAssignments
2181          ),
2182        {idx, 1, Length[Js]}
2183        ];
2184
2185        (* put together the multiplet assignments and the energies
    *)
2186        freeIenergiesAndMultiplets = Transpose/@Transpose[{First/
    @elevatedIntermediateEigensystems, multipletAssignments}];
2187        freeIenergiesAndMultiplets = Flatten[
    freeIenergiesAndMultiplets, 1];
2188        (* calculate the change of basis matrix using the
    intermediate coupling eigenvectors *)
2189        basisChanger = BlockDiagonalMatrix[Transpose/@Last/
    @elevatedIntermediateEigensystems];
2190        basisChanger = SparseArray[basisChanger];
2191        Return[{theIntermediateEigensystems,
2192          multipletAssignments,
2193          elevatedIntermediateEigensystems,
2194          freeIenergiesAndMultiplets,
2195          basisChanger}]
2196      );
2197
2198      PrintFun["Calculating the intermediate eigensystems for ",ln,
    " using free-ion params from LaF3 ..."];
2199      (* calculate intermediate coupling basis using the free-ion
    params for LaF3 *)
2200      {theIntermediateEigensystems, multipletAssignments,
    elevatedIntermediateEigensystems, freeIenergiesAndMultiplets,
    basisChanger} = TheIntermediateEigensystems@@freeBies;
2201
2202      (* use that intermediate coupling basis to compile a function
     for the full Hamiltonian *)
2203      allFreeEnergies = Flatten[First/
    @elevatedIntermediateEigensystems];
2204      (* important that the intermediate coupling basis have
    attached energies, which make possible the truncation *)
2205      ordering = Ordering[allFreeEnergies];
2206      (* sort the free ion energies and determine which indices
    should be included in the truncation *)
2207      allFreeEnergiesSorted          = Sort[allFreeEnergies];
2208      {minFreeEnergy, maxFreeEnergy} = MinMax[allFreeEnergies];
2209      (* determine the index at which the energy is equal or larger
     than the truncation energy *)
2210      sortedTruncationIndex = Which[
2211        truncationEnergy > (maxFreeEnergy-minFreeEnergy),
2212        hamDim,
2213        True,
2214        FirstPosition[allFreeEnergiesSorted - Min[
    allFreeEnergiesSorted],x_/;x>truncationEnergy,{0},1][[1]]
2215      ];
2216      (* the actual energy at which the truncation is made *)
2217      roundedTruncationEnergy = allFreeEnergiesSorted[[
    sortedTruncationIndex]];
2218
2219      (* the indices that participate in the truncation *)
2220      truncationIndices = ordering[[;;sortedTruncationIndex]];
2221      PrintFun["Computing the block structure of the change of
    basis array ..."];
2222      blockSizes         = BlockArrayDimensionsArray[ham];
2223      basisChangerBlocks = ArrayBlocker[basisChanger, blockSizes];
2224      blockShifts        = First /@ Diagonal[blockSizes];
2225      numBlocks          = Length[blockSizes];
2226
2227      (* using the ham (with all the symbols) change the basis to
    the computed one *)
2228      PrintFun["Changing the basis of the Hamiltonian to the
    intermediate coupling basis ..."];
2229      intermediateHam = BlockMatrixMultiply[ham, basisChangerBlocks
```

```
];
     PrintFun["Distributing products inside of symbolic matrix elements to keep complexity in check ..."];
     Do[
       intermediateHam[[rowIdx, colIdx]] = MapToSparseArray[intermediateHam[[rowIdx, colIdx]], Distribute /@ # &];,
       {rowIdx, 1, numBlocks},
       {colIdx, 1, numBlocks}
     ];
     intermediateHam = BlockMatrixMultiply[BlockTranspose[basisChangerBlocks],intermediateHam];
     PrintFun["Distributing products inside of symbolic matrix elements to keep complexity in check ..."];
     Do[
       intermediateHam[[rowIdx, colIdx]] = MapToSparseArray[intermediateHam[[rowIdx, colIdx]], Distribute /@ # &];,
       {rowIdx, 1, numBlocks},
       {colIdx, 1, numBlocks}
     ];
     (* using the truncation indices truncate that one *)
     PrintFun["Truncating the Hamiltonian ..."];
     truncatedIntermediateHam = TruncateBlockArray[intermediateHam, truncationIndices, blockShifts];
     (* these are the basis vectors for the truncated hamiltonian *)
     PrintFun["Saving the truncated intermediate basis ..."];
     truncatedIntermediateBasis = basisChanger[[All, truncationIndices]];

     PrintFun["Compiling a function for the truncated Hamiltonian ..."];
     (* compile a function that will calculate the truncated Hamiltonian given the parameters in allVars, this is the function to be use in fitting *)
     compileIntermediateTruncatedHam = Compile[Evaluate[allVars], Evaluate[truncatedIntermediateHam]];
     (* save the compiled function *)
     PrintFun["Saving the compiled function for the truncated Hamiltonian and the truncated intermediate basis ..."];
     Export[compiledIntermediateFname, {compileIntermediateTruncatedHam, truncatedIntermediateBasis}];
   )
];
)
];

truncationUmbral = Dimensions[truncatedIntermediateBasis][[2]];
PrintFun["The truncated Hamiltonian has a dimension of ", truncationUmbral, "x", truncationUmbral, " ..."];
presentDataIndices = Select[presentDataIndices, # <= truncationUmbral &];
solCompendium["presentDataIndices"] = presentDataIndices;

(* the problemVars are the symbols that will be fitted for *)

PrintFun["Starting up the fitting process using the Levenberg-Marquardt method ..."];
(* using the problemVars I need to create the argument list including _?NumericQ *)
problemVarsQ       = (ToString[#] <> "_?NumericQ") & /@ problemVars;
problemVarsQString = StringJoin[Riffle[problemVarsQ, ", "]];
(* we also need to have the padded arguments with the variables in the right position and the fixed values in the remaining ones *)
problemVarsPositions = Position[allVars, #][[1, 1]] & /@ problemVars;
problemVarsString    = StringJoin[Riffle[ToString /@ problemVars, ", "]];
(* to feed parameters to the Hamiltonian, which includes all parameters, we need to form the set of arguments, with fixed values where needed, and the variables in the right position *)
varsWithConstants                          = standardValues;
varsWithConstants[[problemVarsPositions]] = problemVars;
varsWithConstantsString                    = ToString[varsWithConstants];
```

```
2279
2280    (* this following function serves eigenvalues from the
        Hamiltonian, has memoization so it might grow to use a lot of RAM
        *)
2281    Clear[HamSortedEigenvalues];
2282    hamEigenvaluesTemplate = StringTemplate["
2283    HamSortedEigenvalues[`problemVarsQ`]:=(
2284      ham          = compileIntermediateTruncatedHam@@`
        varsWithConstants`;
2285      eigenValues = Chop[Sort@Eigenvalues@ham];
2286      eigenValues = eigenValues - Min[eigenValues];
2287      HamSortedEigenvalues[`problemVarsString`] = eigenValues;
2288      Return[eigenValues]
2289    )"];
2290    hamString = hamEigenvaluesTemplate[<|
2291        "problemVarsQ"      -> problemVarsQString,
2292        "varsWithConstants" -> varsWithConstantsString,
2293        "problemVarsString" -> problemVarsString
2294        |>];
2295    ToExpression[hamString];
2296
2297    (* we also need a function that will pick the i-th eigenvalue,
        this seems unnecessary but it's needed to form the right
        functional form expected by the Levenberg-Marquardt method *)
2298    eigenvalueDispenserTemplate = StringTemplate["
2299    PartialHamEigenvalues[`problemVarsQ`][i_]:=(
2300      eigenVals = HamSortedEigenvalues[`problemVarsString`];
2301      eigenVals[[i]]
2302    )
2303    "];
2304    eigenValueDispenserString = eigenvalueDispenserTemplate[<|
2305        "problemVarsQ"      -> problemVarsQString,
2306        "problemVarsString" -> problemVarsString
2307        |>];
2308    ToExpression[eigenValueDispenserString];
2309
2310    PrintFun["Determining the free variables after constraints ..."];
2311    constrainedProblemVars     = (problemVars /. constraints);
2312    constrainedProblemVarsList = Variables[constrainedProblemVars];
2313    If[addShift,
2314      PrintFun["Adding a constant shift to the fitting parameters ...
        "];
2315      constrainedProblemVarsList = Append[constrainedProblemVarsList,
        \[Epsilon]]
2316    ];
2317
2318    indepVars = Complement[problemVars, #[[1]] & /@ constraints];
2319    stringPartialVars = ToString/@constrainedProblemVarsList;
2320
2321    paramSols  = {};
2322    rmsHistory = {};
2323    steps      = 0;
2324    problemVarsWithStartValues = KeyValueMap[{#1,#2} &, startValues];
2325    If[addShift,
2326      problemVarsWithStartValues = Append[problemVarsWithStartValues,
        {\[Epsilon] ,0}];
2327    ];
2328    openNotebooks     = If[runningInteractive,
2329              ("WindowTitle"/.NotebookInformation[#]) & /@ Notebooks
        [],
2330              {}];
2331    If[Not[MemberQ[openNotebooks,"Solver Progress"]] && OptionValue["
        ProgressView"],
2332      ProgressNotebook["Basic"->False]
2333    ];
2334    degressOfFreedom = Length[presentDataIndices] - Length[
        problemVars] - 1;
2335    PrintFun["Fitting for ", Length[presentDataIndices], " data
        points with ", Length[problemVars], " free parameters.", " The
        effective degrees of freedom are ", degressOfFreedom, " ..."];
2336
2337    PrintFun["Fitting model to data ..."];
2338    startTime   = Now;
2339    shiftToggle = If[addShift, 1, 0];
2340    sol = FindMinimum[
2341      Sum[(expData[[j]][[1]] - (PartialHamEigenvalues @@
```

