`qlanth`

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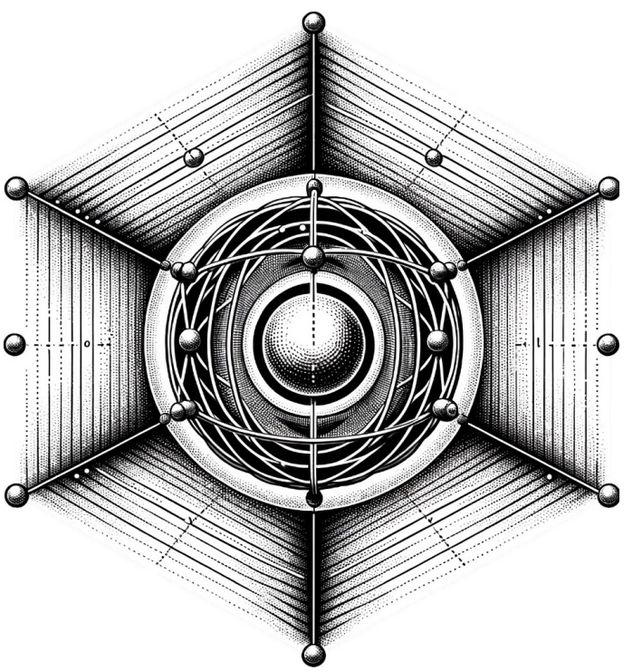

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2025 AD

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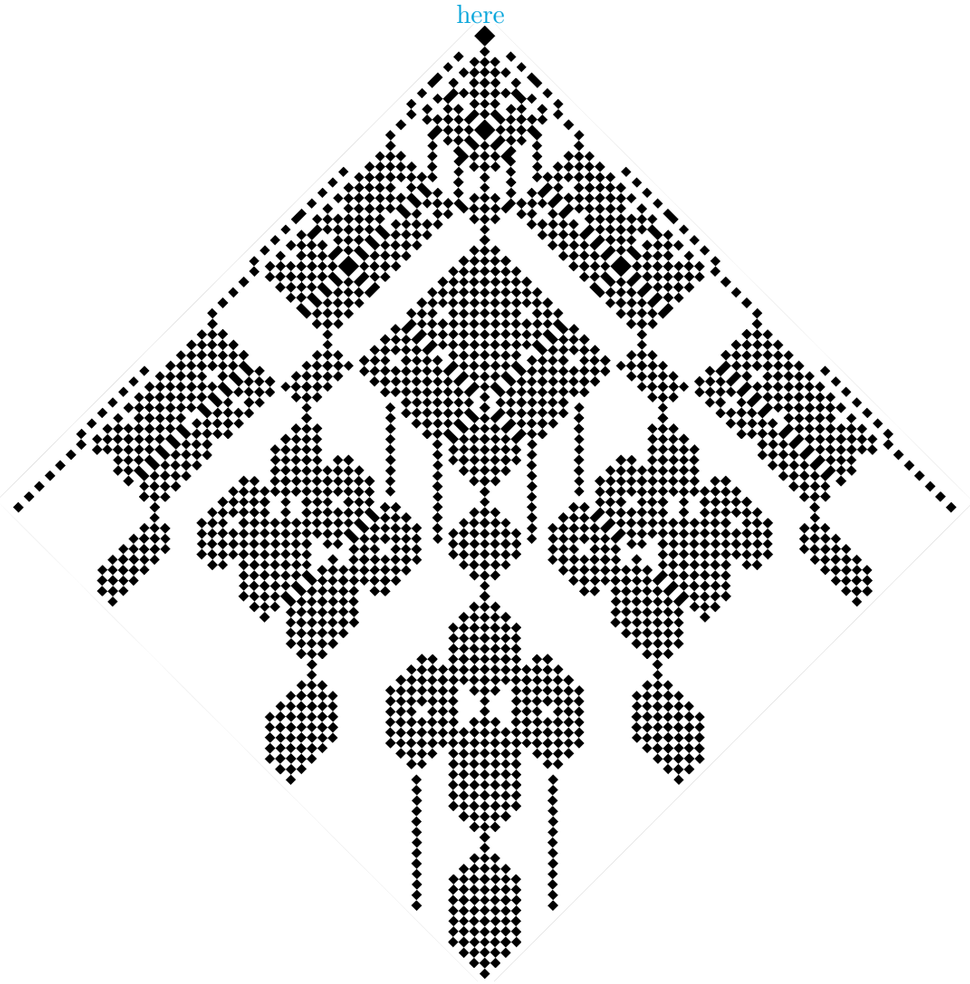

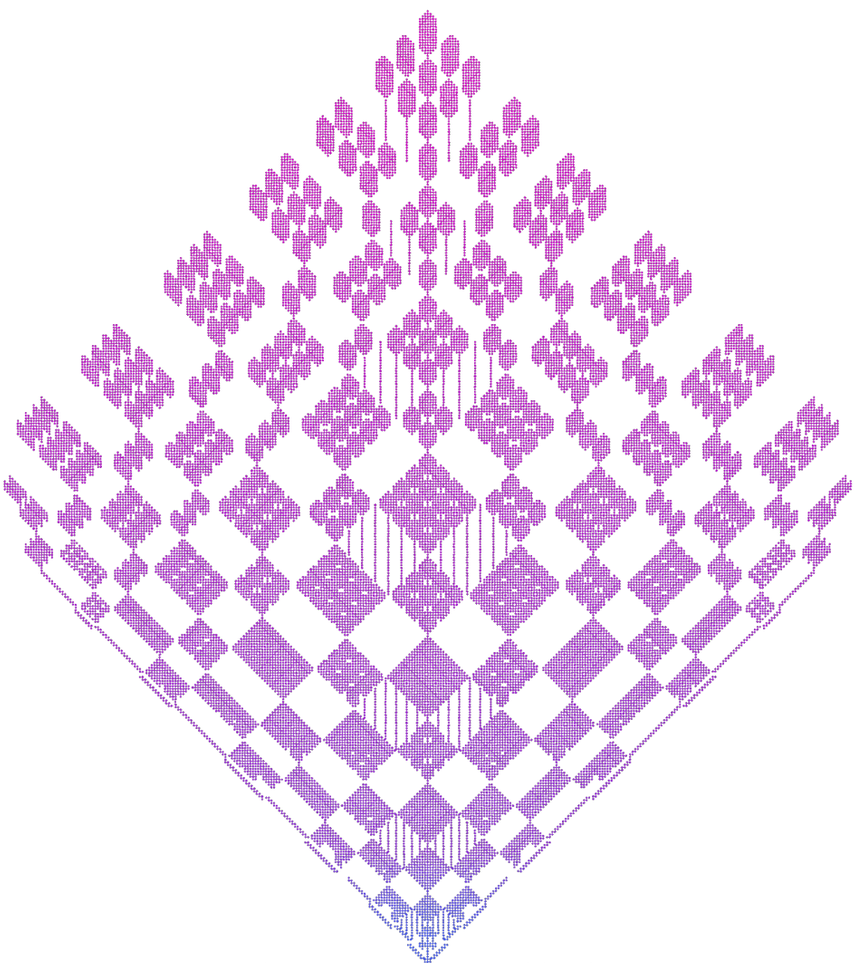

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qlanth is a tool that can be used to estimate the electronic structure of lanthanide ions in crystals. I uses an effective Hamiltonian limited to a single-configuration with included configuration-interaction corrections. This Hamiltonian aims to describe 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 18 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 17 include a summary of notation and definitions used throughout this document. Finally, Section 18 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.

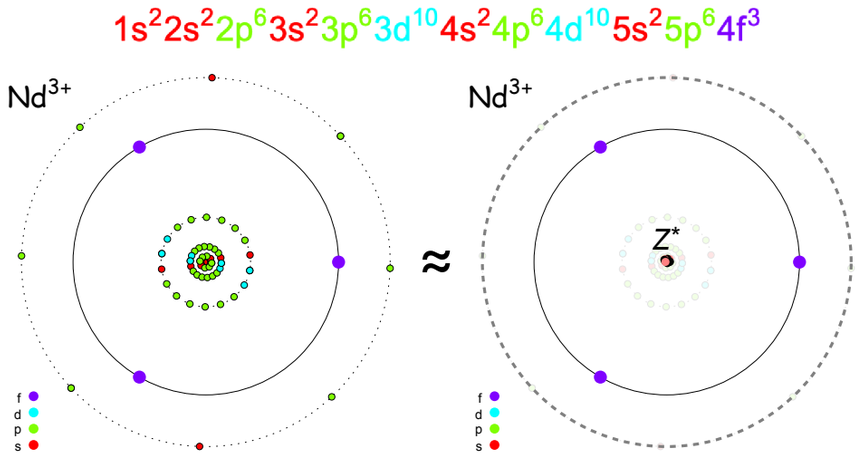

Figure 1: The trivalent neodymium ion shielded by $5s^2$ and $5p^6$ closed shells.

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 Fig-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.

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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 data what results is a Hamiltonian such as the one for Pr^{3+} in LaF_3 shown in Fig-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.

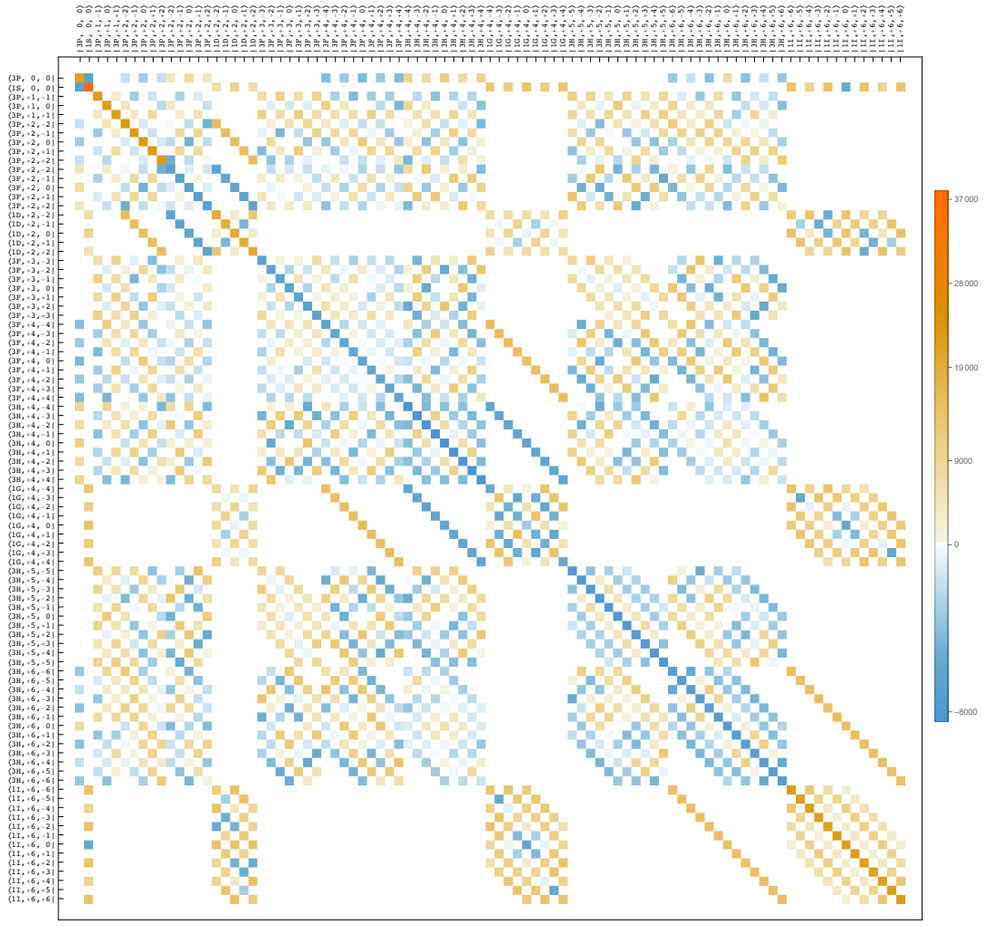

Figure 3: The matrix representation of $\hat{\mathcal{H}}$ for Pr^{3+} in LaF_3 in the $|LSJM_J\rangle$ basis.

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 $^{2S+1}\mathrm{L}$), **levels** correspond to $|LSJ\rangle$ (also noted as $^{2S+1}\mathrm{L}_J$), and **states** correspond to $|LSJM_J\rangle$ (also noted as $^{2S+1}\mathrm{L}_{J,M_J}$). Fig-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.