```
    constrainedProblemVars)[j] - shiftToggle * \[Epsilon])^2,
      {j, presentDataIndices}],
      problemVarsWithStartValues,
      Method        -> "LevenbergMarquardt",
      MaxIterations -> OptionValue["MaxIterations"],
      AccuracyGoal  -> OptionValue["AccuracyGoal"],
      StepMonitor :> (
        steps      += 1;
        currentSqSum = Sum[(expData[[j]][[1]] - (
    PartialHamEigenvalues @@ constrainedProblemVars)[j] - shiftToggle
    * \[Epsilon])^2, {j, presentDataIndices}];
        currentRMS = Sqrt[currentSqSum / degressOfFreedom];
        paramSols  = AddToList[paramSols, constrainedProblemVarsList,
     maxHistory];
        rmsHistory = AddToList[rmsHistory, currentRMS, maxHistory];
        )
      ];
    endTime = Now;
    timeTaken = QuantityMagnitude[endTime - startTime, "Seconds"];
    PrintFun["Solution found in ", timeTaken, "s"];

    solVec = constrainedProblemVars /. sol[[-1]];
    indepSolVec = indepVars /. sol[[-1]];
    If[addShift,
      \[Epsilon]Best = \[Epsilon]/. sol[[-1]],
      \[Epsilon]Best = 0
    ];
    fullSolVec = standardValues;
    fullSolVec[[problemVarsPositions]] = solVec;
    PrintFun["Calculating the truncated numerical Hamiltonian
    corresponding to the solution ..."];
    fullHam = compileIntermediateTruncatedHam @@ fullSolVec;
    PrintFun["Calculating energies and eigenvectors ..."];
    {eigenEnergies, eigenVectors} = Eigensystem[fullHam];
    states = Transpose[{eigenEnergies, eigenVectors}];
    states = SortBy[states, First];
    eigenEnergies = First /@ states;
    PrintFun["Shifting energies to make ground state zero of energy
    ..."];
    eigenEnergies = eigenEnergies - eigenEnergies[[1]];
    PrintFun["Calculating the linear approximant to each eigenvalue
    ..."];
    allVarsVec = Transpose[{allVars}];
    p0 = Transpose[{fullSolVec}];
    linMat = {};
    If[addShift,
      tail = -2,
      tail = -1];
    Do[
      (
        aVarPosition = Position[allVars, aVar][[1, 1]];
        isolationValues = ConstantArray[0, Length[allVars]];
        isolationValues[[aVarPosition]] = 1;
        dependentVars = KeyValueMap[{#1, D[#2, aVar]} &, Association[
    constraints]];
        Do[
          isolationValues[[Position[allVars, dVar[[1]]][[1, 1]]]] =
    dVar[[2]],
          {dVar, dependentVars}
        ];
        perHam  = compileIntermediateTruncatedHam @@ isolationValues;
        lin     = FirstOrderPerturbation[Last /@ states, perHam];
        linMat  = Append[linMat, lin];
      ),
      {aVar, constrainedProblemVarsList[[;;tail]]}
    ];
    PrintFun["Removing the gradient of the ground state ..."];
    linMat = (# - #[[1]] & /@ linMat);
    PrintFun["Transposing derivative matrices into columns ..."];
    linMat  = Transpose[linMat];

    PrintFun["Calculating the eigenvalue vector at solution ..."];
    \[Lambda]0Vec = Transpose[{eigenEnergies[[presentDataIndices]]}];
    PrintFun["Putting together the experimental vector ..."];
    \[Lambda]exp = Transpose[{First /@ expData[[presentDataIndices
    ]]}];
```

```
    problemVarsVec = If[addShift,
      Transpose[{constrainedProblemVarsList[[;;-2]]}],
      Transpose[{constrainedProblemVarsList}]
    ];
    indepSolVecVec = Transpose[{indepSolVec}];
    PrintFun["Calculating the difference between eigenvalues at solution ..."];
    diff = If[linMat == {},
      (\[Lambda]0Vec - \[Lambda]exp)  + \[Epsilon]Best,
      (\[Lambda]0Vec - \[Lambda]exp)  + \[Epsilon]Best + linMat[[presentDataIndices]].(problemVarsVec - indepSolVecVec)
    ];
    PrintFun["Calculating the sum of squares of differences around solution ... "];
    sqdiff        = Expand[(Transpose[diff] . diff)[[1, 1]]];
    PrintFun["Calculating the minimum (which should coincide with sol) ..."];
    minpoly       = sqdiff /. AssociationThread[problemVars -> solVec];
    fmSolAssoc    = Association[sol[[2]]];
    If[\[Sigma]exp == Automatic,
      \[Sigma]exp = Sqrt[minpoly / degressOfFreedom];
    ];
    \[CapitalDelta]\[Chi]2 = Sqrt[degressOfFreedom];
    Amat = (1/\[Sigma]exp^2) * Transpose[linMat[[presentDataIndices]]].linMat[[presentDataIndices]];
    paramIntervals = EllipsoidBoundingBox[Amat, \[CapitalDelta]\[Chi]2];
    PrintFun["Calculating the uncertainty in the parameters ..."];
    solWithUncertainty = Table[
      (
        aVar        = constrainedProblemVarsList[[varIdx]];
        paramBest   = aVar /. fmSolAssoc;
        (aVar -> {paramBest, paramIntervals[[varIdx, 2]]})
      ),
    {varIdx, 1, Length[constrainedProblemVarsList]-shiftToggle}
    ];

    bestRMS    = Sqrt[minpoly / degressOfFreedom];
    bestParams = sol[[2]];
    bestWithConstraints = Association@Join[constraints, bestParams];
    bestWithConstraints = bestWithConstraints /. bestWithConstraints;
    bestWithConstraints = (# + 0.) & /@ bestWithConstraints;

    solCompendium["degreesOfFreedom"]    = degressOfFreedom;
    solCompendium["solWithUncertainty"]  = solWithUncertainty;
    solCompendium["truncatedDim"]        = truncationUmbral;
    solCompendium["fittedLevels"]        = Length[presentDataIndices];
    solCompendium["actualSteps"]         = steps;
    solCompendium["bestRMS"]             = bestRMS;
    solCompendium["problemVars"]         = problemVars;
    solCompendium["paramSols"]           = paramSols;
    solCompendium["rmsHistory"]          = rmsHistory;
    solCompendium["Appendix"]            = OptionValue["AppendToLogFile"];
    solCompendium["timeTaken/s"]         = timeTaken;
    solCompendium["bestParams"]          = bestParams;
    solCompendium["bestParamsWithConstraints"] = bestWithConstraints;

    If[OptionValue["SaveEigenvectors"],
        solCompendium["states"] = {#[[1]] + \[Epsilon]Best, #[[2]]} &/@ (Chop /@ ShiftedLevels[states]),
        (
          finalEnergies = Sort[First /@ states];
          finalEnergies = finalEnergies - finalEnergies[[1]];
          finalEnergies = finalEnergies + \[Epsilon]Best;
          finalEnergies  = Chop /@ finalEnergies;
          solCompendium["energies"] = finalEnergies;
      )
    ];
    If[OptionValue["SaveToLog"],
      PrintFun["Saving the solution to the log file ..."];
      LogSol[solCompendium, logFilePrefix];
    ];
    PrintFun["Finished ..."];
```

```
    Return[solCompendium];
  )
];

caseConstraints::usage="This Association contains the constraints that are not the same across all of the lanthanides. For instance, since the ratio between M2 and M0 is assumed the same for all the trivalent lanthanides, that one is not included here.
This association has keys equal to symbols of lanthanides and values equal to lists of rules that express either a parameter being held fixed or made proportional to another.
In Table I of Carnall 1989 these correspond to cases were values are given in square brackets.";
caseConstraints = <|
"Ce" -> {
    B02 -> -218.,
    B04 -> 738.,
    B06 -> 679.,
    B22 -> -50.,
    B24 -> 431.,
    B26 -> -921.,
    B44 -> 616.,
    B46 -> -348.,
    B66 -> -788.
    },
"Pr" -> {},
"Nd" -> {},
"Sm" -> {
    B22 -> -50.,
    T2 -> 300.,
    T3 -> 36.,
    T4 -> 56.,
    γ -> 1500.
    },
"Eu" -> {
    F4 -> 0.713 F2,
    F6 -> 0.512 F2,
    B22 -> -50.,
    B24 -> 597.,
    B26 -> -706.,
    B44 -> 408.,
    B46 -> -508.,
    B66 -> -692.,
    M0 -> 2.1,
    P2 -> 360.,
    T2 -> 300.,
    T3 -> 40.,
    T4 -> 60.,
    T6 -> -300.,
    T7 -> 370.,
    T8 -> 320.,
    α -> 20.16,
    β  -> -566.9,
    γ -> 1500.
    },
"Pm" -> {
    B02 -> -245.,
    B04 -> 470.,
    B06 -> 640.,
    B22 -> -50.,
    B24 -> 525.,
    B26 -> -750.,
    B44 -> 490.,
    B46 -> -450.,
    B66 -> -760.,
    F2 -> 76400.,
    F4 -> 54900.,
    F6 -> 37700.,
    M0 -> 2.4,
    P2 -> 275.,
    T2 -> 300.,
    T3 -> 35.,
    T4 -> 58.,
    T6 -> -310.,
    T7 -> 350.,
    T8 -> 320.,
```

```
    α -> 20.5,
    β -> -560.,
    γ -> 1475.,
    ζ -> 1025.},
"Gd" -> {
    F4 -> 0.710 F2,
    B02 -> -231.,
    B04 -> 604.,
    B06 -> 280.,
    B22 -> -99.,
    B24 -> 340.,
    B26 -> -721.,
    B44 -> 452.,
    B46 -> -204.,
    B66 -> -509.,
    T2 -> 300.,
    T3 -> 42.,
    T4 -> 62.,
    T6 -> -295.,
    T7 -> 350.,
    T8 -> 310.,
    β -> -600.,
    γ -> 1575.
    },
"Tb" -> {
    F4 -> 0.707 F2,
    T2 -> 320.,
    T3 -> 40.,
    T4 -> 50.,
    γ -> 1650.
    },
"Dy" -> {},
"Ho" -> {
    B02 -> -240.,
    T2 -> 400.,
    γ -> 1800.
    },
"Er" -> {
    T2 -> 400.,
    γ -> 1800.
    },
"Tm" -> {
    T2 -> 400.,
    γ -> 1820.
    },
"Yb" -> {
    B02 -> -249.,
    B04 -> 457.,
    B06 -> 282.,
    B22 -> -105.,
    B24 -> 320.,
    B26 -> -482.,
    B44 -> 428.,
    B46 -> -234.,
    B66 -> -492.
}
|>;

variedSymbols =<|
    "Ce" -> {ζ},
    "Pr" -> {B02, B04, B06, B22, B24, B26, B44, B46, B66,
             F2, F4, F6,
             M0, P2,
             α, β, γ,
             ζ},
    "Nd" -> {B02, B04, B06, B22, B24, B26, B44, B46, B66,
             F2, F4, F6,
             M0, P2,
             T2, T3, T4, T6, T7, T8,
             α, β, γ,
             ζ},
    "Pm" -> {},
    "Sm" -> {B02, B04, B06, B24, B26, B44, B46, B66,
             F2, F4, F6, M0, P2,
             T6, T7, T8,
             α, β, ζ},
```

```
    "Eu" -> {B02, B04, B06,
            F2, F4, F6, ζ},
    "Gd" -> {F2, F4, F6,
            M0, P2,
            α, ζ},
    "Tb" -> {B02, B04, B06, B22, B24, B26, B44, B46, B66,
            F2, F4, F6,
            M0, P2,
            T6, T7, T8,
            α, β, ζ},
    "Dy" -> {B02, B04, B06, B22, B24, B26, B44, B46, B66,
            F2, F4, F6,
            M0, P2,
            T2, T3, T4, T6, T7, T8,
            α, β, γ, ζ},
    "Ho" -> {B04, B06, B22, B24, B26, B44, B46, B66,
            F2, F4, F6,
            M0, P2,
            T3, T4, T6, T7, T8,
            α, β, ζ},
    "Er" -> {B02, B04, B06, B22, B24, B26, B44, B46, B66,
            F2, F4, F6,
            M0, P2,
            T3, T4, T6, T7, T8, α, β, ζ},
    "Tm" -> {B02, B04, B06, B22, B24, B26, B44, B46, B66,
            F2, F4, F6,
            M0, P2,
            α, β, ζ},
    "Yb" -> {ζ}
    |>;

caseConstraintsM0::usage="This association contains the symbols that
    are held constant in fitting different ions. It only contains
    symbols for which the constraint doesn't apply to all ions.
This association has keys equal to symbols of lanthanides and values
    equal to associations with the symbols for the parameters that are
     held fixed or made proportional to another. If the value is to be
     held constant a placeholder value of 0 is given, if a ratio
    constraint is given, the value is constraint.
The operator basis for which this is applicable is the mostly-
    orthogonal basis.
";
caseConstraintsM0 = <|
"Ce"-> {
    B02 -> 0,
    B04 -> 0,
    B06 -> 0,
    B22 -> 0,
    B24 -> 0,
    B26 -> 0,
    B44 -> 0,
    B46 -> 0,
    B66 -> 0
    },
"Pr"-> {},
"Nd"-> {},
"Sm"-> {
    B22 -> 0,
    T2p -> 0,
    T3 -> 0,
    T4 -> 0,
    γp -> 0
    },
"Eu"-> {
    E2p -> 0.0049 E1p,
    E3p -> 0.098 E1p,
    B22 -> 0,
    B24 -> 0,
    B26 -> 0,
    B44 -> 0,
    B46 -> 0,
    B66 -> 0,
    M0  -> 0,
    P2  -> 0,
    T2p -> 0,
    T3  -> 0,
```

```
    T4  -> 0,
    T6  -> 0,
    T7  -> 0,
    T8  -> 0,
    αp -> 0,
    βp -> 0,
    γp -> 0
    },
"Pm"-> {
    B02 -> 0,
    B04 -> 0,
    B06 -> 0,
    B22 -> 0,
    B24 -> 0,
    B26 -> 0,
    B44 -> 0,
    B46 -> 0,
    B66 -> 0,
    E1p -> 0,
    E2p -> 0,
    E3p -> 0,
    M0 -> 0,
    P2 -> 0,
    T2p -> 0,
    T3 -> 0,
    T4 -> 0,
    T6 -> 0,
    T7 -> 0,
    T8 -> 0,
    αp -> 0,
    βp -> 0,
    γp -> 0,
    ζ -> 0
    },
"Gd"-> {
    E2p -> 0.0049 E1p,
    B02 -> 0,
    B04 -> 0,
    B06 -> 0,
    B22 -> 0,
    B24 -> 0,
    B26 -> 0,
    B44 -> 0,
    B46 -> 0,
    B66 -> 0,
    T2p -> 0,
    T3 -> 0,
    T4 -> 0,
    T6 -> 0,
    T7 -> 0,
    T8 -> 0,
    βp -> 0,
    γp -> 0
    },
"Tb"-> {
    E2p -> 0.0049 E1p,
    T2p -> 0,
    T3 -> 0,
    T4 -> 0,
    γp -> 0
    },
"Dy"-> {},
"Ho"-> {
    B02 -> 0,
    T2p -> 0,
    γp -> 0
    },
"Er"-> {
    T2p -> 0,
    γp -> 0
    },
"Tm"-> {
    γp -> 0
    },
"Yb"-> {
    B02 -> 0,
```

```
    B04 -> 0,
    B06 -> 0,
    B22 -> 0,
    B24 -> 0,
    B26 -> 0,
    B44 -> 0,
    B46 -> 0,
    B66 -> 0
}
|>;

variedSymbolsM0 =<|
    "Ce" -> {ζ},
    "Pr" -> {B02, B04, B06, B22, B24, B26, B44, B46, B66,
             E1p, E2p, E3p,
             M0, P2,
             αp, βp, γp,
             ζ},
    "Nd" -> {B02, B04, B06, B22, B24, B26, B44, B46, B66,
             E1p, E2p, E3p,
             M0, P2,
             T2p, T3, T4, T6, T7, T8,
             αp, βp, γp,
             ζ},
    "Pm" -> {},
    "Sm" -> {B02, B04, B06, B24, B26, B44, B46, B66,
             E1p, E2p, E3p, M0, P2,
             T6, T7, T8,
             αp, βp, ζ},
    "Eu" -> {B02, B04, B06,
             E1p, E2p, E3p, ζ},
    "Gd" -> {E1p, E2p, E3p,
             M0, P2,
             αp, ζ},
    "Tb" -> {B02, B04, B06, B22, B24, B26, B44, B46, B66,
             E1p, E2p, E3p,
             M0, P2,
             T6, T7, T8,
             αp, βp, ζ},
    "Dy" -> {B02, B04, B06, B22, B24, B26, B44, B46, B66,
             E1p, E2p, E3p,
             M0, P2,
             T2p, T3, T4, T6, T7, T8,
             αp, βp, γp, ζ},
    "Ho" -> {B04, B06, B22, B24, B26, B44, B46, B66,
             E1p, E2p, E3p,
             M0, P2,
             T3, T4, T6, T7, T8,
             αp, βp, ζ},
    "Er" -> {B02, B04, B06, B22, B24, B26, B44, B46, B66,
             E1p, E2p, E3p,
             M0, P2,
             T3, T4, T6, T7, T8, αp, βp, ζ},
    "Tm" -> {B02, B04, B06, B22, B24, B26, B44, B46, B66,
             E1p, E2p, E3p,
             M0, P2,
             αp, βp, ζ},
    "Yb" -> {ζ}
    |>;

caseConstraintsLiYF4 = <|
  "Ce" -> {
    B04 -> -1043.,
    B44 -> -1249.,
    B06 -> -65.,
    B46 -> -1069.
    },
  "Pr" -> {
    β -> -644.,
    γ -> 1413.,
    M0 -> 1.88,
    P2 -> 244.
    },
  "Nd" -> {
    M0 -> 1.85
    },
```

```
  "Sm" -> {
    α -> 20.5,
    β -> -616.,
    γ -> 1565.,
    T2 -> 282.,
    T3 -> 26.,
    T4 -> 71.,
    T6 -> -257.,
    T7 -> 314.,
    T8 -> 328.,
    M0 -> 2.38,
    P2 -> 336.
    },
  "Eu" -> {
    T2 -> 370.,
    T3 -> 40.,
    T4 -> 40.,
    T6 -> -300.,
    T7 -> 380.,
    T8 -> 370.
    },
  "Tb" -> {
    F4 -> 0.709 F2,
    F6 -> 0.503 F2,
    α -> 17.6,
    β -> -581.,
    γ -> 1792.,
    T2 -> 330.,
    T3 -> 40.,
    T4 -> 45.,
    T6 -> -365.,
    T7 -> 320.,
    T8 -> 349.,
    M0 -> 2.7,
    P2 -> 482.
    },
  "Dy" -> {
    (* F4 -> 0.707 F2,
    F6 -> 0.516 F2, *)
    F2 -> 90421,
    F4 -> 63928,
    F6 -> 46657,
    α -> 17.9,
    β -> -628.,
    γ -> 1790.,
    T2 -> 326.,
    T3 -> 23.,
    T4 -> 83.,
    T6 -> -294.,
    T7 -> 403.,
    T8 -> 340.,
    M0 -> 4.46,
    P2 -> 610.,
    B46 -> -700.
    },
  "Ho" -> {
    α -> 17.2,
    β -> -596.,
    γ -> 1839.,
    T2 -> 365.,
    T3 -> 37.,
    T4 -> 95.,
    T6 -> -274.,
    T7 -> 331.,
    T8 -> 343.,
    P2 -> 582.
    },
  "Er" -> {},
  "Tm" -> {
    α -> 17.3,
    β -> -665.,
    γ -> 1936.,
    M0 -> 4.93,
    P2 -> 730.,
    T2 -> 400.
    },
```

```
  "Yb" -> {
    B06 -> -23.,
    B46 -> -512.
    }
  |>;

variedSymbolsLiYF4 = <|
  "Ce" -> {
    B02, ζ
    },
  "Pr" -> {
    B02, B04, B06, B44, B46,
    F2, F4, F6,
    α, ζ
    },
  "Nd" -> {
    B02, B04, B06, B44, B46,
    F2, F4, F6,
    P2,
    T2, T3, T4, T6, T7, T8,
    α, β, γ, ζ
    },
  "Sm" -> {
    B02, B04, B06, B44, B46,
    F2, F4, F6,
    ζ
    },
  "Eu" -> {
    B02, B04, B06, B44, B46,
    F2, F4, F6,
    M0, P2,
    α, β, γ, ζ
    },
  "Tb" -> {
    B02, B04, B06, B44, B46,
    F2, F4, F6,
    ζ
    },
  "Dy" -> {
    B02, B04, B06, B44,
    F2, F4, F6,
    ζ
    },
  "Ho" -> {
    B02, B04, B06, B44, B46,
    F2, F4, F6,
    M0,
    ζ
    },
  "Er" -> {
    B02, B04, B06, B44, B46,
    F2, F4, F6,
    M0, P2,
    T2, T3, T4, T6, T7,  T8,
    α, β, γ,
    ζ
    },
  "Tm" -> {
    B02, B04, B06, B44, B46,
    F2, F4, F6,
    ζ
    },
  "Yb" -> {
    B02, B04, B44,
    ζ
    }
  |>

paramsChengLiYF4::usage="This association has the model parameters as
    fitted by Cheng et. al \"Crystal-field analyses for trivalent
   lanthanide ions in LiYF4\".";
paramsChengLiYF4 = <|
  "Ce" -> {
    ζ -> 630.,
    B02 -> 354., B04 -> -1043.,
    B44 -> -1249., B06 -> -65.,
```

```
    B46 -> -1069.
    },
 "Pr" -> {
    F2 -> 68955., F4 -> 50505., F6 -> 33098.,
    ζ -> 748.,
    α -> 23.3, β -> -644., γ -> 1413.,
    M0 -> 1.88, P2 -> 244.,
    B02 -> 512.,   B04 -> -1127.,
    B44 -> -1239., B06 -> -85.,
    B46 -> -1205.
    },
"Nd" -> {
    F2 -> 72952., F4 -> 52681., F6 -> 35476.,
    ζ -> 877.,
    α -> 21., β -> -579., γ -> 1446.,
    T2 -> 210., T3 -> 41., T4 -> 74., T6 -> -293., T7 -> 321., T8 ->
   205.,
    M0 -> 1.85, P2 -> 304.,
    B02 -> 391.,   B04 -> -1031.,
    B44 -> -1271., B06 -> -28.,
    B46 -> -1046.
    },
"Sm" -> {
    F2 -> 79515., F4 -> 56766., F6 -> 40078.,
    ζ -> 1168.,
    α -> 20.5, β -> -616., γ -> 1565.,
    T2 -> 282., T3 -> 26., T4 -> 71., T6 -> -257., T7 -> 314., T8 ->
   328.,
    M0 -> 2.38, P2 -> 336.,
    B02 -> 370.,  B04 -> -757.,
    B44 -> -941., B06 -> -67.,
    B46 -> -895.
    },
"Eu" -> {
    F2 -> 82573., F4 -> 59646., F6 -> 43203.,
    ζ -> 1329.,
    α -> 21.6, β -> -482., γ -> 1140.,
    T2 -> 370., T3 -> 40., T4 -> 40., T6 -> -300., T7 -> 380., T8 ->
   370.,
    M0 -> 2.41, P2 -> 332.,
    B02 -> 339.,   B04 -> -733.,
    B44 -> -1067., B06 -> -36.,
    B46 -> -764.
    },
"Tb" -> {
    F2 -> 90972., F4 -> 64499., F6 -> 45759.,
    ζ -> 1702.,
    α -> 17.6, β -> -581., γ -> 1792.,
    T2 -> 330., T3 -> 40., T4 -> 45., T6 -> -365., T7 -> 320., T8 ->
   349.,
    M0 -> 2.7, P2 -> 482.,
    B02 -> 413.,   B04 -> -867.,
    B44 -> -1114., B06 -> -41.,
    B46 -> -736.
    },
"Dy" -> {
    F0 -> 0,
    F2 -> 90421., F4 -> 63928., F6 -> 46657.,
    ζ -> 1895.,
    α -> 17.9, β -> -628., γ -> 1790.,
    T2 -> 326., T3 -> 23., T4 -> 83., T6 -> -294., T7 -> 403., T8 ->
   340.,
    M0 -> 4.46, P2 -> 610.,
    B02 -> 360.,  B04 -> -737.,
    B44 -> -943., B06 -> -35.,
    B46 -> -700.
    },
"Ho" -> {
    F2 -> 93512., F4 -> 66084., F6 -> 49765.,
    ζ -> 2126.,
    α -> 17.2, β -> -596., γ -> 1839.,
    T2 -> 365., T3 -> 37., T4 -> 95., T6 -> -274., T7 -> 331., T8 ->
   343.,
    M0 -> 3.92, P2 -> 582.,
    B02 -> 386.,  B04 -> -629.,
    B44 -> -841., B06 -> -33.,
```