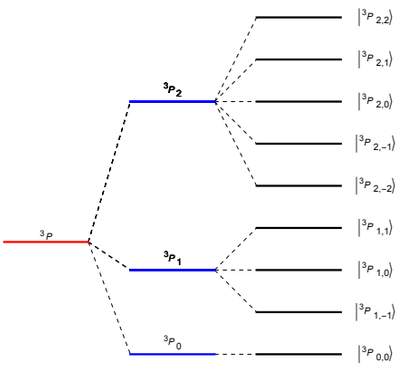

Figure 4: Levels and states associated with the ^3P term in $\underline{\mathrm{f}}^2$.

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)

---

^{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 *multiplets*. (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 subspace, 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 complementary 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 of T2 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 $\underline{f}^n$ 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 $\underline{f}^2$. 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).

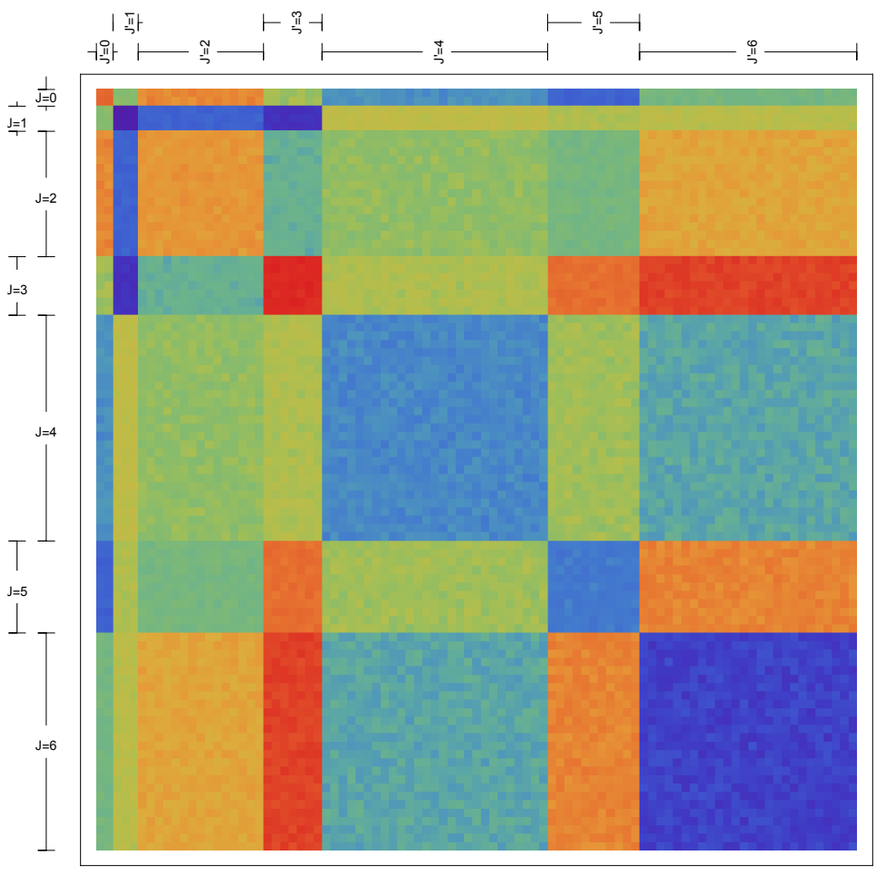


Figure 5: The J-J' block structure for $\underline{f}^2$

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^numE configuration. The matrix is
    returned as a SparseArray.
The function admits an optional parameter ''FilenameAppendix'' which
    can be used to control which variant of the JJBlocks is used to
    assemble the matrix.
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 ..."];
```

```
    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];
  )
];
```

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, &qn-?? 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 shell structure of the atom, the lanthanide ions "see" the nuclear charge as shielded by a

[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)

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];
        esub0 = fsup0;
```

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

```
        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 / TPO[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]:

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

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, 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[
  {V1k1, 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

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

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

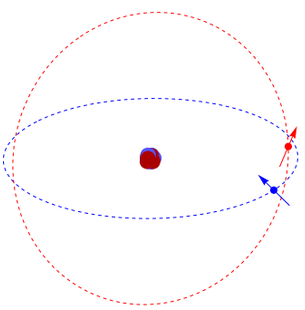
$$\langle \underline{\ell}^n W \alpha S L J M_J | \hat{\mathcal{H}}_{\mathcal{SO}(7)} | \underline{\ell}^n W \alpha' S' L' J' M_J \rangle = \gamma \delta(\alpha S L J M_J, \alpha' S' L' J' M'_J) \Lambda_{\mathcal{SO}(7)}(W) \tag{32}$$

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 done 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 $\hat{\mathscr{H}}_{\mathbf{ecs:o}}$: *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])

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

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

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

Judd *et al.* [JCC68] then go on to list the reduced matrix elements of this operator in the $\underline{\mathrm{f}}^2$ 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

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

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).

```
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 LoadChenDeltas[].";
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];
  )
];
```

```
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
     folder. 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-spin
    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 Eqn-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 done 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.
Rigorously 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 $\hat{\mathcal{H}}_{\lambda}$: 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)

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

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):

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

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]);

  (* Parse table 1 from Judd 1984 *)
```

```
judd1984Fname1 = FileNameJoin[{moduleDir, "data", "Judd1984-1.csv"}];
data = Import[judd1984Fname1, "CSV", "Numeric" -> False];
headers = data[[1]];
data = data[[2 ;;]];
data = Transpose[data];
\[Psi] = Select[data[[1]], # != "" &];
\[Psi]p = Select[data[[2]], # != "" &];
matrixKeys = Transpose[{\[Psi], \[Psi]p}];
data = data[[3 ;;]];
cols = Table[ParseJuddTab1 /@ Select[col, # != "" &], {col, data}];
cols = Select[cols, Length[#] == 21 &];
tab1 = Prepend[Prepend[cols, \[Psi]p], \[Psi]];
tab1 = Transpose[Prepend[Transpose[tab1], headers]];

(* Parse table 2 from Judd 1984 *)
judd1984Fname2 = FileNameJoin[{moduleDir, "data", "Judd1984-2.csv"}];
data = Import[judd1984Fname2, "CSV", "Numeric" -> False];
headers = data[[1]];
data = data[[2 ;;]];
data = Transpose[data];
{operatorLabels, WUlabels, multiFactorSymbols, multiFactorValues} = data[[;; 4]];
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];
);

ThreeBodyTable::usage="ThreeBodyTable is an association containing
    the LS-reduced matrix elements for the three-body operators for f^
    n configurations. The keys are lists of the form {n, SL, SpLp}.";
```

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

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

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

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

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

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

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:

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

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 $C_q^{(k)}$ 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]
    calculates the matrix element of the crystal field in terms of Bqk
     and Sqk parameters for configuration f^numE. It is calculated as
    an association with keys of the form {n, NKSL, J, M, NKSLp, Jp, Mp
    }.";
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 programmatically 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).

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

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

Crystal field expressions

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

(END) Crystal field expressions (END)

```
CrystalFieldForm::usage = "CrystalFieldForm[symmetryGroup] returns an
     association that describes the crystal field parameters that are
    necessary to describe 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 $\hat{\mathscr{H}}_{\mathbf{Z}}$: 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 $\vec{\eta}' = \left(\mathcal{M}^{-1}\right)^T \vec{\eta}$.)

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^numE configuration. The matrix is
    returned as a SparseArray.
2 The function admits an optional parameter ''FilenameAppendix'' which
    can be used to control which variant of the JJBlocks is used to
    assemble the matrix.
```

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

```
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

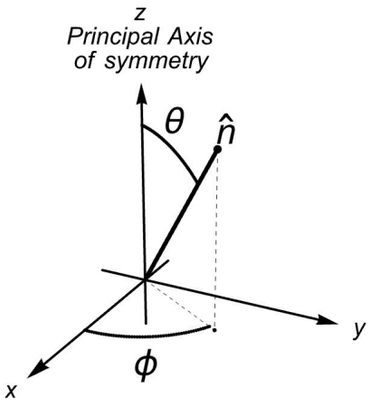
Before adding interactions lacking spherical symmetry, the orientation of the coordinate system is irrelevant. At the point when the crystal field is added, this orientation becomes relevant in the sense that only certain orientations of the coordinate system yield an expression for the crystal field 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 illuminated with a broad spectrum light source, and the wavelength dependent absorption

[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.



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 the examples included in this code 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])

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

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 $\hat{\mathcal{S}}(\psi, \psi')$.

```
1 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
    .
2 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 hPlanckHartree)) * 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]

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

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 dipole 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 $|\alpha' SL'J'\rangle$, the oscillator

[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.

strength between them is approximated as [Jud62]

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

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 association 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 levels are
    described by eigenSys in configuration f^numE. 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.
    - 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 18.2). 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 18.2.

- `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.

```
1 ClassicalFit::usage="ClassicalFit[numE, expData, excludeDataIndices,
    problemVars, startValues, constraints] 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.
2
3 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.
4
5 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.
6
```

```
7  The Hamiltonian used for fitting is a 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''.
8
9  The option ''Experimental Uncertainty in K'' 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.
10
11 The list startValues is a list with all of the parameters needed to
     define the Hamiltonian (including the initial values for
     problemVars).
12
13 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.
14
15 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.
16
17 The function returns an association with the following keys:
18
19 - ''bestRMS'' which is the best \[Sigma] value found.
20 - ''bestParams'' which is the best set of parameters found for the
     variables that were not constrained.
21 - ''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.
22 - ''paramSols'' which is a list of the parameters trajectories during
       the stepping of the fitting algorithm.
23 - ''timeTaken/s'' which is the time taken to find the best fit.
24 - ''simplifier'' which is the simplifier used to simplify the
     Hamiltonian.
25 - ''excludeDataIndices'' as given in the input.
26 - ''startValues'' as given in the input.
27
28 - ''freeIonSymbols'' which are the symbols used in the intermediate
     coupling basis.
29 - ''truncationEnergy'' which is the energy used to truncate the
     Hamiltonian, if it was set to Automatic, the value here is the
     actual energy used.
30 - ''numE'' which is the number of electrons in the f^numE
     configuration.
31 - ''expData'' which is the experimental data used for fitting.
32 - ''problemVars'' which are the symbols considered for fitting
33
34 - ''maxIterations'' which is the maximum number of iterations used by
       NMinimize.
35 - ''hamDim'' which is the dimension of the full Hamiltonian.
36 - ''allVars'' which are all the symbols defining the Hamiltonian
     under the aggregate simplifications.
37 - ''freeBies'' which are the free-ion parameters used to define the
     intermediate coupling basis.
38 - ''truncatedDim'' which is the dimension of the truncated
     Hamiltonian.
39 - ''compiledIntermediateFname'' the file name of the compiled
     function used for the truncated Hamiltonian.
40
41 - ''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, which has to match one-to-one what is provided by
     startValues.
- ''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,
  "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,
    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, solCompendium,
    openNotebooks, ordering, otherSimplifier, p0,
    paramBest, perHam,
    presentDataIndices, PrintFun, problemVarsPositions,
    problemVarsQ, problemVarsQString, problemVarsVec,
    problemVarsWithStartValues, reducedModelSymbols,
    roundedTruncationEnergy,
    runningInteractive, shiftToggle, simplifier,
    sol, solWithUncertainty,
    sortedTruncationIndex, sqdiff, standardValues,
    startTime,
    states, steps, symmetrySimplifier,
    theIntermediateEigensystems, TheIntermediateEigensystems,
    TheTruncatedAndSignedPathGenerator, timeTaken,
    truncatedIntermediateBasis, truncatedIntermediateHam,
    truncationEnergy, truncationIndices, RefParams,
    truncationUmbral, varHash,
    varsWithConstants, \[Lambda]0Vec,
    \[Lambda]exp
    },
  (
    \[Sigma]exp   = OptionValue["Energy Uncertainty in K"];
    solCompendium = <||>;
    refParamsVintage = OptionValue["RefParamsVintage"];
    RefParams = Which[
      refParamsVintage === "LaF3",
      LoadLaF3Parameters,
      refParamsVintage === "LiYF4",
      LoadLiYF4Parameters,
      True,
      refParamsVintage
    ];
    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 *)
    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  = Sort[Select[paramSymbols, Not[MemberQ[Join[
    racahSymbols, juddOfeltIntensitySymbols, chenSymbols,{t2Switch, \[
    Epsilon], gs, nE}],#]]&]];
    (* 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[""];
      )
```