```
    B46 -> -687.
    },
 "Er" -> {
    F2 -> 97326., F4 -> 67987., F6 -> 53651.,
    ζ -> 2377.,
    α -> 18.1, β -> -599., γ -> 1870.,
    T2 -> 380., T3 -> 41., T4 -> 69., T6 -> -356., T7 -> 239., T8 ->
    390.,
    M0 -> 4.41, P2 -> 795.,
    B02 -> 325.,   B04 -> -749.,
    B44 -> -1014., B06 -> -19.,
    B46 -> -635.
    },
 "Tm" -> {
    F0 -> 0.,
    T2 -> 0.,
    F2 -> 101938., F4 -> 71553., F6 -> 51359.,
    ζ -> 2632.,
    α -> 17.3, β -> -665., γ -> 1936.,
    M0 -> 4.93, P2 -> 730.,
    B02 -> 339.,  B04 -> -627.,
    B44 -> -913., B06 -> -39.,
    B46 -> -584.
    },
 "Yb" -> {
    ζ -> 2916.,
    B02 -> 446.,  B04 -> -560.,
    B44 -> -843., B06 -> -23.,
    B46 -> -512.
    }
  |>

StringToSLJ[string_] := Module[
  {stringed = string, LS, J, LSindex},
  (
  If[StringContainsQ[stringed, "+"],
   Return["mixed"]
   ];
  LS = StringTake[stringed, {1, 2}];
  If[StringContainsQ[stringed, "("],
   (
     LSindex =
     StringCases[stringed, RegularExpression["\\(([^)]*)\\)"] :> "$1"
   ];
    LS = LS <> LSindex;
    stringed = StringSplit[stringed, ")"][[-1]];
    J = ToExpression[stringed];
    ),
   (
    J = ToExpression@StringTake[stringed, {3, -1}];
    )
   ];
  {LS, J}
  )
];

FreeIonSolver::usage="This function takes a list of experimental data
    and the number of electrons in the lanthanide ion and returns the
    free-ion parameters that best fit the data. The options are:
  - F4F6_SlaterRatios: a list of two numbers that represent the ratio
    of F4 to F2 and F6 to F2, respectively.
  - PrintFun: a function that will be used to print the progress of
    the fitting process.
  - MaxIterations: the maximum number of iterations that the fitting
    process will run.
  - MaxMultiplets: the maximum number of multiplets that will be used
    in the fitting process.
  - MaxPercent: the maximum percentage of the data that can be off by
    the fitting.
  - SubSetBounds: a list of two numbers that represent the minimum
    and maximum number of multiplets that will be used in the fitting
    process.
The function returns an association with the following keys:
  - bestParams: the best parameters found in the fitting.
  - worstRelativeError: the worst relative error in the fitting.
  - SlaterRatios: the Slater ratios used in the fitting.
```

```
  - usedBaricenters: the baricenters used in the fitting.
If no acceptable solution is found, the function will return all solutions that are not worse than 10*MaxPercent. A solution is acceptable if the worst relative error is less than the MaxPercent option.
";
Options[FreeIonSolver] = {
  "F4F6_SlaterRatios" -> {0.707, 0.516},
  "PrintFun" -> PrintTemporary,
  "MaxIterations" -> 10000,
  "MaxMultiplets" -> 12,
  "MaxPercent" -> 3.,
  "SubSetBounds" -> {5, 12}
  };
FreeIonSolver[expData_, numE_, OptionsPattern[]] := Module[
  (* {maxMultiplets, maxPercent, F4overF2, F6overF2, PrintFun,
   minSubSetSize, maxSubSetSize, multipletEnergies, numMultiplets,
   allEqns, subsetSizes, ln, solutions, subsets, subset, eqns, m, b,
   meritFun, sol, goodThings, bestThings, bestOfAll, finalSol,
   usedMultiplets, usedBaricenters}, *)
  {},
  (
    maxMultiplets = OptionValue["MaxMultiplets"];
    maxIterations = OptionValue["MaxIterations"];
    maxPercent    = OptionValue["MaxPercent"];
    F4overF2      = OptionValue["F4F6_SlaterRatios"][[1]];
    F6overF2      = OptionValue["F4F6_SlaterRatios"][[2]];
    PrintFun      = OptionValue["PrintFun"];
    minSubSetSize = OptionValue["SubSetBounds"][[1]];
    maxSubSetSize = OptionValue["SubSetBounds"][[2]];
    freeIonParams = {F0, F2, F4, F6, ζ};
    ln            = theLanthanides[[numE]];

    PrintFun["Parsing the barycenters of the different multiplets ..."];
    multipletEnergies = Map[First, #] & /@ GroupBy[expData, #[[2]] &];
    multipletEnergies = Mean[Select[#, NumberQ]] & /@ multipletEnergies;
    multipletEnergies = Select[multipletEnergies, FreeQ[#, Mean] &];
    multipletEnergies = KeySelect[KeyMap[StringToSLJ, multipletEnergies], # =!= "mixed" &];
    multipletEnergies = KeyMap[Prepend[#, numE] &, multipletEnergies];
    numMultiplets     = Length[multipletEnergies];

    PrintFun["Composing the system of equations for the free-ion energies ..."];
    allEqns     = KeyValueMap[FreeIonTable[#1] == #2 &, multipletEnergies];
    allEqns     = Append[Coefficient[#[[1]], {F0, F2, F4, F6, ζ}], #[[2]]] & /@ allEqns;
    allEqns     = allEqns[[;; Min[Length[allEqns], maxMultiplets]]];
    subsetSizes = Range[1, numMultiplets];
    numSubsets = {#, Binomial[numMultiplets, #]} & /@ subsetSizes;
    numSubsets = Transpose@SortBy[numSubsets, Last];
    accSizes = Accumulate[numSubsets[[2]]];
    numSubsets = Transpose@Append[numSubsets, accSizes];
    lastSub = SelectFirst[numSubsets, #[[3]] > 1000 &, Last[numSubsets]];
    lastPosition = Position[numSubsets, lastSub][[1, 1]];
    chosenSubsetSizes = #[[1]] & /@ numSubsets[[;; lastPosition]];
    solutions   = <||>;

    PrintFun["Selecting subsets of different lengths and fitting with ratio-constraints ..."];
    Do[
      subsets = Subsets[Range[1, Length[allEqns]], {subsetSize}];
      PrintFun["Considering ", Length[subsets], " barycenter subsets of size ", subsetSize, " ..."];
      Do[
      (
        subset = subsets[[subsetIndex]];
        eqns   = allEqns[[subset]];
        m      = #[[;; 5]] & /@ eqns;
        b      = #[[6]] & /@ eqns;
```

```
        meritFun = Max[100 * Expand[Abs[(m . freeIonParams - b)]/b]];
        sol = NMinimize[{meritFun,
          F0 > 0,
          F2 > 0,
          F4 == F4overF2 * F2,
          F6 == F6overF2 * F2,
          ζ > 0},
          freeIonParams,
          MaxIterations -> maxIterations,
          Method -> "Convex"];
        solutions[{subsetSize, subset}] = sol;
        )
      , {subsetIndex, 1, Length[subsets]}
      ],
      {subsetSize, chosenSubsetSizes}
    ];

    PrintFun["Collecting solutions of different subset size ..."];
    goodThings = Table[Normal[Sort[KeySelect[#[[1]] & /@
            solutions, #[[1]] == subSize &]]][[1]],
            {subSize, subsetSizes}];

    PrintFun["Picking the solutions that are not worse than ",
    maxPercent, "% ..."];
    bestThings = Select[goodThings, #[[2]] <= maxPercent &];
    If[bestThings == {},
      Print["No acceptable solution found, consider increasing
    maxPercent or inspecting the given data ..."];
      Return[goodThings];
    ];

    PrintFun["Keeping the solution with the largest number of used
    barycenters ..."];
    bestOfAll = bestThings[[-1]];
    sol       = solutions[bestOfAll[[1]]];
    subset    = bestOfAll[[1, 2]];
    eqns      = allEqns[[subset]];
    m         = #[[;; 5]] & /@ eqns;
    b         = #[[6]] & /@ eqns;
    usedMultiplets  = Keys[multipletEnergies][[subset]];
    usedBaricenters = {#, multipletEnergies[#]} & /@ usedMultiplets;
    uniqeLS = DeleteDuplicates[#[[2]] &/@ Keys[multipletEnergies]];
    solAssoc = Association[sol[[2]]];
    usedLaF3 = False;
    If[Length[uniqeLS] == 1,
      (
        Print["There is too little data to find Slater parameters,
    using the ones for LaF3, and keeping the fitted spin-orbit zeta
    ..."];
        laf3params = LoadLaF3Parameters[ln];
        usedLaF3   = True;
        solAssoc[F0] = laf3params[F0];
        solAssoc[F2] = laf3params[F2];
        solAssoc[F4] = laf3params[F4];
        solAssoc[F6] = laf3params[F6];
      )
    ];
    finalSol = <|
      "bestParams" -> solAssoc,
      "usedLaF3" -> usedLaF3,
      "worstRelativeError" -> sol[[1]],
      "SlaterRatios" -> {F4overF2, F6overF2},
      "usedBaricenters" -> usedBaricenters|>;
    Return[finalSol];
)
];
```

17.3 qplotter.m

This module has a few useful plotting routines.