```
269     ];
270
271     (* calculate the basis *)
272     PrintFun["Retrieving the LSJMJ basis for f^", numE, " ..."];
273     basis    = BasisLSJMJ[numE];
274
275     Which[refParamsVintage === Automatic,
276       (
277         PrintFun["Using the automatic vintage with freshly fitted
    free-ion parameters and others as in LaF3 ..."];
278         lnParams      = LoadLaF3Parameters[ln];
279         freeIonSol    = FreeIonSolver[expData, numE];
280         freeIonParams = freeIonSol["bestParams"];
281         lnParams      = Join[lnParams, freeIonParams];
282       ),
283       MemberQ[{List, Association}, Head[RefParams]],
284       (
285         RefParams = Association[RefParams];
286         PrintFun["Using the given parameters as a starting point ..."
    ];
287         lnParams    = RefParams;
288         extraParams = LoadLaF3Parameters[ln];
289         lnParams    = Join[extraParams, lnParams];
290       ),
291       True,
292       (
293         (* get the reference parameters from the given vintage *)
294         PrintFun["Getting reference free-ion parameters for ", ln, "
    using ", refParamsVintage, " ..."];
295         lnParams = ParamPad[RefParams[ln], "PrintFun" -> PrintFun];
296       )
297     ];
298     freeBies = Prepend[Values[(#->(#/.lnParams)) &/@ freeIonSymbols],
      numE];
299     (* a more explicit alias *)
300     allVars            = reducedModelSymbols;
301     numericConstraints = Association@Select[constraints, NumericQ
    [#[[2]]] &];
302     standardValues     = allVars /. Join[lnParams, numericConstraints
    ];
303     solCompendium["allVars"]  = allVars;
304     solCompendium["freeBies"] = freeBies;
305
306     (* reload compiled version if found *)
307     varHash                   = Hash[{numE, allVars, freeBies,
    truncationEnergy, simplifier}];
308     compiledIntermediateFname = ln <> "-compiled-intermediate-
    truncated-ham-" <> ToString[varHash] <> ".mx";
309     compiledIntermediateFname = FileNameJoin[{moduleDir, "compiled",
    compiledIntermediateFname}];
310     solCompendium["compiledIntermediateFname"] =
    compiledIntermediateFname;
311
312     If[FileExistsQ[compiledIntermediateFname],
313       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 ..."];
314       PrintFun["Using : ", compiledIntermediateFname];
315       {compileIntermediateTruncatedHam, truncatedIntermediateBasis} =
     Import[compiledIntermediateFname];,
316     (
317       If[truncationEnergy == Infinity,
318       (
319           ham = HamMatrixAssembly[numE, "ReturnInBlocks" -> False];
320           theSimplifier = simplifier;
321           ham = Normal@ReplaceInSparseArray[ham, simplifier];
322           PrintFun["Compiling a function for the Hamiltonian with no
    truncation ..."];
323           (* compile a function that will calculate the truncated
    Hamiltonian given the parameters in allVars, this is the function
    to be use in fitting *)
324           compileIntermediateTruncatedHam = Compile[Evaluate[allVars
    ], Evaluate[ham]];
325           truncatedIntermediateBasis = SparseArray@IdentityMatrix[
    Binomial[14, numE]];
```

```
326           (* save the compiled function *)
327           PrintFun["Saving the compiled function for the Hamiltonian
      with no truncation and a placeholder intermediate basis ..."];
328           Export[compiledIntermediateFname, {
      compileIntermediateTruncatedHam, truncatedIntermediateBasis}];
329         ),
330         (
331           (* grab the Hamiltonian preserving the block structure *)
332           PrintFun["Assembling the Hamiltonian for f^",numE," keeping
      the block structure ..."];
333           ham          = HamMatrixAssembly[numE, "ReturnInBlocks"->True
      ];
334           (* apply the simplifier *)
335           PrintFun["Simplifying using the aggregate set of
      simplification rules ..."];
336           ham          = Map[ReplaceInSparseArray[#, simplifier]&, ham,
       {2}];
337           PrintFun["Zeroing out every symbol in the Hamiltonian that is
       not a free-ion parameter ..."];
338           (* Get the free ion symbols *)
339           freeIonSimplifier = (#->0) & /@ Complement[
      reducedModelSymbols, freeIonSymbols];
340           (* Take the diagonal blocks for the intermediate analysis *)
341           PrintFun["Grabbing the diagonal blocks of the Hamiltonian ...
      "];
342           diagonalBlocks       = Diagonal[ham];
343           (* simplify them to only keep the free ion symbols *)
344           PrintFun["Simplifying the diagonal blocks to only keep the
      free ion symbols ..."];
345           diagonalScalarBlocks = ReplaceInSparseArray[#,
      freeIonSimplifier]&/@diagonalBlocks;
346           (* these include the MJ quantum numbers, remove that *)
347           PrintFun["Contracting the basis vectors by removing the MJ
      quantum numbers from the diagonal blocks ..."];
348           diagonalScalarBlocks = FreeHam[diagonalScalarBlocks, numE];
349
350           argsOfTheIntermediateEigensystems         = StringJoin[Riffle
      [Prepend[(ToString[#]<>"v_") & /@ freeIonSymbols,"numE_"],", "]];
351           argsForEvalInsideOfTheIntermediateSystems = StringJoin[Riffle
      [(ToString[#]<>"v") & /@ freeIonSymbols,", "]];
352           PrintFun["argsOfTheIntermediateEigensystems = ",
      argsOfTheIntermediateEigensystems];
353           PrintFun["argsForEvalInsideOfTheIntermediateSystems = ",
      argsForEvalInsideOfTheIntermediateSystems];
354           PrintFun["(if the following fails, it might help to see if
      the arguments of TheIntermediateEigensystems match the ones shown
      above)"];
355
356           (* 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 *)
357           (* compile one function for each of the blocks *)
358           PrintFun["Compiling functions for the diagonal blocks of the
      Hamiltonian ..."];
359           compiledDiagonal = Compile[Evaluate[freeIonSymbols], Evaluate
      [N[Normal[#]]]]&/@diagonalScalarBlocks;
360           (* use that to create a function that will calculate the free
      -ion eigensystem *)
361           TheIntermediateEigensystems[numEv_, F0v_, F2v_, F4v_, F6v_, ζ
      v_] := (
362             theNumericBlocks = (#[F0v, F2v, F4v, F6v, ζv]&) /@
      compiledDiagonal;
363             theIntermediateEigensystems = Eigensystem /@
      theNumericBlocks;
364             Js     = AllowedJ[numEv];
365             basisJ = BasisLSJMJ[numEv,"AsAssociation"->True];
366             (* having calculated the eigensystems with the removed
      degeneracies, put the degeneracies back in explicitly *)
367             elevatedIntermediateEigensystems = MapIndexed[EigenLever
      [#1, 2Js[[#2[[1]]]]+1 ]&, theIntermediateEigensystems];
368             (* Identify a single MJ to keep *)
369             pivot          = If[EvenQ[numEv],0,-1/2];
370             LSJmultiplets = (#[[1]]<>ToString[InputForm[#[[2]]]])&/
      @Select[BasisLSJMJ[numEv],#[[-1]]== pivot &];
371             (* calculate the multiplet assignments that the
      intermediate basis eigenvectors have *)
```

```
372           needlePosition = 0;
373           multipletAssignments = Table[
374             (
375               J          = Js[[idx]];
376               eigenVecs = theIntermediateEigensystems[[idx]][[2]];
377               majorComponentIndices         = Ordering[Abs[#]][[-1]]&/
    @eigenVecs;
378               majorComponentIndices       += needlePosition;
379               needlePosition              += Length[
    majorComponentIndices];
380               majorComponentAssignments    = LSJmultiplets[[#]]&/
    @majorComponentIndices;
381               (* All of the degenerate eigenvectors belong to the
    same multiplet*)
382               elevatedMultipletAssignments = ListRepeater[
    majorComponentAssignments, 2J+1];
383               elevatedMultipletAssignments
384             ),
385           {idx, 1, Length[Js]}
386           ];
387           (* put together the multiplet assignments and the energies
    *)
388           freeIenergiesAndMultiplets = Transpose/@Transpose[{First/
    @elevatedIntermediateEigensystems, multipletAssignments}];
389           freeIenergiesAndMultiplets = Flatten[
    freeIenergiesAndMultiplets, 1];
390           (* calculate the change of basis matrix using the
    intermediate coupling eigenvectors *)
391           basisChanger = BlockDiagonalMatrix[Transpose/@Last/
    @elevatedIntermediateEigensystems];
392           basisChanger = SparseArray[basisChanger];
393           Return[{theIntermediateEigensystems, multipletAssignments,
    elevatedIntermediateEigensystems, freeIenergiesAndMultiplets,
    basisChanger}]
394         );
395
396         PrintFun["Calculating the intermediate eigensystems for ",ln,
    " using free-ion params from LaF3 ..."];
397         (* calculate intermediate coupling basis using the free-ion
    params for LaF3 *)
398         {theIntermediateEigensystems, multipletAssignments,
    elevatedIntermediateEigensystems, freeIenergiesAndMultiplets,
    basisChanger} = TheIntermediateEigensystems@@freeBies;
399
400         (* use that intermediate coupling basis to compile a function
     for the full Hamiltonian *)
401         allFreeEnergies = Flatten[First/
    @elevatedIntermediateEigensystems];
402         (* important that the intermediate coupling basis have
    attached energies, which make possible the truncation *)
403         ordering = Ordering[allFreeEnergies];
404         (* sort the free ion energies and determine which indices
    should be included in the truncation *)
405         allFreeEnergiesSorted          = Sort[allFreeEnergies];
406         {minFreeEnergy, maxFreeEnergy} = MinMax[allFreeEnergies];
407         (* determine the index at which the energy is equal or larger
     than the truncation energy *)
408         sortedTruncationIndex = Which[
409           truncationEnergy > (maxFreeEnergy-minFreeEnergy),
410           hamDim,
411           True,
412           FirstPosition[allFreeEnergiesSorted - Min[
    allFreeEnergiesSorted],x_/;x>truncationEnergy,{0},1][[1]]
413         ];
414         (* the actual energy at which the truncation is made *)
415         roundedTruncationEnergy = allFreeEnergiesSorted[[
    sortedTruncationIndex]];
416
417         (* the indices that participate in the truncation *)
418         truncationIndices = ordering[[;;sortedTruncationIndex]];
419         PrintFun["Computing the block structure of the change of
    basis array ..."];
420         blockSizes          = BlockArrayDimensionsArray[ham];
421         basisChangerBlocks = ArrayBlocker[basisChanger, blockSizes];
422         blockShifts         = First /@ Diagonal[blockSizes];
423         numBlocks           = Length[blockSizes];
```

```
424         (* using the ham (with all the symbols) change the basis to
    the computed one *)
425         PrintFun["Changing the basis of the Hamiltonian to the
    intermediate coupling basis ..."];
426         intermediateHam = BlockMatrixMultiply[ham, basisChangerBlocks
    ];
427         PrintFun["Distributing products inside of symbolic matrix
    elements to keep complexity in check ..."];
428         Do[
429           intermediateHam[[rowIdx, colIdx]] = MapToSparseArray[
    intermediateHam[[rowIdx, colIdx]], Distribute /@ # &];,
430           {rowIdx, 1, numBlocks},
431           {colIdx, 1, numBlocks}
432         ];
433         intermediateHam = BlockMatrixMultiply[BlockTranspose[
    basisChangerBlocks],intermediateHam];
434         PrintFun["Distributing products inside of symbolic matrix
    elements to keep complexity in check ..."];
435         Do[
436           intermediateHam[[rowIdx, colIdx]] = MapToSparseArray[
    intermediateHam[[rowIdx, colIdx]], Distribute /@ # &];,
437           {rowIdx, 1, numBlocks},
438           {colIdx, 1, numBlocks}
439         ];
440         (* using the truncation indices truncate that one *)
441         PrintFun["Truncating the Hamiltonian ..."];
442         truncatedIntermediateHam = TruncateBlockArray[intermediateHam
    , truncationIndices, blockShifts];
443         (* these are the basis vectors for the truncated hamiltonian
    *)
444         PrintFun["Saving the truncated intermediate basis ..."];
445         truncatedIntermediateBasis = basisChanger[[All,
    truncationIndices]];
446
447         PrintFun["Compiling a function for the truncated Hamiltonian
    ..."];
448         (* compile a function that will calculate the truncated
    Hamiltonian given the parameters in allVars, this is the function
    to be use in fitting *)
449         compileIntermediateTruncatedHam = Compile[Evaluate[allVars],
    Evaluate[truncatedIntermediateHam]];
450         (* save the compiled function *)
451         PrintFun["Saving the compiled function for the truncated
    Hamiltonian and the truncated intermediate basis ..."];
452         Export[compiledIntermediateFname, {
    compileIntermediateTruncatedHam, truncatedIntermediateBasis}];
453       )
454     ];
455     )
456     ];
457
458     truncationUmbral = Dimensions[truncatedIntermediateBasis][[2]];
459     PrintFun["The truncated Hamiltonian has a dimension of ",
    truncationUmbral, "x", truncationUmbral, " ..."];
460     presentDataIndices = Select[presentDataIndices, # <=
    truncationUmbral &];
461     solCompendium["presentDataIndices"] = presentDataIndices;
462
463     (* the problemVars are the symbols that will be fitted for *)
464
465     PrintFun["Starting up the fitting process using the Levenberg-
    Marquardt method ..."];
466     (* using the problemVars I need to create the argument list
    including _?NumericQ *)
467     problemVarsQ       = (ToString[#] <> "_?NumericQ") & /@
    problemVars;
468     problemVarsQString = StringJoin[Riffle[problemVarsQ, ", "]];
469     (* we also need to have the padded arguments with the variables
    in the right position and the fixed values in the remaining ones
    *)
470     problemVarsPositions = Position[allVars, #][[1, 1]] & /@
    problemVars;
471     problemVarsString    = StringJoin[Riffle[ToString /@ problemVars,
     ", "]];
472     (* 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 *)
eigenvalueDispenserTemplate = StringTemplate["
PartialHamEigenvalues[`problemVarsQ`][i_]:=(
  eigenVals = HamSortedEigenvalues[`problemVarsString`];
  eigenVals[[i]]
)
"];
eigenValueDispenserString = eigenvalueDispenserTemplate[<|
    "problemVarsQ"      -> problemVarsQString,
    "problemVarsString" -> problemVarsString
    |>];
ToExpression[eigenValueDispenserString];

PrintFun["Determining the free variables after constraints ..."];
constrainedProblemVars     = (problemVars /. constraints);
constrainedProblemVarsList = Variables[constrainedProblemVars];
If[addShift,
  PrintFun["Adding a constant shift to the fitting parameters ...
"];
  constrainedProblemVarsList = Append[constrainedProblemVarsList,
 \[Epsilon]]
];

indepVars = Complement[problemVars, #[[1]] & /@ constraints];
stringPartialVars = ToString/@constrainedProblemVarsList;

paramSols  = {};
rmsHistory = {};
steps      = 0;
problemVarsWithStartValues = KeyValueMap[{#1,#2} &, startValues];
If[addShift,
  problemVarsWithStartValues = Append[problemVarsWithStartValues,
 {\[Epsilon] ,0}];
];
openNotebooks     = If[runningInteractive,
          ("WindowTitle"/.NotebookInformation[#]) & /@ Notebooks
[],
          {}];
If[Not[MemberQ[openNotebooks,"Solver Progress"]] && OptionValue["
ProgressView"],
  ProgressNotebook["Basic"->False]
];
degressOfFreedom = Length[presentDataIndices] - Length[
problemVars] - 1;
PrintFun["Fitting for ", Length[presentDataIndices], " data
points with ", Length[problemVars], " free parameters.", " The
effective degrees of freedom are ", degressOfFreedom, " ..."];

```

```
PrintFun["Fitting model to data ..."];
startTime   = Now;
shiftToggle = If[addShift, 1, 0];
sol = FindMinimum[
  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];

```

```
601     PrintFun["Calculating the eigenvalue vector at solution ..."];
602     \[Lambda]0Vec = Transpose[{eigenEnergies[[presentDataIndices]]}];
603     PrintFun["Putting together the experimental vector ..."];
604     \[Lambda]exp = Transpose[{First /@ expData[[presentDataIndices
        ]]}];
605     problemVarsVec = If[addShift,
606       Transpose[{constrainedProblemVarsList[[;;-2]]}],
607       Transpose[{constrainedProblemVarsList}]
608     ];
609     indepSolVecVec = Transpose[{indepSolVec}];
610     PrintFun["Calculating the difference between eigenvalues at
        solution ..."];
611     diff = If[linMat == {},
612       (\[Lambda]0Vec - \[Lambda]exp)  + \[Epsilon]Best,
613       (\[Lambda]0Vec - \[Lambda]exp)  + \[Epsilon]Best + linMat[[
        presentDataIndices]].(problemVarsVec - indepSolVecVec)
614     ];
615     PrintFun["Calculating the sum of squares of differences around
        solution ... "];
616     sqdiff         = Expand[(Transpose[diff] . diff)[[1, 1]]];
617     PrintFun["Calculating the minimum (which should coincide with sol
        ) ..."];
618     minpoly        = sqdiff /. AssociationThread[problemVars -> solVec
        ];
619     fmSolAssoc     = Association[sol[[2]]];
620     If[\[Sigma]exp == Automatic,
621       \[Sigma]exp = Sqrt[minpoly / degressOfFreedom];
622     ];
623     \[CapitalDelta]\[Chi]2 = Sqrt[degressOfFreedom];
624     Amat = (1/\[Sigma]exp^2) * Transpose[linMat[[presentDataIndices
        ]]].linMat[[presentDataIndices]];
625     paramIntervals = EllipsoidBoundingBox[Amat, \[CapitalDelta]\[Chi
        ]2];
626     PrintFun["Calculating the uncertainty in the parameters ..."];
627     solWithUncertainty = Table[
628       (
629         aVar         = constrainedProblemVarsList[[varIdx]];
630         paramBest    = aVar /. fmSolAssoc;
631         (aVar -> {paramBest, paramIntervals[[varIdx, 2]]})
632       ),
633     {varIdx, 1, Length[constrainedProblemVarsList]-shiftToggle}
634     ];
635
636     bestRMS     = Sqrt[minpoly / degressOfFreedom];
637     bestParams = sol[[2]];
638     bestWithConstraints = Association@Join[constraints, bestParams];
639     bestWithConstraints = bestWithConstraints /. bestWithConstraints;
640     bestWithConstraints = (# + 0.) & /@ bestWithConstraints;
641
642     solCompendium["degreesOfFreedom"]     = degressOfFreedom;
643     solCompendium["solWithUncertainty"]   = solWithUncertainty;
644     solCompendium["truncatedDim"]         = truncationUmbral;
645     solCompendium["fittedLevels"]         = Length[presentDataIndices
        ];
646     solCompendium["actualSteps"]          = steps;
647     solCompendium["bestRMS"]              = bestRMS;
648     solCompendium["problemVars"]          = problemVars;
649     solCompendium["paramSols"]            = paramSols;
650     solCompendium["rmsHistory"]           = rmsHistory;
651     solCompendium["Appendix"]             = OptionValue["
        AppendToLogFile"];
652     solCompendium["timeTaken/s"]          = timeTaken;
653     solCompendium["bestParams"]           = bestParams;
654     solCompendium["bestParamsWithConstraints"] = bestWithConstraints;
655
656     If[OptionValue["SaveEigenvectors"],
657         solCompendium["states"] = {#[[1]] + \[Epsilon]Best, #[[2]]}
        &/@ (Chop /@ ShiftedLevels[states]),
658         (
659           finalEnergies = Sort[First /@ states];
660           finalEnergies = finalEnergies - finalEnergies[[1]];
661           finalEnergies = finalEnergies + \[Epsilon]Best;
662           finalEnergies  = Chop /@ finalEnergies;
663           solCompendium["energies"] = finalEnergies;
664       )
665     ];
```

```
    If[OptionValue["SaveToLog"],
      PrintFun["Saving the solution to the log file ..."];
      LogSol[solCompendium, logFilePrefix];
    ];
    PrintFun["Finished ..."];
    Return[solCompendium];
  )
];
```

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. These notebooks assume that the `paclet` has been installed according to the instructions given in Section 16.

- `/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.

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}
    ";
```

```
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];
  ];
);
```

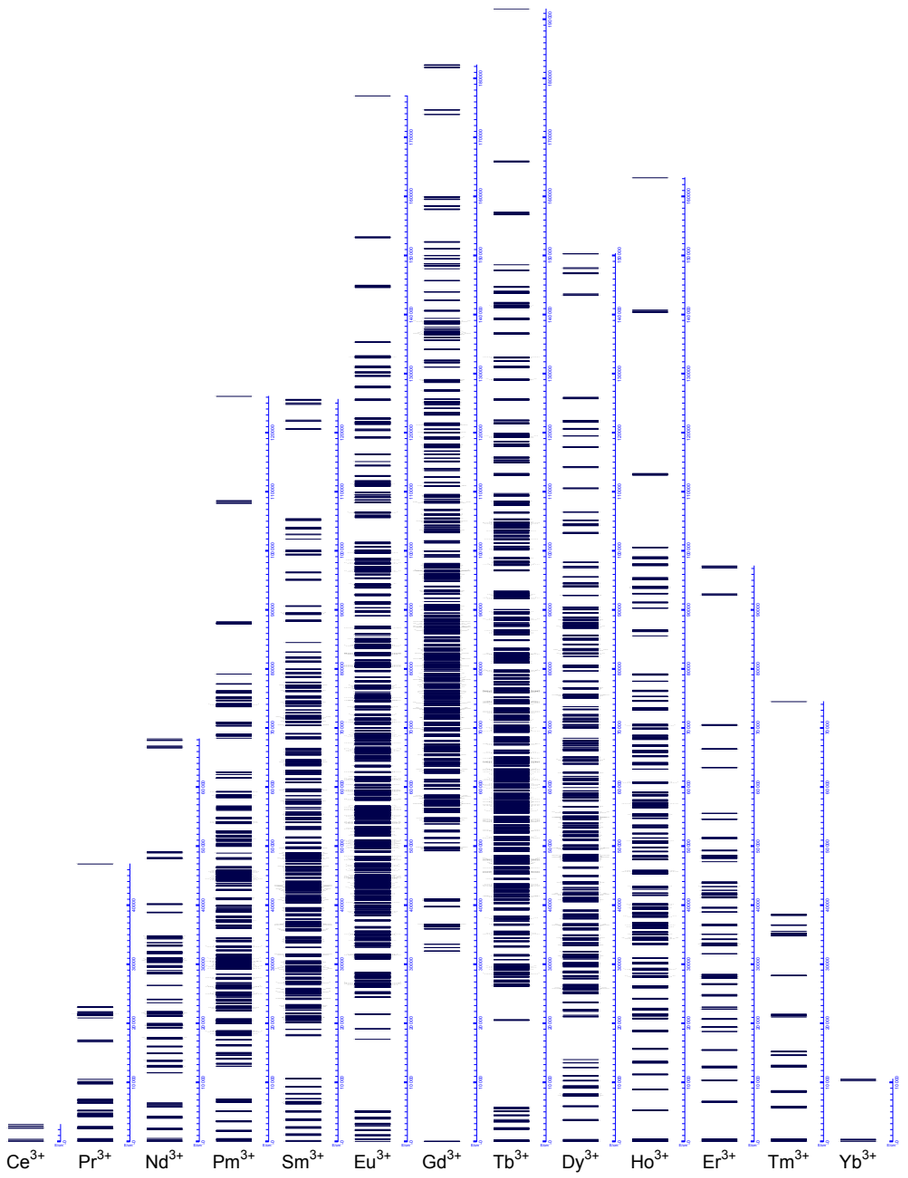


Figure 6: Energy levels in LaF_3.

```
LoadLaF3Parameters::usage="LoadLaF4Parameters[ln] gives the models
    for the semi-empirical Hamiltonian for the trivalent lanthanide
    ion with symbol ln.";
Options[LoadLaF3Parameters] = {
  "Vintage"  -> "Standard",
  "With Uncertainties"->False
};
LoadLaF3Parameters[Ln_String, OptionsPattern[]]:= Module[
  {paramData, params},
  (
    paramData = Which[
      OptionValue["Vintage"] == "Carnall",
      Import[FileNameJoin[{moduleDir, "data", "carnallParams.m"}]],
      OptionValue["Vintage"] == "Standard",
      Import[FileNameJoin[{moduleDir, "data", "standardParams.m"}]]
    ];
  params = If[OptionValue["With Uncertainties"],
    paramData[Ln],
    If[Head[#] === Around, #["Value"], #] & /@ paramData[Ln]
  ];
  Return[params];
  )
];
```

```
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
```