```
BeginPackage["qplotter`"];

GetColor;
IndexMappingPlot;
```

```
ListLabelPlot;
AutoGraphicsGrid;
SpectrumPlot;
WaveToRGB;

Begin["`Private`"];

  AutoGraphicsGrid::usage="AutoGraphicsGrid[graphsList] takes a list
    of graphics and creates a GraphicsGrid with them. The number of
    columns and rows is chosen automatically so that the grid has a
    squarish shape.";
  Options[AutoGraphicsGrid] = Options[GraphicsGrid];
  AutoGraphicsGrid[graphsList_, opts : OptionsPattern[]] :=
    (
      numGraphs = Length[graphsList];
      width = Floor[Sqrt[numGraphs]];
      height = Ceiling[numGraphs/width];
      groupedGraphs = Partition[graphsList, width, width, 1, Null];
      GraphicsGrid[groupedGraphs, opts]
    )

  Options[IndexMappingPlot] = Options[Graphics];
  IndexMappingPlot::usage =
    "IndexMappingPlot[pairs] take a list of pairs of integers and
    creates a visual representation of how they are paired. The first
    indices being depicted in the bottom and the second indices being
    depicted on top.";
  IndexMappingPlot[pairs_, opts : OptionsPattern[]] := Module[{width,
     height}, (
    width = Max[First /@ pairs];
    height = width/3;
    Return[
      Graphics[{{Tooltip[Point[{#[[1]], 0}],#[[1]]]}, Tooltip[Point
    [{#[[2]], height}],#[[2]]],
          Line[{{#[[1]], 0}, {#[[2]], height}}]} & /@ pairs, opts,
    ImageSize -> 800]]
    )
    ]

  TickCompressor[fTicks_] :=
  Module[{avgTicks, prevTickLabel, groupCounter, groupTally, idx,
    tickPosition, tickLabel, avgPosition, groupLabel}, (avgTicks =
    {};
    prevTickLabel = fTicks[[1, 2]];
    groupCounter = 0;
    groupTally = 0;
    idx = 1;
    Do[({tickPosition, tickLabel} = tick;
      If[
      tickLabel === prevTickLabel,
      (groupCounter += 1;
        groupTally += tickPosition;
        groupLabel = tickLabel;),
      (
        avgPosition = groupTally/groupCounter;
        avgTicks = Append[avgTicks, {avgPosition, groupLabel}];
        groupCounter = 1;
        groupTally = tickPosition;
        groupLabel = tickLabel;
        )
      ];
      If[idx != Length[fTicks],
      prevTickLabel = tickLabel;
      idx += 1;]
      ), {tick, fTicks}];
    If[Or[Not[prevTickLabel === tickLabel], groupCounter > 1],
    (
      avgPosition = groupTally/groupCounter;
      avgTicks = Append[avgTicks, {avgPosition, groupLabel}];
      )
    ];
    Return[avgTicks];)]

  GetColor[s_Style] := s /. Style[_, c_] :> c
  GetColor[_] := Black

```

```
ListLabelPlot::usage="ListLabelPlot[data, labels] takes a list of numbers with corresponding labels. The data is grouped according to the labels and a ListPlot is created with them so that each group has a different color and their corresponding label is shown in the horizontal axis.";
Options[ListLabelPlot] = Join[Options[ListPlot], {"TickCompression"->True,
"LabelLevels"->1}];
ListLabelPlot[data_, labels_, opts : OptionsPattern[]] := Module[
  {uniqueLabels, pallete, groupedByTerm, groupedKeys, scatterGroups,
   groupedColors, frameTicks, compTicks, bottomTicks, topTicks},
   (
   uniqueLabels  = DeleteDuplicates[labels];
   pallete = Table[ColorData["Rainbow", i], {i, 0, 1,
       1/(Length[uniqueLabels] - 1)}];
   uniqueLabels  = (#[[1]] -> #[[2]]) & /@ Transpose[{RandomSample[uniqueLabels], pallete}];
   uniqueLabels  = Association[uniqueLabels];
   groupedByTerm = GroupBy[Transpose[{labels, Range[Length[data]], data}], First];
   groupedKeys   = Keys[groupedByTerm];
   scatterGroups = Transpose[Transpose[#][[2 ;; 3]]] & /@ Values[groupedByTerm];
   groupedColors = uniqueLabels[#] & /@ groupedKeys;
   frameTicks    = {Transpose[{Range[Length[data]],
     Style[Rotate[#, 90 Degree], uniqueLabels[#]] & /@ labels}],
     Automatic};
     If[OptionValue["TickCompression"], (
         compTicks = TickCompressor[frameTicks[[1]]];
         bottomTicks =
             MapIndexed[
             If[EvenQ[First[#2]], {#1[[1]],
                 Tooltip[Style["\[SmallCircle]", GetColor[#1[[2]]]],#1[[2]]]
                 }, #1] &, compTicks];
         topTicks =
             MapIndexed[
             If[OddQ[First[#2]], {#1[[1]],
                 Tooltip[Style["\[SmallCircle]", GetColor[#1[[2]]]],#1[[2]]]
                 }, #1] &, compTicks];
         frameTicks = {{Automatic, Automatic}, {bottomTicks, topTicks}};)
   ];
   ListPlot[scatterGroups,
     opts,
     Frame      -> True,
     AxesStyle  -> {Directive[Black, Dotted], Automatic},
     PlotStyle  -> groupedColors,
     FrameTicks -> frameTicks]
   )
   ]

WaveToRGB::usage="WaveToRGB[wave, gamma] takes a wavelength in nm and returns the corresponding RGB color. The gamma parameter is optional and defaults to 0.8. The wavelength wave is assumed to be in nm. If the wavelength is below 380 the color will be the same as for 380 nm. If the wavelength is above 750 the color will be the same as for 750 nm. The function returns an RGBColor object. REF: https://www.noah.org/wiki/wave_to_rgb_in_Python. ";
WaveToRGB[wave_, gamma_ : 0.8] := (
  wavelength = (wave);
  Which[
    wavelength < 380,
      wavelength = 380,
    wavelength > 750,
      wavelength = 750
  ];
  Which[380 <= wavelength <= 440,
  (
    attenuation = 0.3 + 0.7*(wavelength - 380)/(440 - 380);
    R = ((-(wavelength - 440)/(440 - 380))*attenuation)^gamma;
    G = 0.0;
    B = (1.0*attenuation)^gamma;
    ),
```

```
    440 <= wavelength <= 490,
    (
      R = 0.0;
      G = ((wavelength - 440)/(490 - 440))^gamma;
      B = 1.0;
      ),
    490 <= wavelength <= 510,
    (
      R = 0.0;
      G = 1.0;
      B = (-(wavelength - 510)/(510 - 490))^gamma;
      ),
    510 <= wavelength <= 580,
    (
      R = ((wavelength - 510)/(580 - 510))^gamma;
      G = 1.0;
      B = 0.0;
      ),
    580 <= wavelength <= 645,
    (
      R = 1.0;
      G = (-(wavelength - 645)/(645 - 580))^gamma;
      B = 0.0;
      ),
    645 <= wavelength <= 750,
    (
      attenuation = 0.3 + 0.7*(750 - wavelength)/(750 - 645);
      R = (1.0*attenuation)^gamma;
      G = 0.0;
      B = 0.0;
      ),
    True,
    (
      R = 0;
      G = 0;
      B = 0;
      )];
    Return[RGBColor[R, G, B]]
    )

  FuzzyRectangle::usage = "FuzzyRectangle[xCenter, width, ymin,
    height, color] creates a polygon with a fuzzy edge. The polygon is
     centered at xCenter and has a full horizontal width of width. The
     bottom of the polygon is at ymin and the height is height. The
    color of the polygon is color. The left edge and the right edge of
     the resulting polygon will be transparent and the middle will be
    colored. The polygon is returned as a list of polygons.";
  FuzzyRectangle[xCenter_, width_, ymin_, height_, color_, intensity_
    :1] := Module[
    {intenseColor, nocolor, ymax, polys},
  (
    nocolor = Directive[Opacity[0], color];
    ymax = ymin + height;
    intenseColor = Directive[Opacity[intensity], color];
    polys = {
      Polygon[{
        {xCenter - width/2, ymin},
        {xCenter, ymin},
        {xCenter, ymax},
        {xCenter - width/2, ymax}},
        VertexColors -> {
          nocolor,
          intenseColor,
          intenseColor,
          nocolor,
          nocolor}],
      Polygon[{
        {xCenter, ymin},
        {xCenter + width/2, ymin},
        {xCenter + width/2, ymax},
        {xCenter, ymax}},
        VertexColors -> {
          intenseColor,
          nocolor,
          nocolor,
          intenseColor,
```

```
          intenseColor}]
      };
    Return[polys]
    );
]

Options[SpectrumPlot] = Options[Graphics];
Options[SpectrumPlot] = Join[Options[SpectrumPlot], {"Intensities"
  -> {},"Tooltips" -> True, "Comments" -> {}, "SpectrumFunction" ->
  WaveToRGB}];
SpectrumPlot::usage="SpectrumPlot[lines, widthToHeightAspect,
  lineWidth] takes a list of spectral lines and creates a visual
  representation of them. The lines are represented as fuzzy
  rectangles with a width of lineWidth and a height that is
  determined by the overall condition that the width to height ratio
   of the resulting graph is widthToHeightAspect. The color of the
  lines is determined by the wavelength of the line. The function
  assumes that the lines are given in nm.
If the lineWidth parameter is a single number, then every line
  shares that width. If the lineWidth parameter is a list of numbers
  , then each line has a different width. The function returns a
  Graphics object. The function also accepts any options that
  Graphics accepts. The background of the plot is black by default.
  The plot range is set to the minimum and maximum wavelength of the
   given lines.
Besides the options for Graphics the function also admits the
  option Intensities. This option is a list of numbers that
  determines the intensity of each line. If the Intensities option
  is not given, then the lines are drawn with full intensity. If the
   Intensities option is given, then the lines are drawn with the
  given intensity. The intensity is a number between 0 and 1.
The function also admits the option \"Tooltips\". If this option is
   set to True, then the lines will have a tooltip that shows the
  wavelength of the line. If this option is set to False, then the
  lines will not have a tooltip. The default value for this option
  is True.
If \"Tooltips\" is set to True and the option \"Comments\" is a non
  -empty list, then the tooltip will append the wavelength and the
  values in the comments list for the tooltips.
The function also admits the option \"SpectrumFunction\". This
  option is a function that takes a wavelength and returns a color.
  The default value for this option is WaveToRGB.
";
SpectrumPlot[lines_, widthToHeightAspect_, lineWidth_, opts :
  OptionsPattern[]] := Module[
  {minWave, maxWave, height, fuzzyLines},
(
  colorFun = OptionValue["SpectrumFunction"];
  {minWave, maxWave} = MinMax[lines];
  height     = (maxWave - minWave)/widthToHeightAspect;
  fuzzyLines = Which[
    NumberQ[lineWidth] && Length[OptionValue["Intensities"]] == 0,
      FuzzyRectangle[#, lineWidth, 0, height, colorFun[#]] &/@
  lines,
    Not[NumberQ[lineWidth]] && Length[OptionValue["Intensities"]]
  == 0,
      MapThread[FuzzyRectangle[#1, #2, 0, height, colorFun[#1]] &,
  {lines, lineWidth}],
    NumberQ[lineWidth] && Length[OptionValue["Intensities"]] > 0,
      MapThread[FuzzyRectangle[#1, lineWidth, 0, height, colorFun
  [#1], #2] &, {lines, OptionValue["Intensities"]}],
    Not[NumberQ[lineWidth]] && Length[OptionValue["Intensities"]] >
   0,
      MapThread[FuzzyRectangle[#1, #2, 0, height, colorFun[#1], #3]
   &, {lines, lineWidth, OptionValue["Intensities"]}]
  ];
  comments = Which[
    Length[OptionValue["Comments"]] > 0,
      MapThread[StringJoin[ToString[#1]<>" nm","\n",ToString[#2]]&,
      {lines, OptionValue["Comments"]}],
    Length[OptionValue["Comments"]] == 0,
      ToString[#] <>" nm" & /@ lines,
    True,
    {}
  ];
  If[OptionValue["Tooltips"],
```

```
      fuzzyLines = MapThread[Tooltip[#1, #2] &, {fuzzyLines, comments
    }];
    ];
    graphicsOpts = FilterRules[{opts}, Options[Graphics]];
    Graphics[fuzzyLines,
        graphicsOpts,
        Background -> Black,
        PlotRange -> {{minWave, maxWave}, {0, height}}]
    )
  ];

End[];

EndPackage[];
```

17.4 misc.m

This module includes a few functions useful for data-handling.