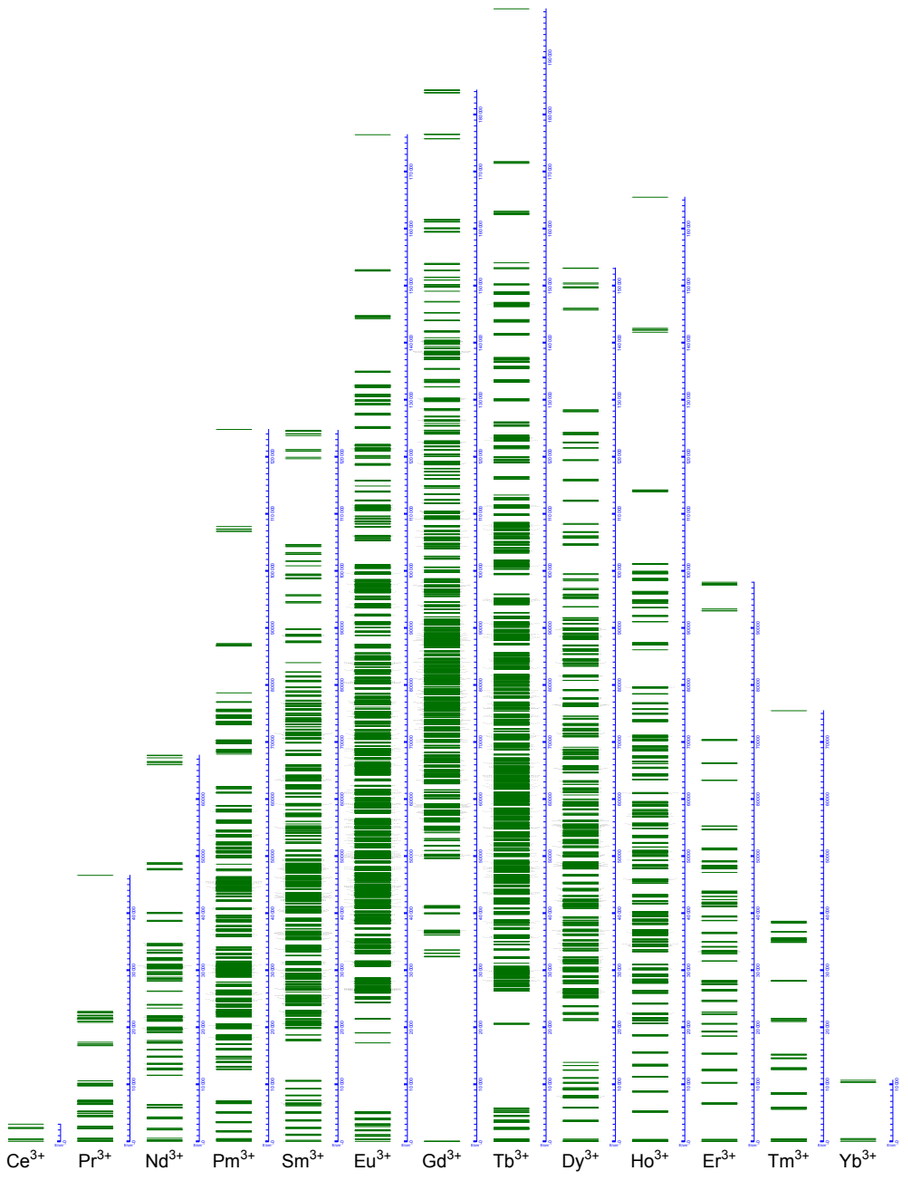


Figure 7: Energy levels in $LiYF_4$.

```
    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 14.1 on MacOS Sequoia 15.2.

14 Units

Following the tradition of the spectroscopic community, all the matrix elements of the Hamiltonian are calculated using the Kayser ($\mathcal{K} \equiv \mathrm{cm}^{-1}$) 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["DavidLizarazo`qonstants`"];

(* 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

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

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 Mathematica `paclet`

The simplest way of adding **qlanth** to *Mathematica* is by installing the corresponding paclet. After having cloned the repository to a local machine, execute the code below in a *Mathematica* session. After this, the downloaded files may be removed, and **qlanth** may be loaded into a notebook by issuing the command `Needs["DavidLizarazo`qlanth`"]` .

```
Needs["PacletTools‘"];
pathToRepo = "~/Downloads/qlanth/"; (* change accordingly *)
pathToBuild = FileNameJoin[{pathToRepo,"build","DavidLizarazo__qlanth"}];
CreatePacletArchive[pathToBuild];
pathToPaclet = FileNames[
```

```
FileNameJoin[{pathToRepo, "build", "/*.paclet"}][[1]];
PacletInstall[pathToPaclet, ForceVersionInstall -> True];
```

The paclet includes documentation in standard *Mathematica* format (see Fig-8a).

17 Definitions

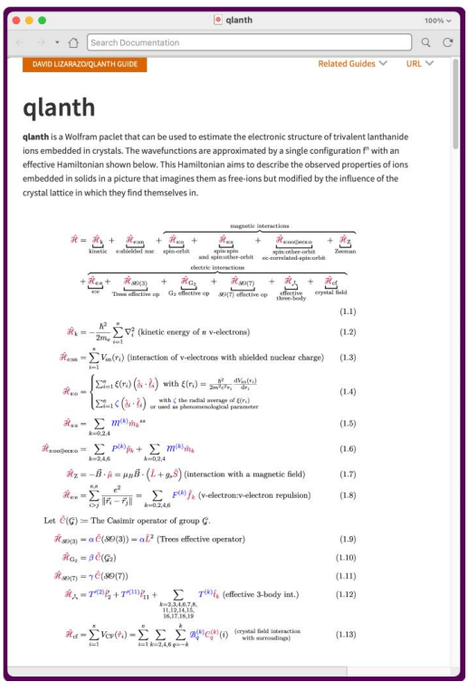
$$\overbrace{[\![x]\!]}^{\text{two plus one}} := 2x+1 \tag{135}$$

$$\overbrace{\hat{u}^{(k)}}^{\text{irreducible unit tensor operator of rank k}} \tag{136}$$

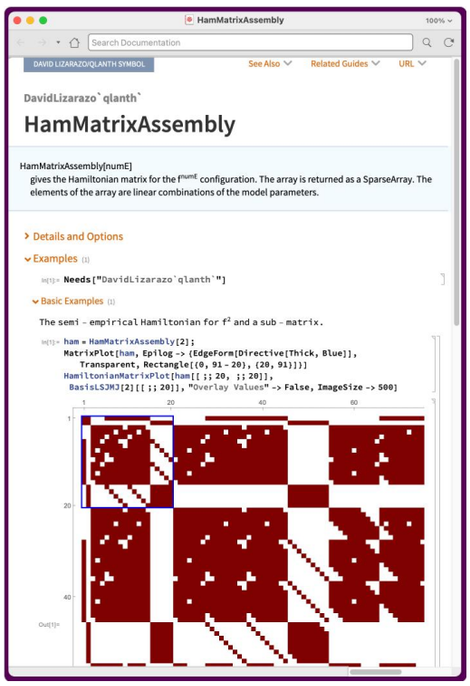
$$\overbrace{\hat{U}^{(k)} := \sum_{i=1}^{n} \hat{u}^{(k)}}^{\text{symmetric unit tensor operator for n equivalent electrons}} \tag{137}$$

$$\overbrace{C_q^{(k)} := \sqrt{\frac{4\pi}{2k+1}} \mathrm{Y}_q^{(k)}}^{\text{Renormalized spherical harmonics}} \tag{138}$$

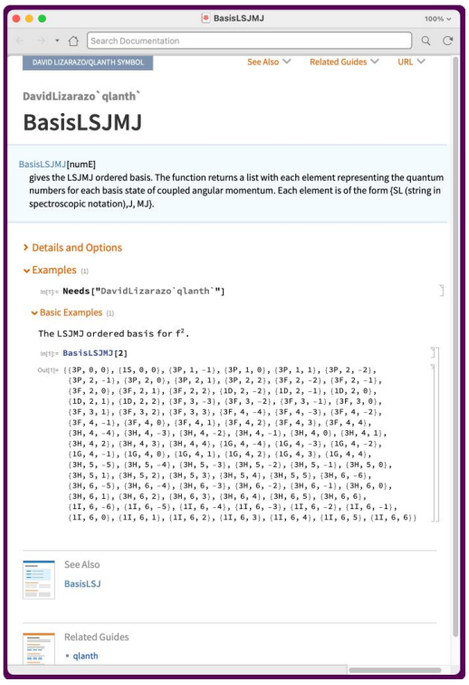
$$\overbrace{\triangleleft(j_1,j_2,j_3) := \begin{cases} 1 & \text{if } j_1 = (j_2+j_3),(j_2+j_3-1),\ldots,|j_2-j_3| \\ 0 & \text{otherwise} \end{cases}}^{\text{Triangle “delta” between } j_1,j_2,j_3} \tag{139}$$



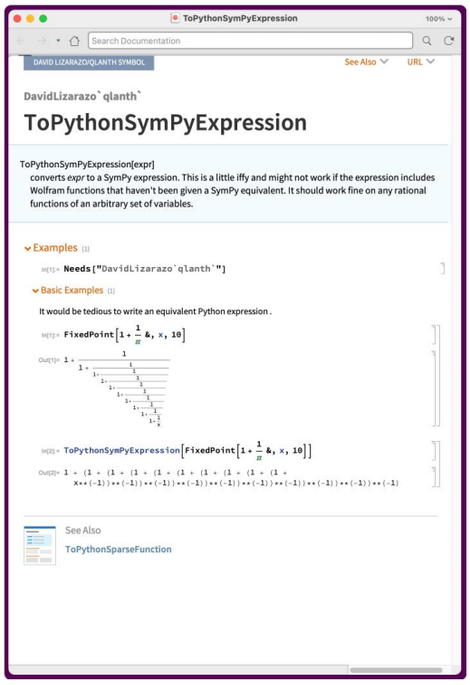
(a) **qlanth** guide in *Mathematica*



(b) Documentation for `HamMatrixAssembly`



(c) Doc. for `BasisLSJMJ`



(d) Doc. for `ToPythonSymPyExpression`

18 code

18.1 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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------------------------------------------------------------------- *)

BeginPackage["DavidLizarazo`qlanth`"];
Get[FileNameJoin[{DirectoryName[$InputFileName], "qonstants.m"}]]
Get[FileNameJoin[{DirectoryName[$InputFileName], "misc.m"}]]

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

```

```
ζ: spin-orbit strength parameter.

F0: Direct Slater integral F^0, produces an overall shift of all
    energy levels.
F2: Direct Slater integral F^2
F4: Direct Slater integral F^4, possibly constrained by ratio to F^2
F6: Direct Slater integral F^6, possibly constrained by ratio to F^2

M0: 0th Marvin integral
M2: 2nd Marvin integral
M4: 4th Marvin integral
\[Sigma]SS: spin-spin override, if 0 spin-spin is omitted, if 1 then
    spin-spin is included

T2:  three-body effective operator parameter T^2  (non-orthogonal)
T2p: three-body effective operator parameter T^2' (orthogonalized T2)
T3:  three-body effective operator parameter T^3
T4:  three-body effective operator parameter T^4
T6:  three-body effective operator parameter T^6
T7:  three-body effective operator parameter T^7
T8:  three-body effective operator parameter T^8

T11p: three-body effective operator parameter T^11' (orthogonalized
    T11)
T12:  three-body effective operator parameter T^12
T14:  three-body effective operator parameter T^14
T15:  three-body effective operator parameter T^15
T16:  three-body effective operator parameter T^16
T17:  three-body effective operator parameter T^17
T18:  three-body effective operator parameter T^18
T19:  three-body effective operator parameter T^19

P0: pseudo-magnetic parameter P^0
P2: pseudo-magnetic parameter P^2
P4: pseudo-magnetic parameter P^4
P6: pseudo-magnetic parameter P^6

gs: electronic gyromagnetic ratio

α: Trees' parameter α describing configuration interaction via the
    Casimir operator of SO(3)
β: Trees' parameter β describing configuration interaction via the
    Casimir operator of G(2)
γ: Trees' parameter γ describing configuration interaction via the
    Casimir operator of SO(7)

B02: crystal field parameter B_0^2 (real)
B04: crystal field parameter B_0^4 (real)
B06: crystal field parameter B_0^6 (real)
B12: crystal field parameter B_1^2 (real)
B14: crystal field parameter B_1^4 (real)

B16: crystal field parameter B_1^6 (real)
B22: crystal field parameter B_2^2 (real)
B24: crystal field parameter B_2^4 (real)
B26: crystal field parameter B_2^6 (real)
B34: crystal field parameter B_3^4 (real)

B36: crystal field parameter B_3^6 (real)
B44: crystal field parameter B_4^4 (real)
B46: crystal field parameter B_4^6 (real)
B56: crystal field parameter B_5^6 (real)
B66: crystal field parameter B_6^6 (real)

S12: crystal field parameter S_1^2 (real)
S14: crystal field parameter S_1^4 (real)
S16: crystal field parameter S_1^6 (real)
S22: crystal field parameter S_2^2 (real)

S24: crystal field parameter S_2^4 (real)
S26: crystal field parameter S_2^6 (real)
S34: crystal field parameter S_3^4 (real)
S36: crystal field parameter S_3^6 (real)

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;
Bqk;
CFP;

CFPAssoc;
CFPTable;
CFPTerms;
Carnall;
CasimirG2;

CasimirSO3;
CasimirSO7;
Ck;
Cqk;
CrystalField;
CrystalFieldForm;

Dk;
EigenLever;
Electrostatic;
ElectrostaticConfigInteraction;
ElectrostaticTable;

EnergyLevelDiagram;
EnergyStates;
EtoF;
ExportMZip;

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

GG2U;
GSO7W;
GenerateCFP;
GenerateCFPAssoc;
GenerateCFPTable;

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

GenerateSOOandECSOTable;
GenerateSpinOrbitTable;
GenerateSpinSpinTable;
GenerateT22Table;
GenerateThreeBodyTables;

```

```
GroundMagDipoleOscillatorStrength;
HamMatrixAssembly;
HamiltonianForm;

HamiltonianMatrixPlot;
HoleElectronConjugation;
ImportMZip;
IonSolver;
JJBlockMagDip;

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

LevelElecDipoleOscillatorStrength;
LevelJJBlockMagDipole;
LevelMagDipoleLineStrength;
LevelMagDipoleMatrixAssembly;
LevelMagDipoleOscillatorStrength;

LevelMagDipoleSpontaneousDecayRates;
LevelSimplerSymbolicHamMatrix;
LevelSolver;
LoadAll;

LoadCFP;
LoadCarnall;
LoadChenDeltas;
LoadElectrostatic;
LoadFreeIon;

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

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;

ReducedV1k;
ReducedV1kTable;
Reducedt11inf2;
ReplaceInSparseArray;
ScalarLSJMFromLS;
SOOandECSO;
SOOandECSOLSTable;

SOOandECSOTable;
Seniority;
ShiftedLevels;
SimplerSymbolicHamMatrix;

SixJay;
SpinOrbit;
SpinOrbitTable;
SpinSpin;
SpinSpinTable;

Sqk;
SquarePrimeToNormal;
TPO;
T22Table;
TabulateJJBlockMagDipTable;
TabulateJJBlockMatrixTable;

TabulateManyJJBlockMagDipTables;
TabulateManyJJBlockMatrixTables;
ThreeBodyTable;
ThreeBodyTables;
ThreeJay;

chenDeltas;
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`"]

  ListRepeater;
  TotalCFIters;

  moduleDir = ParentDirectory[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, 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
  "}];
  If[OptionValue["Export"],
    (
      Print["Exporting to file "<>ToString[exportFname]];
```

```
740         Export[exportFname, ReducedV1kTable];
741       )
742     ];
743     Return[ReducedV1kTable];
744   )
745
746   (* ###################### Racah Algebra ###################### *)
747   (* ########################################################### *)
748
749   (* ########################################################### *)
750   (* ###################### Electrostatic ###################### *)
751
752   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.";
753   fsubk[numE_, orbital_, NKSL_, NKSLp_, k_] := Module[
754     {terms, S, L, Sp, Lp,
755     termsWithSameSpin, SL,
756     fsubkVal, spinMultiplicity,
757     prefactor, summand1, summand2},
758     (
759       {S, L}   = FindSL[NKSL];
760       {Sp, Lp} = FindSL[NKSLp];
761       terms    = AllowedNKSLTerms[numE];
762       (* sum for summand1 is over terms with same spin *)
763       spinMultiplicity  = 2*S + 1;
764       termsWithSameSpin = StringCases[terms, ToString[
      spinMultiplicity] ~~ __];
765       termsWithSameSpin = Flatten[termsWithSameSpin];
766       If[Not[{S, L} == {Sp, Lp}],
767         Return[0]
768       ];
769       prefactor = 1/2 * Abs[Ck[orbital, k]]^2;
770       summand1 = Sum[(
771           ReducedUkTable[{numE, orbital, SL, NKSL,  k}] *
772           ReducedUkTable[{numE, orbital, SL, NKSLp, k}]
773           ),
774         {SL, termsWithSameSpin}
775       ];
776       summand1 = 1 / TPO[L] * summand1;
777       summand2 = (
778         KroneckerDelta[NKSL, NKSLp] *
779           (numE *(4*orbital + 2 - numE)) /
780           ((2*orbital + 1) * (4*orbital + 1))
781         );
782       fsubkVal = prefactor*(summand1 - summand2);
783       Return[fsubkVal];
784     )
785   ];
786
787   fsupk::usage = "fsupk[numE, orbital, SL, SLp, k] gives the
        superscripted Slater integral f^k = Subscript[f, k] * Subscript[D,
         k].";
788   fsupk[numE_, orbital_, NKSL_, NKSLp_ ,k_] := (
789     Dk[k] * fsubk[numE, orbital, NKSL, NKSLp, k]
790     )
791
792   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.";
793   Dk[k_] := {1, 225, 1089, 184041/25}[[k/2+1]];
794
795   FtoE::usage = "FtoE[F0, F2, F4, F6] calculates the Racah parameters
         {E0, E1, E2, E3} corresponding to the given Slater integrals.
796   See eqn. 2-80 in Wybourne.
797   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.";
798   FtoE[F0_, F2_, F4_, F6_] := Module[
799     {E0, E1, E2, E3},
800     (
801       E0 = (F0 - 10*F2/Dk[2] - 33*F4/Dk[4] - 286*F6/Dk[6]);
802       E1 = (70*F2/Dk[2] + 231*F4/Dk[4] + 2002*F6/Dk[6])/9;
803       E2 = (F2/Dk[2] - 3*F4/Dk[4] + 7*F6/Dk[6])/9;
```

```
804       E3 = (5*F2/Dk[2] + 6*F4/Dk[4] - 91*F6/Dk[6])/3;
805       Return[{E0, E1, E2, E3}];
806     )
807   ];
808
809   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.";
810   EtoF[E0_, E1_, E2_, E3_] := Module[
811     {F0, F2, F4, F6},
812     (
813       F0 = 1/7       (7 E0 + 9 E1);
814       F2 = 75/14     (E1 + 143 E2 + 11 E3);
815       F4 = 99/7      (E1 - 130 E2 + 4 E3);
816       F6 = 5577/350 (E1 + 35 E2 - 7 E3);
817       Return[{F0, F2, F4, F6}];
818     )
819   ];
820
821   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\".";
822   Options[Electrostatic] = {"Coefficients" -> "Slater"};
823   Electrostatic[{numE_, NKSL_, NKSLp_}, OptionsPattern[]] := Module[
824     {fsub0, fsub2, fsub4, fsub6,
825      esub0, esub1, esub2, esub3,
826      fsup0, fsup2, fsup4, fsup6,
827      eMatrixVal, orbital},
828     (
829       orbital = 3;
830       Which[
831         OptionValue["Coefficients"] == "Slater",
832         (
833           fsub0 = fsubk[numE, orbital, NKSL, NKSLp, 0];
834           fsub2 = fsubk[numE, orbital, NKSL, NKSLp, 2];
835           fsub4 = fsubk[numE, orbital, NKSL, NKSLp, 4];
836           fsub6 = fsubk[numE, orbital, NKSL, NKSLp, 6];
837           eMatrixVal = fsub0*F0 + fsub2*F2 + fsub4*F4 + fsub6*F6;
838         ),
839         OptionValue["Coefficients"] == "Racah",
840         (
841           fsup0 = fsupk[numE, orbital, NKSL, NKSLp, 0];
842           fsup2 = fsupk[numE, orbital, NKSL, NKSLp, 2];
843           fsup4 = fsupk[numE, orbital, NKSL, NKSLp, 4];
844           fsup6 = fsupk[numE, orbital, NKSL, NKSLp, 6];
845           esub0 = fsup0;
846           esub1 = 9/7*fsup0 +   1/42*fsup2 +   1/77*fsup4 +  1/462*
      fsup6;
847           esub2 =             143/42*fsup2 - 130/77*fsup4 + 35/462*
      fsup6;
848           esub3 =              11/42*fsup2 + 4/77*fsup4   -  7/462*
      fsup6;
849           eMatrixVal = esub0*E0 + esub1*E1 + esub2*E2 + esub3*E3;
850         )
851       ];
852       Return[eMatrixVal];
853     )
854   ];
855
856   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.";
857   Options[GenerateElectrostaticTable] = {"Export" -> True, "
        Coefficients" -> "Slater"};
858   GenerateElectrostaticTable[numEmax_Integer:7, OptionsPattern[]] :=
        (
859     ElectrostaticTable = Table[
860       {numE, SL, SpLp} -> SimplifyFun[Electrostatic[{numE, SL, SpLp},
       "Coefficients" -> OptionValue["Coefficients"]]],
```

```
861       {numE, 1, numEmax},
862       {SL, AllowedNKSLTerms[numE]},
863       {SpLp, AllowedNKSLTerms[numE]}
864     ];
865     ElectrostaticTable = Association[Flatten[ElectrostaticTable]];
866     If[OptionValue["Export"],
867       Export[FileNameJoin[{moduleDir, "data", "ElectrostaticTable.m"}],
868       ElectrostaticTable];
869     ];
870     Return[ElectrostaticTable];
871   );
872
873   (* ###################### Electrostatic ###################### *)
874   (* ########################################################## *)
875
876   (* ########################################################## *)
877   (* ########################## Bases ######################### *)
878
879   BasisTableGenerator::usage = "BasisTableGenerator[numE] gives an
        association whose keys are lists of the form {numE, J} and whose
        values are lists with elements of the form {LS, J, MJ}
        representing the elements of the LSJM coupled basis.";
880   BasisTableGenerator[numE_] := Module[
881     {energyStatesTable, allowedJ, J, Jp},
882     (
883       energyStatesTable = <||>;
884       allowedJ = AllowedJ[numE];
885       Do[
886       (
887         energyStatesTable[{numE, J}] = EnergyStates[numE, J];
888       ),
889       {Jp, allowedJ},
890       {J,  allowedJ}];
891       Return[energyStatesTable]
892     )
893   ];
894
895   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}.
896   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.
897   ";
898   Options[BasisLSJMJ] = {"AsAssociation" -> False};
899   BasisLSJMJ[numE_, OptionsPattern[]] := Module[
900     {energyStatesTable, basis, idx1},
901     (
902       energyStatesTable = BasisTableGenerator[numE];
903       basis = Table[
904         energyStatesTable[{numE, AllowedJ[numE][[idx1]]}],
905         {idx1, 1, Length[AllowedJ[numE]]}];
906       basis = Flatten[basis, 1];
907       If[OptionValue["AsAssociation"],
908         (
909           Js    = AllowedJ[numE];
910           basis = Table[(J -> Select[basis, #[[2]] == J &]), {J, Js}];
911           basis = Association[basis];
912         )
913       ];
914       Return[basis];
915     )
916   ];
917
918   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}.
919   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 = (
```

```
980             (parentS + parentL + daughterS + daughterL) -
981             (orbital + spin)
982         );
983         phase = Phaser[-expo];
984         If[parent == 2*orbital,
985             phase = phase * Phaser[(daughterSeniority-1)/2]];
986         Return[phase];
987       )
988     ];
989
990     Options[CFPExpander] = {"Export" -> True, "PhaseFunction" -> "NK"};
991     CFPExpander::usage = "Using the coefficients of fractional
          parentage up to f7 this function calculates them up to f14.
992     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.";
993     CFPExpander[OptionsPattern[]] := Module[
994       {orbital, halfFilled, fullShell, parentMax, PhaseFun,
995       complementaryCFPs, daughter, conjugateDaughter,
996       conjugateParent, parentTerms, daughterTerms,
997       parentCFPs, daughterSeniority, daughterS, daughterL,
998       parentCFP, parentTerm, parentCFPval,
999       parentS, parentL, parentSeniority, phase, prefactor,
1000      newCFPval, key, extendedCFPs, exportFname},
1001      (
1002        orbital    = 3;
1003        halfFilled = 2 * orbital + 1;
1004        fullShell  = 2 * halfFilled;
1005        parentMax  = 2 * orbital;
1006
1007        PhaseFun  = <|
1008            "Judd" -> JuddCFPPhase,
1009            "NK" -> NKCFPPhase|>[OptionValue["PhaseFunction"]];
1010        PrintTemporary["Calculating CFPs using the phase system from ",
           PhaseFun];
1011        (* Initialize everything with lists to be filled in the next Do
          *)
1012        complementaryCFPs =
1013            Table[
1014            ({numE, term} -> {term}),
1015            {numE, halfFilled + 1, fullShell - 1, 1},
1016            {term, AllowedNKSLTerms[numE]
1017            }];
1018        complementaryCFPs = Association[Flatten[complementaryCFPs]];
1019        Do[(
1020            daughter          = parent + 1;
1021            conjugateDaughter = fullShell - parent;
1022            conjugateParent   = conjugateDaughter - 1;
1023            parentTerms       = AllowedNKSLTerms[parent];
1024            daughterTerms     = AllowedNKSLTerms[daughter];
1025            Do[
1026            (
1027                parentCFPs             = Rest[CFP[{daughter,
          daughterTerm}]];
1028                daughterSeniority      = Seniority[daughterTerm];
1029                {daughterS, daughterL} = FindSL[daughterTerm];
1030                Do[
1031                (
1032                    {parentTerm, parentCFPval} = parentCFP;
1033                    {parentS, parentL}         = FindSL[parentTerm];
1034                    parentSeniority            = Seniority[parentTerm];
1035                    phase = PhaseFun[parent, parentS, parentL,
1036                                     daughterS, daughterL,