```
BeginPackage["misc`"];
(* Needs["MaTeX`"]; *)

ArrayBlocker;
BlockAndIndex;
BlockArrayDimensionsArray;
BlockMatrixMultiply;
BlockTranspose;

EllipsoidBoundingBox;
EllipsoidBoundingBox2;
ExportToH5;
ExtractSymbolNames;
FirstOrderPerturbation;
FlattenBasis;

FlowMatching;
GetModificationDate;
GreedyMatching;
HamTeX;
HelperNotebook;

RecoverBasis;
RemoveTrailingDigits;
ReplaceDiagonal;
RobustMissingQ;
RobustMissingQ;

RoundToSignificantFigures;
RoundValueWithUncertainty;
SecondOrderPerturbation;
StochasticMatching;
SuperIdentity;

TextBasedProgressBar;
ToPythonSparseFunction;
ToPythonSymPyExpression;
TruncateBlockArray;

Begin["`Private`"];

  RemoveTrailingDigits[s_String] := StringReplace[s,
    RegularExpression["\\d+$"] -> ""];

  BlockTranspose[anArray_]:=(
    Map[Transpose, Transpose[anArray], {2}]
  );

  BlockMatrixMultiply::usage="BlockMatrixMultiply[A,B] gives the
    matrix multiplication of A and B, with A and B having a compatible
     block structure that allows for matrix multiplication into a
    congruent block structure.";
  BlockMatrixMultiply[Amat_,Bmat_]:=Module[{rowIdx,colIdx,sumIdx},
  (
    Table[
      Sum[Amat[[rowIdx,sumIdx]].Bmat[[sumIdx,colIdx]],{sumIdx,1,
    Dimensions[Amat][[2]]}],
```

```
    {rowIdx,1,Dimensions[Amat][[1]]},
    {colIdx,1,Dimensions[Bmat][[2]]}
    ]
  )
  ];

  BlockAndIndex::usage="BlockAndIndex[blockSizes, index] takes a list
     of bin widths and index. The function return in which block the
    index would be, were the bins to be layed out from left to right.
    The function also returns the position within the bin in which it
    is accomodated. The function returns these two numbers as a list
    of two elements {blockIndex, blockSubIndex}";
  BlockAndIndex[blockSizes_List, index_Integer]:=Module[{
    accumulatedBlockSize,blockIndex, blockSubIndex},
  (
    accumulatedBlockSize = Accumulate[blockSizes];
    If[accumulatedBlockSize[[-1]]-index<0,
      Print["Index out of bounds"];
      Abort[]
    ];
    blockIndex    = Flatten[Position[accumulatedBlockSize-index,n_ /;
     n>=0]][[1]];
    blockSubIndex = Mod[index-accumulatedBlockSize[[blockIndex]],
    blockSizes[[blockIndex]],1];
    Return[{blockIndex,blockSubIndex}]
  )
  ];

  TruncateBlockArray::usage="TruncateBlockArray[blockArray,
    truncationIndices, blockWidths] takes a an array of blocks and
    selects the columns and rows corresponding to truncationIndices.
    The indices being given in what would be the ArrayFlatten[
    blockArray] version of the array. They blocks in the given array
    may be SparseArray. This is equivalent to FlattenArray[blockArray
    ][truncationIndices, truncationIndices] but may be more efficient
    blockArray is sparse.";
  TruncateBlockArray[blockArray_,truncationIndices_,blockWidths_]:=
    Module[
  {truncatedArray,blockCol,blockRow,blockSubCol,blockSubRow},(
  truncatedArray = Table[
    {blockCol,blockSubCol} = BlockAndIndex[blockWidths,fullColIndex];
    {blockRow,blockSubRow} = BlockAndIndex[blockWidths,fullRowIndex];
    blockArray[[blockRow,blockCol]][[blockSubRow,blockSubCol]],
  {fullColIndex,truncationIndices},
  {fullRowIndex,truncationIndices}
  ];
  Return[truncatedArray]
  )
  ];

  BlockArrayDimensionsArray::usage="BlockArrayDimensionsArray[
    blockArray] returns the array of block sizes in a given blocked
    array.";
  BlockArrayDimensionsArray[blockArray_]:=(
    Map[Dimensions,blockArray,{2}]
  );

  ArrayBlocker::usage="ArrayBlocker[anArray, blockSizes] takes a flat
     2d array and a congruent 2D array of block sizes, and with them
    it returns the original array with the block structure imposed by
    blockSizes. The resulting array satisfies ArrayFlatten[
    blockedArray] == anArray, and also Map[Dimensions, blockedArray
    ,{2}] == blockSizes.";
  ArrayBlocker[anArray_,blockSizes_]:=Module[{rowStart,colStart,
    colEnd,numBlocks,blockedArray,blockSize,rowEnd,aBlock,idxRow,
    idxCol},(
    rowStart   = 1;
    colStart   = 1;
    colEnd     = 1;
    numBlocks  = Length[blockSizes];
    blockedArray = Table[(
      blockSize  = blockSizes[[idxRow,idxCol]];
      rowEnd     = rowStart+blockSize[[1]]-1;
      colEnd     = colStart+blockSize[[2]]-1;
      aBlock     = anArray[[rowStart;;rowEnd,colStart;;colEnd]];
      colStart   = colEnd+1;
```

```
      If[idxCol==numBlocks,
        rowStart=rowEnd+1;
        colStart=1;
      ];
      aBlock
    ),
    {idxRow,1,numBlocks},
    {idxCol,1,numBlocks}
    ];
    Return[blockedArray]
  )
  ];

  ReplaceDiagonal::usage =
    "ReplaceDiagonal[matrix, repValue] replaces all the diagonal of
    the given array to the given value. The array is assumed to be
    square and the replacement value is assumed to be a number. The
    returned value is the array with the diagonal replaced. This
    function is useful for setting the diagonal of an array to a given
     value. The original array is not modified. The given array may be
     sparse.";
  ReplaceDiagonal[matrix_, repValue_] :=
    ReplacePart[matrix,
      Table[{i, i} -> repValue, {i, 1, Length[matrix]}]];

  Options[RoundValueWithUncertainty] = {"SetPrecision" -> False};
  RoundValueWithUncertainty::usage = "RoundValueWithUncertainty[x,dx]
     given a number x together with an uncertainty dx this function
    rounds x to the first significant figure of dx and also rounds dx
    to have a single significant figure.
  The returned value is a list with the form {roundedX, roundedDx}.
  The option \"SetPrecision\" can be used to control whether the
    Mathematica precision of x and dx is also set accordingly to these
     rules, otherwise the rounded numbers still have the original
    precision of the input values.
  If the position of the first significant figure of x is after the
    position of the first significant figure of dx, the function
    returns {0,dx} with dx rounded to one significant figure.";
  RoundValueWithUncertainty[x_, dx_, OptionsPattern[]] := Module[
    {xExpo, dxExpo, sigFigs, roundedX, roundedDx, returning},
    (
      xExpo   = RealDigits[x][[2]];
      dxExpo  = RealDigits[dx][[2]];
      sigFigs = (xExpo - dxExpo) + 1;
      {roundedX, roundedDx} = If[sigFigs <= 0,
        {0., N@RoundToSignificantFigures[dx, 1]},
        N[
        {
          RoundToSignificantFigures[x, xExpo - dxExpo + 1],
          RoundToSignificantFigures[dx, 1]}
        ]
        ];
      returning = If[
        OptionValue["SetPrecision"],
        {SetPrecision[roundedX, Max[1, sigFigs]],
        SetPrecision[roundedDx, 1]},
        {roundedX, roundedDx}
        ];
      Return[returning]
      )
    ];

  RoundToSignificantFigures::usage =
    "RoundToSignificantFigures[x, sigFigs] rounds x so that it only
    has \
  sigFigs significant figures.";
  RoundToSignificantFigures[x_, sigFigs_] :=
    Sign[x]*N[FromDigits[RealDigits[x, 10, sigFigs]]];

  RobustMissingQ[expr_] := (FreeQ[expr, _Missing] === False);

  TextBasedProgressBar[progress_, totalIterations_, prefix_:""] :=
    Module[
      {progMessage},
      progMessage = ToString[progress] <> "/" <> ToString[
    totalIterations];
```

```
      If[progress < totalIterations,
          WriteString["stdout", StringJoin[prefix, progMessage, "\r"]],
          WriteString["stdout", StringJoin[prefix, progMessage, "\n"]]
      ];
];

FirstOrderPerturbation::usage="Given the eigenVectors of a matrix A (which doesn't need to be given) together with a corresponding perturbation matrix perMatrix, this function calculates the first derivative of the eigenvalues with respect to the scale factor of the perturbation matrix. In the sense that the eigenvalues of the matrix A + β perMatrix are to first order equal to \[Lambda] + \[Delta]_i β, where the \[Delta]_i are the returned values. This assuming that the eigenvalues are non-degenerate.";
FirstOrderPerturbation[eigenVectors_, perMatrix_] := (Chop@Diagonal[
  Conjugate@eigenVectors . perMatrix . Transpose[eigenVectors]])

SecondOrderPerturbation::usage="Given the eigenValues and eigenVectors of a matrix A (which doesn't need to be given) together with a corresponding perturbation matrix perMatrix, this function calculates the second derivative of the eigenvalues with respect to the scale factor of the perturbation matrix. In the sense that the eigenvalues of the matrix A + β perMatrix are to second order equal to \[Lambda] + \[Delta]_i β + \[Delta]_i^{(2)}/2 β^2, where the \[Delta]_i^{(2)} are the returned values. The eigenvalues and eigenvectors are assumed to be given in the same order, i.e. the ith eigenvalue corresponds to the ith eigenvector. This assuming that the eigenvalues are non-degenerate.";
SecondOrderPerturbation[eigenValues_, eigenVectors_, perMatrix_] := (
  dim = Length[perMatrix];
  eigenBras = Conjugate[eigenVectors];
  eigenKets = eigenVectors;
  matV = Abs[eigenBras . perMatrix . Transpose[eigenKets]]^2;
  OneOver[x_, y_] := If[x == y, 0, 1/(x - y)];
  eigenDiffs = Outer[OneOver, eigenValues, eigenValues, 1];
  pProduct = Transpose[eigenDiffs]*matV;
  Return[2*(Total /@ Transpose[pProduct])];
  )

SuperIdentity::usage="SuperIdentity[args] returns the arguments passed to it. This is useful for defining a function that does nothing, but that can be used in a composition.";
SuperIdentity[args___] := {args};

FlattenBasis::usage="FlattenBasis[basis] takes a basis in the standard representation and separates out the strings that describe the LS part of the labels and the additional numbers that define the values of J MJ and MI. It returns a list with two elements {flatbasisLS, flatbasisNums}. This is useful for saving the basis to an h5 file where the strings and numbers need to be separated.";
FlattenBasis[basis_] := Module[{flatbasis, flatbasisLS, flatbasisNums},
  (
    flatbasis = Flatten[basis];
    flatbasisLS = flatbasis[[1 ;; ;; 4]];
    flatbasisNums = Select[flatbasis, Not[StringQ[#]] &];
    Return[{flatbasisLS, flatbasisNums}]
    )
  ];

RecoverBasis::usage="RecoverBasis[{flatBasisLS, flatbasisNums}] takes the output of FlattenBasis and returns the original basis. The input is a list with two elements {flatbasisLS, flatbasisNums}.";
RecoverBasis[{flatbasisLS_, flatbasisNums_}] := Module[{recBasis},
  (
  recBasis = {{{#[[1]], #[[2]]}, #[[3]]}, #[[4]]} & /@ (Flatten /@
      Transpose[{flatbasisLS,
        Partition[Round[2*#]/2 & /@ flatbasisNums, 3]}]);
  Return[recBasis];
  )
```

```
207     ]
208
209   ExtractSymbolNames[expr_Hold] := Module[
210     {strSymbols},
211     strSymbols = ToString[expr, InputForm];
212     StringCases[strSymbols, RegularExpression["\\w+"]][[2 ;;]]
213     ]
214
215   ExportToH5::usage =
216     "ExportToH5[fname, Hold[{symbol1, symbol2, ...}]] takes an .h5
      filename and a held list of symbols and export to the .h5 file the
       values of the symbols with keys equal the symbol names. The
      values of the symbols cannot be arbitrary, for instance a list
      with mixes numbers and string will fail, but an Association with
      mixed values exports ok. Do give it a try.
217     If the file is already present in disk, this function will
      overwrite it by default. If the value of a given symbol contains
      symbolic numbers, e.g. \[Pi], these will be converted to floats in
       the exported file.";
218   Options[ExportToH5] = {"Overwrite" -> True};
219   ExportToH5[fname_String, symbols_Hold, OptionsPattern[]] := (
220     If[And[FileExistsQ[fname], OptionValue["Overwrite"]],
221     (
222       Print["File already exists, overwriting ..."];
223       DeleteFile[fname];
224       )
225     ];
226     symbolNames = ExtractSymbolNames[symbols];
227     Do[(Print[symbolName];
228       Export[fname, ToExpression[symbolName], {"Datasets", symbolName
      },
229       OverwriteTarget -> "Append"]
230       ), {symbolName, symbolNames}]
231     )
232
233   GreedyMatching::usage="GreedyMatching[aList, bList] returns a list
      of pairs of elements from aList and bList that are closest to each
       other, this is returned in a list together with a mapping of
      indices from the aList to those in bList to which they were
      matched. The option \"alistLabels\" can be used to specify labels
      for the elements in aList. The option \"blistLabels\" can be used
      to specify labels for the elements in bList. If these options are
      used, the function returns a list with three elements the pairs of
       matched elements, the pairs of corresponding matched labels, and
      the mapping of indices.";
234   Options[GreedyMatching] = {
235       "alistLabels" -> {},
236       "blistLabels" -> {}};
237   GreedyMatching[aValues0_, bValues0_, OptionsPattern[]] := Module[{
238     aValues = aValues0,
239     bValues = bValues0,
240     bValuesOriginal = bValues0,
241     bestLabels, bestMatches,
242     bestLabel, aElement, givenLabels,
243     aLabels, aLabel,
244     diffs, minDiff,
245     bLabels,
246     minDiffPosition, bestMatch},
247     (
248     aLabels     = OptionValue["alistLabels"];
249     bLabels     = OptionValue["blistLabels"];
250     bestMatches = {};
251     bestLabels  = {};
252     givenLabels = (Length[aLabels] > 0);
253     Do[
254       (
255       aElement        = aValues[[idx]];
256       diffs           = Abs[bValues - aElement];
257       minDiff         = Min[diffs];
258       minDiffPosition = Position[diffs, minDiff][[1, 1]];
259       bestMatch       = bValues[[minDiffPosition]];
260       bestMatches     = Append[bestMatches, {aElement, bestMatch}];
261       If[givenLabels,
262         (
263         aLabel     = aLabels[[idx]];
264         bestLabel  = bLabels[[minDiffPosition]];
```

```
        bestLabels = Append[bestLabels, {aLabel, bestLabel}];
        bLabels    = Drop[bLabels, {minDiffPosition}];
        )
        ];
      bValues = Drop[bValues, {minDiffPosition}];
      If[Length[bValues] == 0, Break[]];
      ),
      {idx, 1, Length[aValues]}
      ];
    pairedIndices = MapIndexed[{#2[[1]], Position[bValuesOriginal,
    #1[[2]]][[1, 1]]} &, bestMatches];
    If[givenLabels,
      Return[{bestMatches, bestLabels, pairedIndices}],
      Return[{bestMatches, pairedIndices}]
      ]
    )
    ]

  StochasticMatching::usage="StochasticMatching[aValues, bValues]
    finds a better assignment by randomly shuffling the elements of
    aValues and then applying the greedy assignment algorithm. The
    function prints what is the range of total absolute differences
    found during shuffling, the standard deviation of all of them, and
     the number of shuffles that were attempted. The option \"
    alistLabels\" can be used to specify labels for the elements in
    aValues. The option \"blistLabels\" can be used to specify labels
    for the elements in bValues. If these options are used, the
    function returns a list with three elements the pairs of matched
    elements, the pairs of corresponding matched labels, and the
    mapping of indices.";
  Options[StochasticMatching] = {"alistLabels" -> {},
    "blistLabels" -> {}};
  StochasticMatching[aValues0_, bValues0_, numShuffles_ : 200,
    OptionsPattern[]] := Module[{
    aValues = aValues0,
    bValues = bValues0,
    matchingLabels, ranger, matches, noShuff, bestMatch, highestCost,
     lowestCost, dev, sorter, bestValues,
    pairedIndices, bestLabels, matchedIndices, shuffler
    },
    (
    matchingLabels = (Length[OptionValue["alistLabels"]] > 0);
    ranger = Range[1, Length[aValues]];
    matches = If[Not[matchingLabels], (
        Table[(
          shuffler = If[i == 1, ranger, RandomSample[ranger]];
          {bestValues, matchedIndices} =
          GreedyMatching[aValues[[shuffler]], bValues];
          cost = Total[Abs[#[[1]] - #[[2]]] & /@ bestValues];
          {cost, {bestValues, matchedIndices}}
          ), {i, 1, numShuffles}]
        ),
      Table[(
        shuffler = If[i == 1, ranger, RandomSample[ranger]];
        {bestValues, bestLabels, matchedIndices} =
          GreedyMatching[aValues[[shuffler]], bValues,
          "alistLabels" -> OptionValue["alistLabels"][[shuffler]],
          "blistLabels" -> OptionValue["blistLabels"]];
        cost = Total[Abs[#[[1]] - #[[2]]] & /@ bestValues];
        {cost, {bestValues, bestLabels, matchedIndices}}
        ), {i, 1, numShuffles}]
      ];
    noShuff = matches[[1, 1]];
    matches = SortBy[matches, First];
    bestMatch = matches[[1, 2]];
    highestCost = matches[[-1, 1]];
    lowestCost = matches[[1, 1]];
    dev = StandardDeviation[First /@ matches];
    Print[lowestCost, " <-> ", highestCost, " | \[Sigma]=", dev,
      " | N=", numShuffles, " | null=", noShuff];
    If[matchingLabels,
      (
      {bestValues, bestLabels, matchedIndices} = bestMatch;
      sorter = Ordering[First /@ bestValues];
      bestValues = bestValues[[sorter]];
      bestLabels = bestLabels[[sorter]];
```

```
      pairedIndices =
        MapIndexed[{#2[[1]], Position[bValues, #1[[2]]][[1, 1]]} &,
        bestValues];
      Return[{bestValues, bestLabels, pairedIndices}]
      ),
      (
      {bestValues, matchedIndices} = bestMatch;
      sorter = Ordering[First /@ bestValues];
      bestValues = bestValues[[sorter]];
      pairedIndices =
        MapIndexed[{#2[[1]], Position[bValues, #1[[2]]][[1, 1]]} &,
        bestValues];
      Return[{bestValues, pairedIndices}]
      )
      ];
    )
    ]

  FlowMatching::usage="FlowMatching[aList, bList] returns a list of
    pairs of elements from aList and bList that are closest to each
    other, this is returned in a list together with a mapping of
    indices from the aList to those in bList to which they were
    matched. The option \"alistLabels\" can be used to specify labels
    for the elements in aList. The option \"blistLabels\" can be used
    to specify labels for the elements in bList. If these options are
    used, the function returns a list with three elements the pairs of
     matched elements, the pairs of corresponding matched labels, and
    the mapping of indices. This is basically a wrapper around
    Mathematica's FindMinimumCostFlow function. By default the option
    \"notMatched\" is zero, and this means that all elements of aList
    must be matched to elements of bList. If this is not the case, the
     option \"notMatched\" can be used to specify how many elements of
     aList can be left unmatched. By default the cost function is Abs
    [#1-#2]&, but this can be changed with the option \"CostFun\",
    this function needs to take two arguments.";
  Options[FlowMatching] = {"alistLabels" -> {}, "blistLabels" -> {},
    "notMatched" -> 0, "CostFun"-> (Abs[#1-#2] &)};
  FlowMatching[aValues0_, bValues0_, OptionsPattern[]] := Module[{
    aValues = aValues0, bValues = bValues0, edgesSourceToA,
    capacitySourceToA, nA, nB,
    costSourceToA, midLayer, midLayerEdges, midCapacities,
    midCosts, edgesBtoSink, capacityBtoSink, costBtoSink,
    allCapacities, allCosts, allEdges, graph,
    flow, bestValues, bestLabels, cFun,
    aLabels, bLabels, pairedIndices, matchingLabels},
    (
    matchingLabels = (Length[OptionValue["alistLabels"]] > 0);
    aLabels = OptionValue["alistLabels"];
    bLabels = OptionValue["blistLabels"];
    cFun    = OptionValue["CostFun"];
    nA      = Length[aValues];
    nB      = Length[bValues];
    (*Build up the edges costs and capacities*)
    (*From source to the nodes representing the values of the first \
  list*)
    edgesSourceToA    = ("source" \[DirectedEdge] {"A", #}) & /@
    Range[1, nA];
    capacitySourceToA = ConstantArray[1, nA];
    costSourceToA     = ConstantArray[0, nA];

    (*From all the elements of A to all the elements of B*)
    midLayer = Table[{{"A", i} \[DirectedEdge] ({"B", j}), 1, cFun[
    aValues[[i]], bValues[[j]]]}, {i, 1, nA}, {j, 1, nB}];
    midLayer = Flatten[midLayer, 1];
    {midLayerEdges, midCapacities, midCosts} = Transpose[midLayer];

    (*From the elements of B to the sink*)
    edgesBtoSink    = ({"B", #} \[DirectedEdge] "sink") & /@ Range[1,
     nB];
    capacityBtoSink = ConstantArray[1, nB];
    costBtoSink     = ConstantArray[0, nB];

    (*Put it all together*)
    allCapacities = Join[capacitySourceToA, midCapacities,
    capacityBtoSink];
    allCosts      = Join[costSourceToA, midCosts, costBtoSink];
```

```
    allEdges      = Join[edgesSourceToA, midLayerEdges, edgesBtoSink
   ];
    graph         = Graph[allEdges, EdgeCapacity -> allCapacities,
      EdgeCost -> allCosts];