1037                                     parentSeniority, daughterSeniority
          ];
1038                    prefactor = (daughter * TPO[daughterS, daughterL])
          /
1039                                (conjugateDaughter * TPO[parentS,
          parentL]);
1040                    prefactor = Sqrt[prefactor];
1041                    newCFPval = phase * prefactor * parentCFPval;
1042                    key = {conjugateDaughter, parentTerm};
1043                    complementaryCFPs[key] = Append[complementaryCFPs[
```

```
    key], {daughterTerm, newCFPval}]
1044              ),
1045              {parentCFP, parentCFPs}
1046              ]
1047          ),
1048          {daughterTerm, daughterTerms}
1049          ]
1050          ),
1051      {parent, 1, parentMax}
1052      ];
1053
1054      complementaryCFPs[{14, "1S"}] = {"1S", {"2F",1}};
1055      extendedCFPs          = Join[CFP, complementaryCFPs];
1056      If[OptionValue["Export"];,
1057      (
1058          exportFname = FileNameJoin[{moduleDir, "data", "
    CFPs_extended.m"}];
1059          Print["Exporting to ", exportFname];
1060          Export[exportFname, extendedCFPs];
1061      )
1062      ];
1063      Return[extendedCFPs];
1064  )
1065  ];
1066
1067  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.
1068  The data being parsed here is the file attachment B1F_ALL.TXT which
       comes from Velkov's thesis.";
1069  Options[GenerateCFPTable] = {"Export" -> True};
1070  GenerateCFPTable[OptionsPattern[]] := Module[
1071    {rawText, rawLines, leadChar, configIndex, line, daughter,
1072    lineParts, numberCode, parsedNumber, toAppend, CFPTablefname},
1073    (
1074      CleanWhitespace[string_]     := StringReplace[string,
    RegularExpression["\\s+"]->" "];
1075      AddSpaceBeforeMinus[string_] := StringReplace[string,
    RegularExpression["(?<!\\s)-"]->" -"];
1076      ToIntegerOrString[list_]     := Map[If[StringMatchQ[#,
    NumberString], ToExpression[#], #] &, list];
1077      CFPTable      = ConstantArray[{},7];
1078      CFPTable[[1]] = {{"2F",{"1S",1}}};
1079
1080      (* Cleaning before processing is useful *)
1081      rawText  = Import[FileNameJoin[{moduleDir, "data", "B1F_ALL.TXT
    "}]];
1082      rawLines = StringTrim/@StringSplit[rawText,"\n"];
1083      rawLines = Select[rawLines,#!=""&];
1084      rawLines = CleanWhitespace/@rawLines;
1085      rawLines = AddSpaceBeforeMinus/@rawLines;
1086
1087      Do[(
1088        (* the first character can be used to identify the start of a
     block *)
1089        leadChar=StringTake[line,{1}];
1090        (* ..FN, N is at position 50 in that line *)
1091        If[leadChar=="[",
1092        (
1093          configIndex=ToExpression[StringTake[line,{50}]];
1094          Continue[];
1095        )
1096        ];
1097        (* Identify which daughter term is being listed *)
1098        If[StringContainsQ[line,"[DAUGHTER TERM]"],
1099          daughter=StringSplit[line,"["][[1]];
1100          CFPTable[[configIndex]]=Append[CFPTable[[configIndex]],{
    daughter}];
1101          Continue[];
1102        ];
1103        (* Once we get here we are already parsing a row with
    coefficient data *)
1104        lineParts    = StringSplit[line," "];
1105        parent       = lineParts[[1]];
1106        numberCode   = ToIntegerOrString[lineParts[[3;;]]];
```

```
1107          parsedNumber = SquarePrimeToNormal[numberCode];
1108          toAppend     = {parent,parsedNumber};
1109          CFPTable[[configIndex]][[-1]] = Append[CFPTable[[configIndex
      ]][[-1]], toAppend]
1110        ),
1111        {line,rawLines}];
1112        If[OptionValue["Export"],
1113          (
1114          CFPTablefname = FileNameJoin[{moduleDir, "data", "CFPTable.m"
      }];
1115          Export[CFPTablefname, CFPTable];
1116          )
1117        ];
1118        Return[CFPTable];
1119      )
1120    ];
1121
1122    GenerateCFPAssoc::usage = "GenerateCFPAssoc[] converts the
       coefficients of fractional parentage into an association in which
       zero values are explicit. If the option \"Export\" is set to True,
        the association is exported to the file /data/CFPAssoc.m. This
       function requires that the association CFP be defined.";
1123    Options[GenerateCFPAssoc] = {"Export" -> True};
1124    GenerateCFPAssoc[OptionsPattern[]] := (
1125      CFPAssoc = Association[];
1126      Do[
1127        (daughterTerms = AllowedNKSLTerms[numE];
1128        parentTerms    = AllowedNKSLTerms[numE - 1];
1129        Do[
1130          (
1131          cfps = CFP[{numE, daughter}];
1132          cfps = cfps[[2 ;;]];
1133          parents = First /@ cfps;
1134          Do[
1135            (
1136            key = {numE, daughter, parent};
1137            cfp = If[
1138              MemberQ[parents, parent],
1139              (
1140                idx = Position[parents, parent][[1, 1]];
1141                cfps[[idx]][[2]]
1142              ),
1143              0
1144              ];
1145            CFPAssoc[key] = cfp;
1146            ),
1147            {parent, parentTerms}
1148            ]
1149          ),
1150          {daughter, daughterTerms}
1151          ]
1152        ),
1153        {numE, 1, 14}
1154        ];
1155      If[OptionValue["Export"],
1156        (
1157        CFPAssocfname = FileNameJoin[{moduleDir, "data", "CFPAssoc.m"
      }];
1158        Export[CFPAssocfname, CFPAssoc];
1159        )
1160      ];
1161      Return[CFPAssoc];
1162    );
1163
1164    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.
1165    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.
1166    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];
  )
];

SpinOrbitTable::usage="An association containing the matrix
  elements for the spin-orbit interaction for f^n configurations.
  The keys are lists of the form {n, SL, SpLp, J}.";

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
```

```
    folder. 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];
  )
];

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

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

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]);

  (* Parse table 1 from Judd 1984 *)
  judd1984Fname1 = FileNameJoin[{moduleDir, "data", "Judd1984-1.csv"}];
  data = Import[judd1984Fname1, "CSV", "Numeric" -> False];
  headers = data[[1]];
  data = data[[2 ;;]];
  data = Transpose[data];
  \[Psi] = Select[data[[1]], # != "" &];
  \[Psi]p = Select[data[[2]], # != "" &];
  matrixKeys = Transpose[{\[Psi], \[Psi]p}];
  data = data[[3 ;;]];
  cols = Table[ParseJuddTab1 /@ Select[col, # != "" &], {col, data}];
  cols = Select[cols, Length[#] == 21 &];
  tab1 = Prepend[Prepend[cols, \[Psi]p], \[Psi]];
  tab1 = Transpose[Prepend[Transpose[tab1], headers]];

  (* Parse table 2 from Judd 1984 *)
  judd1984Fname2 = FileNameJoin[{moduleDir, "data", "Judd1984-2.csv"}];
  data = Import[judd1984Fname2, "CSV", "Numeric" -> False];
  headers = data[[1]];
  data = data[[2 ;;]];
  data = Transpose[data];
  {operatorLabels, WUlabels, multiFactorSymbols, multiFactorValues} = data[[;; 4]];
  multiFactorValues = ParseJuddTab1 /@ multiFactorValues;
  multiFactorValues = AssociationThread[multiFactorSymbols ->
```

```
    multiFactorValues];
1297
1298    (*scale values of table 1 given the values in table 2*)
1299    oppyS = {};
1300    normalTable =
1301      Table[header = col[[1]];
1302        If[StringContainsQ[header, " "],
1303          (
1304            multiplierSymbol = StringSplit[header, " "][[1]];
1305            multiplierValue = multiFactorValues[multiplierSymbol];
1306            operatorSymbol = StringSplit[header, " "][[2]];
1307            oppyS = Append[oppyS, operatorSymbol];
1308          ),
1309          (
1310            multiplierValue = 1;
1311            operatorSymbol = header;
1312          )
1313        ];
1314        normalValues = 1/multiplierValue*col[[2 ;;]];
1315        Join[{operatorSymbol}, normalValues], {col, tab1[[3 ;;]]}
1316      ];
1317
1318    (*Create an association for the reduced matrix elements in the f
      ^3 config*)
1319    juddOperators = Association[];
1320    Do[(
1321      col      = normalTable[[colIndex]];
1322      opLabel  = col[[1]];
1323      opValues = col[[2 ;;]];
1324      opMatrix = AssociationThread[matrixKeys -> opValues];
1325      Do[(
1326        opMatrix[Reverse[mKey]] = opMatrix[mKey]
1327        ),
1328      {mKey, matrixKeys}
1329      ];
1330      juddOperators[{3, opLabel}] = opMatrix),
1331      {colIndex, 1, Length[normalTable]}
1332    ];
1333
1334    (* special case of t2 in f3 *)
1335    (* this is the same as getting the reduced matrix elements from
      Judd 1966 *)
1336    numE = 3;
1337    e3Op      = juddOperators[{3, "e_{3}"}];
1338    t2prime   = juddOperators[{3, "t_{2}^{'}"}];
1339    prefactor = 1/(70 Sqrt[2]);
1340    t2Op = (# -> (t2prime[#] + prefactor*e3Op[#])) & /@ Keys[t2prime
      ];
1341    t2Op = Association[t2Op];
1342    juddOperators[{3, "t_{2}"}] = t2Op;
1343
1344    (*Special case of t11 in f3*)
1345    t11 = juddOperators[{3, "t_{11}"}];
1346    eβprimeOp = juddOperators[{3, "e_{\\beta}^{'}"}];
1347    t11primeOp = (# -> (t11[#] + Sqrt[3/385] eβprimeOp[#])) & /@ Keys
      [t11];
1348    t11primeOp = Association[t11primeOp];
1349    juddOperators[{3, "t_{11}^{'}"}] = t11primeOp;
1350    If[OptionValue["Export"],
1351      (
1352        (*export them*)
1353        PrintTemporary["Exporting ..."];
1354        exportFname = FileNameJoin[{moduleDir, "data", "juddOperators
      .m"}];
1355        Export[exportFname, juddOperators];
1356      )
1357    ];
1358    Return[juddOperators];
1359  );
1360
1361  ThreeBodyTable::usage="ThreeBodyTable is an association containing
      the LS-reduced matrix elements for the three-body operators for f^
      n configurations. The keys are lists of the form {n, SL, SpLp}.";
1362
1363  ThreeBodyTables::usage="ThreeBodyTables is an association whose
      keys are integers n from 1 to 7 and whose values are associations
```

```
    whose keys are symbols for the different three-body operators, and
     whose keys are of the form {LS, LpSp} where LS and LpSp are
    strings for LS-terms in f^n.";
1364
1365  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."
    ;
1366  Options[GenerateThreeBodyTables] = {"Export" -> False};
1367  GenerateThreeBodyTables[OptionsPattern[]] := (
1368    tiKeys        = (StringReplace[ToString[#], {"T" -> "t_{", "p" ->
     "}^{'"}] <> "}") & /@ TSymbols;
1369    TSymbolsAssoc = AssociationThread[tiKeys -> TSymbols];
1370    juddOperators = ParseJudd1984[];
1371    (* 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. *)
1372    op3MatrixElement[SL_, SpLp_, opSymbol_] := (
1373      jOP = juddOperators[{3, opSymbol}];
1374      key = {SL, SpLp};
1375      val = If[MemberQ[Keys[jOP], key],
1376        jOP[key],
1377        0];
1378      Return[val];
1379    );
1380    (* 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. *)
1381    Options[ti] = {"Fast" -> True};
1382    ti[nE_, SL_, SpLp_, tiKey_, opOrder_ : 3, OptionsPattern[]] :=
    Module[
1383      {nn, S, L, Sp, Lp,
1384      cfpSL, cfpSpLp,
1385      parentSL, parentSpLp,
1386      tnk, tnks},
1387      (
1388        {S, L}   = FindSL[SL];
1389        {Sp, Lp} = FindSL[SpLp];
1390        fast     = OptionValue["Fast"];
1391        numH     = 14 - nE;
1392        If[fast && Not[MemberQ[{"t_{2}","t_{11}"},tiKey]] && nE > 7,
1393          Return[-tktable[{numH, SL, SpLp, tiKey}]]
1394        ];
1395        If[(S == Sp && L == Lp),