    (*Solve it*)
    flow          = FindMinimumCostFlow[graph, "source", "sink", nA -
     OptionValue["notMatched"], "OptimumFlowData"];
    (*Collect the pairs of matched indices*)
    pairedIndices = Select[flow["EdgeList"], And[Not[#[[1]] === "
   source"], Not[#[[2]] === "sink"]] &];
    pairedIndices = {#[[1, 2]], #[[2, 2]]} & /@ pairedIndices;
    (*Collect the pairs of matched values*)
    bestValues    = {aValues[[#[[1]]]], bValues[[#[[2]]]]} & /@
   pairedIndices;
    (*Account for having been given labels*)
    If[matchingLabels,
      (
      bestLabels = {aLabels[[#[[1]]]], bLabels[[#[[2]]]]} & /@
   pairedIndices;
      Return[{bestValues, bestLabels, pairedIndices}]
      ),
      (
      Return[{bestValues, pairedIndices}]
      )
      ];
    )
    ]

  HelperNotebook::usage="HelperNotebook[nbName] creates a separate
    notebook and returns a function that can be used to print to the
    bottom of it. The name of the notebook, nbName, is optional and
    defaults to OUT.";
  HelperNotebook[nbName_:"OUT"] :=
  Module[{screenDims, screenWidth, screenHeight, nbWidth, leftMargin,
    PrintToOutputNb}, (
    screenDims =
      SystemInformation["Devices", "ScreenInformation"][[1, 2, 2]];
    screenWidth = screenDims[[1, 2]];
    screenHeight = screenDims[[2, 2]];
    nbWidth = Round[screenWidth/3];
    leftMargin = screenWidth - nbWidth;
    outputNb = CreateDocument[{}, WindowTitle -> nbName,
      WindowMargins -> {{leftMargin, Automatic}, {Automatic,
          Automatic}}, WindowSize -> {nbWidth, screenHeight}];
    PrintToOutputNb[text_] :=
      (
          SelectionMove[outputNb, After, Notebook];
          NotebookWrite[outputNb, Cell[BoxData[ToBoxes[text]], "
   Output"]];
      );
    Return[PrintToOutputNb]
    )
    ]

  GetModificationDate::usage="GetModificationDate[fname] returns the
    modification date of the given file.";
  GetModificationDate[theFileName_] := FileDate[theFileName, "
    Modification"];

  (*Helper function to convert Mathematica expressions to standard
    form*)
  StandardFormExpression[expr0_] := Module[{expr=expr0}, ToString[
    expr, InputForm]];

  (*Helper function to translate to Python/SymPy expressions*)
  ToPythonSymPyExpression::usage="ToPythonSymPyExpression[expr]
    converts a Mathematica expression to a SymPy expression. This is a
     little iffy and might break if the expression includes
    Mathematica functions that haven't been given a SymPy equivalent."
    ;
  ToPythonSymPyExpression[expr0_] := Module[{standardForm, expr=expr0
    },
    standardForm = StandardFormExpression[expr];
    StringReplace[standardForm, {
      "Power[" -> "Pow(",
```

```
      "Sqrt[" -> "sq(",
      "[" -> "(",
      "]" -> ")",
      "\\" -> "",
      "I" -> "1j",
      (*Remove special Mathematica backslashes*)
      "/" -> "/" (*Ensure division is represented with a slash*)}]];

ToPythonSparseFunction[sparseArray_SparseArray, funName_] :=
  Module[{
    rowPointers,
    dimensions,
    pyCode,
    vars,
    varList,
    dataPyList,
    colIndicesPyList
    },
   (* Extract unique symbolic variables from the SparseArray *)
   vars        = Union[Cases[Normal[sparseArray], _Symbol, Infinity
  ]];
   varList     = StringRiffle[ToString /@ vars, ", "];
   (* Convert data to SymPy compatible strings *)
   dataPyList = StringRiffle[ToPythonSymPyExpression /@ Normal[
  sparseArray["NonzeroValues"]], ",\n        "];
   colIndicesPyList = StringRiffle[
     ToPythonSymPyExpression /@ (Flatten[Normal[sparseArray["
  ColumnIndices"]] - 1]), ", "];
   (* Extract sparse array properties *)
   rowPointers = Normal[sparseArray["RowPointers"]];
   dimensions = Dimensions[sparseArray];
   (*Create Python code string*)
   pyCode = StringJoin[
     "#!/usr/bin/env python3\n\n",
     "from scipy.sparse import csr_matrix\n",
     "import numpy as np\n",
     "\n",
     "sq = np.sqrt\n",
     "\n",
     "def ", funName, "(",
     varList,
     "):\n",
     "    data = np.array([\n        ", dataPyList, "\n        ])\n"
  ,
     "    indices = np.array([",
     colIndicesPyList,
     "])\n",
     "    indptr = np.array([",
     StringRiffle[ToString /@ rowPointers, ", "], "])\n",
     "    shape = (", StringRiffle[ToString /@ dimensions, ", "],
     ")\n",
     "    return csr_matrix((data, indices, indptr), shape=shape)\n"
  ];
   pyCode
   ];

Options[HamTeX] = {"T2" -> False};
HamTeX::usage="HamTeX[numE] returns an image with parsed LaTeX code
    for the Hamiltonian of the given number of electrons. The option
   \"T2\" can be used to specify whether the T2 term should be
   included in the Hamiltonian for the f^12 configuration. The
   default is False and the option is ignored if the number of
   electrons is not 12. The function requires the MaTeX package.";
HamTeX[nE_, OptionsPattern[]] := (
  tex = Which[
    MemberQ[{1, 13}, nE],
      "\\zeta \\sum_{i=1}^{n}\\left(\\hat{s}_i \\cdot \
  \\hat{l}_i\\right) \
  +\\sum_{i=1}^n\\sum_{k=2,4,6}\\sum_{q=-k}^{k}B_q^{(k)}\\mathcal{C
  }(i)_\
  q^{(k)} + \\epsilon",
    nE == 2,
      "\\hat{H}&=\\sum_{k=2,4,6}F^{(k)}\\hat{f}_k
  +\\alpha \\hat{L}^2
  +\\beta \\,\\mathcal{C}\\left(\\mathcal{G}(2)\\right)
  +\\gamma \\,\\mathcal{C}\\left(\\mathcal{SO}(7)\\right)\\\\
```

```
    &\\quad + \\zeta \\sum_{i=1}^{n}\\left(\\hat{s}_i \\cdot \
    \\hat{l}_i\\right)
    +\\sum_{k=0,2,4}M^{(k)}\\hat{m}_k
    +\\sum_{k=2,4,6}P^{(k)}\\hat{p}_k \\\\
    &\\quad\\quad\\quad\\quad\\quad+\\sum_{i=1}^n\\sum_{k=2,4,6}\\
    sum_{q=-\
    k}^{k}B_q^{(k)}\\mathcal{C}(i)_q^{(k)} + \\epsilon",
      And[nE == 12, OptionValue["T2"]],
        "\\hat{H}&=\\sum_{k=2,4,6}F^{(k)}\\hat{f}_k
        +T^{(2)}\\hat{t}_2
        +\\alpha \\hat{L}^2
        +\\beta \\,\\mathcal{C}\\left(\\mathcal{G}(2)\\right)
        +\\gamma \\,\\mathcal{C}\\left(\\mathcal{SO}(7)\\right)\\\\
        &\\quad\\quad\\quad + \\zeta \\sum_{i=1}^{n}\\left(\\hat{s}_i
     \\cdot \
        \\hat{l}_i\\right)
        +\\sum_{k=0,2,4}M^{(k)}\\hat{m}_k
        +\\sum_{k=2,4,6}P^{(k)}\\hat{p}_k \\\\
        &\\quad\\quad\\quad\\quad\\quad\\quad\\quad+\\sum_{i=1}^n\\
    sum_{k=2,4,\
        6}\\sum_{q=-k}^{k}B_q^{(k)}\\mathcal{C}(i)_q^{(k)} + \\
    epsilon",
      And[nE == 12, Not@OptionValue["T2"]],
        "\\hat{H}&=\\sum_{k=2,4,6}F^{(k)}\\hat{f}_k
        +\\alpha \\hat{L}^2
        +\\beta \\,\\mathcal{C}\\left(\\mathcal{G}(2)\\right)
        +\\gamma \\,\\mathcal{C}\\left(\\mathcal{SO}(7)\\right)\\\\
        &\\quad + \\zeta \\sum_{i=1}^{n}\\left(\\hat{s}_i \\cdot \
        \\hat{l}_i\\right)
        +\\sum_{k=0,2,4}M^{(k)}\\hat{m}_k
        +\\sum_{k=2,4,6}P^{(k)}\\hat{p}_k \\\\
        &\\quad\\quad\\quad+\\sum_{i=1}^n\\sum_{k=2,4,6}\\sum_{q=-k
    }^{k}B_q^{(\
    k)}\\mathcal{C}(i)_q^{(k)} + \\epsilon",
      True,
        "\\hat{H}&=\\sum_{k=2,4,6}F^{(k)}\\hat{f}_k
        +\\sum_{k=2,3,4,6,7,8}T^{(k)}\\hat{t}_k
        +\\alpha \\hat{L}^2
        +\\beta \\,\\mathcal{C}\\left(\\mathcal{G}(2)\\right)
        +\\gamma \\,\\mathcal{C}\\left(\\mathcal{SO}(7)\\right)\\\\
        &\\quad\\quad\\quad\\quad + \\zeta \\sum_{i=1}^{n}\\left(\\
    hat{s}_i \\
        \\cdot \\hat{l}_i\\right)
        +\\sum_{k=0,2,4}M^{(k)}\\hat{m}_k
        +\\sum_{k=2,4,6}P^{(k)}\\hat{p}_k \\\\
        &\\quad\\quad\\quad\\quad\\quad\\quad\\quad\\quad+\\sum_{i
    =1}^n\\sum_{\
        k=2,4,6}\\sum_{q=-k}^{k}B_q^{(k)}\\mathcal{C}(i)_q^{(k)} + \\
    epsilon"
      ];
    MaTeX[StringJoin[{"\\begin{aligned}\n", tex, "\n\\end{aligned}"
    }]]
    )

  EllipsoidBoundingBox::usage = "EllipsoidBoundingBox[A,\[Kappa]]
    gives the coordinate intervals that contain the ellipsoid
    determined by r^T.A.r==\[Kappa]^2. The matrix A must be square NxN
    , symmetric, and positive definite. The function returns a list
    with N pairs of numbers, each pair being of the form {-x_i, x_i}."
    ;
  EllipsoidBoundingBox[Amat_,\[Kappa]_]:=Module[
    {invAmat, stretchFactors, boundingPlanes, quad},
      (
      invAmat = Inverse[Amat];
      stretchFactors = Sqrt[1/Diagonal[invAmat]];
      boundingPlanes = DiagonalMatrix[stretchFactors].invAmat;
      (* The solution is proportional to \[Kappa] *)
      boundingPlanes = \[Kappa] * boundingPlanes;
      boundingPlanes = Max /@ Transpose[boundingPlanes];
      Return[{-#,#}& /@ boundingPlanes]
    )
  ];

End[];
EndPackage[];
```

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[CFR68b] WT Carnall, PR Fields, and K Rajnak. "Electronic energy levels of the trivalent lanthanide aquo ions. IV. Eu3+". In: *The Journal of Chemical Physics* 49.10 (1968). Publisher: American Institute of Physics, pp. 4450–4455.

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