1396        (
1397          cfpSL   = CFP[{nE, SL}];
1398          cfpSpLp = CFP[{nE, SpLp}];
1399          tnks = Table[(
1400              parentSL   = cfpSL[[nn, 1]];
1401              parentSpLp = cfpSpLp[[mm, 1]];
1402              cfpSL[[nn, 2]] * cfpSpLp[[mm, 2]] *
1403              tktable[{nE - 1, parentSL, parentSpLp, tiKey}]
1404              ),
1405              {nn, 2, Length[cfpSL]},
1406              {mm, 2, Length[cfpSpLp]}
1407              ];
1408          tnk = Total[Flatten[tnks]];
1409          ),
1410        tnk = 0;
1411        ];
1412        Return[ nE / (nE - opOrder) * tnk];
1413      )
1414    ];
1415    (* Calculate the reduced matrix elements of t^i for n up to 14 *)
1416    tktable = <||>;
1417    Do[(
1418      Do[(
1419        tkValue = Which[numE <= 2,
1420          (*Initialize n=1,2 with zeros*)
1421          0,
1422          numE == 3,
1423          (* Grab matrix elem in f^3 from Judd 1984 *)
```

```
1424            SimplifyFun[op3MatrixElement[SL, SpLp, opKey]],
1425            True,
1426            SimplifyFun[ti[numE, SL, SpLp, opKey, If[opKey == "e_{3}",
    2, 3]]]
1427            ];
1428          tktable[{numE, SL, SpLp, opKey}] = tkValue;
1429          ),
1430        {SL, AllowedNKSLTerms[numE]},
1431        {SpLp, AllowedNKSLTerms[numE]},
1432        {opKey, Append[tiKeys, "e_{3}"]}
1433        ];
1434        PrintTemporary[StringJoin["\[ScriptF]", ToString[numE], "
    configuration complete"]];
1435        ),
1436        {numE, 1, 14}
1437      ];
1438
1439      (* Now use those reduced matrix elements to determine their sum
    as weighted by their corresponding strengths Ti *)
1440      ThreeBodyTable = <||>;
1441      Do[
1442        Do[
1443        (
1444          ThreeBodyTable[{numE, SL, SpLp}] = (
1445            Sum[(
1446              If[tiKey == "t_{2}", t2Switch, 1] *
1447              tktable[{numE, SL, SpLp, tiKey}] *
1448              TSymbolsAssoc[tiKey] +
1449              If[tiKey == "t_{2}", 1 - t2Switch, 0] *
1450              (-tktable[{14 - numE, SL, SpLp, tiKey}]) *
1451              TSymbolsAssoc[tiKey]
1452              ),
1453            {tiKey, tiKeys}
1454            ]
1455          );
1456        ),
1457        {SL, AllowedNKSLTerms[numE]},
1458        {SpLp, AllowedNKSLTerms[numE]}
1459        ];
1460      PrintTemporary[StringJoin["\[ScriptF]", ToString[numE], " matrix
    complete"]];,
1461      {numE, 1, 7}
1462      ];
1463
1464      ThreeBodyTables = Table[(
1465        terms = AllowedNKSLTerms[numE];
1466        singleThreeBodyTable =
1467          Table[
1468            {SL, SLp} -> ThreeBodyTable[{numE, SL, SLp}],
1469            {SL, terms},
1470            {SLp, terms}
1471          ];
1472        singleThreeBodyTable  = Flatten[singleThreeBodyTable];
1473        singleThreeBodyTables = Table[(
1474            notNullPosition = Position[TSymbols, notNullSymbol][[1,
    1]];
1475            reps = ConstantArray[0, Length[TSymbols]];
1476            reps[[notNullPosition]] = 1;
1477            rep = AssociationThread[TSymbols -> reps];
1478            notNullSymbol -> Association[(singleThreeBodyTable /. rep)]
1479            ),
1480          {notNullSymbol, TSymbols}
1481          ];
1482        singleThreeBodyTables = Association[singleThreeBodyTables];
1483        numE -> singleThreeBodyTables),
1484        {numE, 1, 7}
1485    ];
1486
1487    ThreeBodyTables = Association[ThreeBodyTables];
1488    If[OptionValue["Export"],
1489      (
1490        threeBodyTablefname = FileNameJoin[{moduleDir, "data", "
    ThreeBodyTable.m"}];
1491        Export[threeBodyTablefname, ThreeBodyTable];
1492        threeBodyTablesfname = FileNameJoin[{moduleDir, "data", "
    ThreeBodyTables.m"}];
```

```
      Export[threeBodyTablesfname, ThreeBodyTables];
    )
  ];
Return[{ThreeBodyTable, ThreeBodyTables}];
);


(* ################## 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.
Rigorously 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 done 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};
```

```
1735        {S, L}    = FindSL[SL];
1736        {Sp, Lp} = FindSL[SpLp];
1737        t = 2;
1738        cfpSL     = CFP[{numE, SL}];
1739        cfpSpLp   = CFP[{numE, SpLp}];
1740        Tnkk = Sum[(
1741          parentSL    = cfpSL[[idx2, 1]];
1742          parentSpLp = cfpSpLp[[idx1, 1]];
1743          {Sb, Lb}    = FindSL[parentSL];
1744          {Sbp, Lbp} = FindSL[parentSpLp];
1745          phase = Phaser[Sb + Lb + spin + orbital + Sp + Lp];
1746          (
1747            phase *
1748            cfpSpLp[[idx1, 2]] * cfpSL[[idx2, 2]] *
1749            SixJay[{S, t, Sp}, {Sbp, spin, Sb}] *
1750            SixJay[{L, t, Lp}, {Lbp, orbital, Lb}] *
1751            T22Table[{numE - 1, parentSL, parentSpLp}]
1752          )
1753        ),
1754        {idx1, 2, Length[cfpSpLp]},
1755        {idx2, 2, Length[cfpSL]}
1756        ];
1757        Tnkk *= numE / (numE - 2) * Sqrt[TPO[S,Sp,L,Lp]];
1758        Return[Tnkk];
1759      )
1760    ];
1761
1762    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.
1763    This is an intermediate step to the calculation of the reduced
       matrix elements of the spin-spin operator.";
1764    Options[GenerateT22Table] = {"Export" -> True, "Progress" -> True};
1765    GenerateT22Table[nmax_Integer, OptionsPattern[]] := (
1766      If[And[OptionValue["Progress"], frontEndAvailable],
1767        (
1768          numItersai = Association[Table[numE->Length[AllowedNKSLTerms[
       numE]]^2, {numE, 1, nmax}]];
1769          counters = Association[Table[numE->0, {numE, 1, nmax}]];
1770          totalIters = Total[Values[numItersai[[1;;nmax]]]];
1771          template1 = StringTemplate["Iteration `numiter` of `totaliter
       `"];
1772          template2 = StringTemplate["`remtime` min remaining"];
       template3 = StringTemplate["Iteration speed = `speed` ms/it"];
1773          template4 = StringTemplate["Time elapsed = `runtime` min"];
1774          progBar = PrintTemporary[
1775            Dynamic[
1776              Pane[
1777                Grid[{{Superscript["f", numE]},
1778                      {template1[<|"numiter"->numiter, "totaliter"->
       totalIters|>]},
1779                      {template4[<|"runtime"->Round[QuantityMagnitude[
       UnitConvert[(Now-startTime), "min"]], 0.1]|>]},
1780                      {template2[<|"remtime"->Round[QuantityMagnitude[
       UnitConvert[(Now-startTime)/(numiter)*(totalIters-numiter), "min"
       ]], 0.1]|>]},
1781                      {template3[<|"speed"->Round[QuantityMagnitude[Now
       -startTime, "ms"]/(numiter), 0.01]|>]},
1782                      {ProgressIndicator[Dynamic[numiter], {1,
       totalIters}]}},
1783                      Frame->All],
1784                      Full,
1785                      Alignment->Center]
1786                  ]
1787                ];
1788        )
1789      ];
1790      T22Table = <||>;
1791      startTime = Now;
1792      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];
  );

  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]
    )
  ];

  GenerateSpinSpinTable::usage = "GenerateSpinSpinTable[nmax] generates the reduced matrix elements in the |LSJ> basis for the spin-spin 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 LoadChenDeltas[].";
  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 ################## *)
  (* ############################################################ *)
```

```
1963
1964  (* ############################################################ *)
1965  (* ##################### Free-Ion Energies ##################### *)
1966
1967  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.";
1968  Options[GenerateFreeIonTable] = {"Export" -> False};
1969  GenerateFreeIonTable[OptionsPattern[]] := Module[
1970    {terms, numEH, zetaSign, fname, FreeIonTable},
1971    (
1972      If[Not[ValueQ[ElectrostaticTable]],
1973        LoadElectrostatic[]
1974        ];
1975      If[Not[ValueQ[SpinOrbitTable]],
1976        LoadSpinOrbit[]
1977      ];
1978      If[Not[ValueQ[ReducedUkTable]],
1979        LoadUk[]
1980      ];
1981      FreeIonTable = <||>;
1982      Do[
1983        (
1984          terms = AllowedNKSLJTerms[nE];
1985          numEH = Min[nE, 14 - nE];
1986          zetaSign = If[nE > 7, -1, 1];
1987          Do[
1988            FreeIonTable[{nE,term[[1]],term[[2]]}] = (
1989              Electrostatic[{numEH, term[[1]], term[[1]]}] +
1990                zetaSign * SpinOrbitTable[{numEH, term[[1]], term
      [[1]], term[[2]]}]
1991                ),
1992            {term, terms}];
1993        ),
1994        {nE, 1, 14}
1995      ];
1996      If[OptionValue["Export"],
1997        (
1998          fname = FileNameJoin[{moduleDir, "data", "FreeIonTable.m"
      }];
1999          Export[fname, FreeIonTable];
2000        )
2001      ];
2002      Return[FreeIonTable];
2003    )
2004  ];
2005
2006  LoadFreeIon::usage = "LoadFreeIon[] loads the free-ion energies
        from the data folder. The values are stored in the association
        FreeIonTable.";
2007  LoadFreeIon[] := (
2008    If[ValueQ[FreeIonTable],
2009      Return[]
2010    ];
2011    PrintTemporary["Loading the association of free-ion energies ..."
      ];
2012    FreeIonTableFname = FileNameJoin[{moduleDir, "data", "
      FreeIonTable.m"}];
2013    FreeIonTable = If[!FileExistsQ[FreeIonTableFname],
2014      (
2015        PrintTemporary[">> FreeIonTable.m not found, generating ..."
      ];
2016        GenerateFreeIonTable["Export" -> True]
2017      ),
```

```
        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]
  calculates the matrix element of the crystal field in terms of Bqk
   and Sqk parameters for configuration f^numE. It is calculated as
  an association with keys of the form {n, NKSL, J, M, NKSLp, Jp, Mp
  }.";
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}
  ]
```

```
2088  )
2089
2090  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.";
2091  TotalCFIters[i_, j_] := (
2092    numIters = {196, 8281, 132496, 1002001, 4008004, 9018009,
        11778624};
2093    Return[Total[numIters[[i ;; j]]]];
2094    )
2095
2096  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.";
2097  Options[GenerateCrystalFieldTable] = {"Export" -> False, "Progress"
       -> True, "Compress" -> True, "Overwrite" -> False};
2098  GenerateCrystalFieldTable[numEs_List:{1,2,3,4,5,6,7},
        OptionsPattern[]] := (
2099    ExportFun =
2100    If[OptionValue["Compress"],
2101      ExportMZip,
2102      Export
2103    ];
2104    numiter = 1;
2105    template1 = StringTemplate["Iteration `numiter` of `totaliter`"];
2106    template2 = StringTemplate["`remtime` min remaining"];
2107    template3 = StringTemplate["Iteration speed = `speed` ms/it"];
2108    template4 = StringTemplate["Time elapsed = `runtime` min"];
2109    totalIter = Total[TotalCFIters[#, #] & /@ numEs];
2110    freebies = 0;
2111    startTime = Now;
2112    If[And[OptionValue["Progress"], frontEndAvailable],
2113      progBar = PrintTemporary[
2114        Dynamic[
2115          Pane[
2116            Grid[
2117              {
2118                {Superscript["f", numE]},
2119                {template1[<|"numiter" -> numiter, "totaliter" ->
      totalIter|>]},
2120                {template4[<|"runtime" -> Round[QuantityMagnitude[
      UnitConvert[(Now - startTime), "min"]], 0.1]|>]},
2121                {template2[<|"remtime" -> Round[QuantityMagnitude[
      UnitConvert[(Now - startTime)/(numiter - freebies) * (totalIter -
      numiter), "min"]], 0.1]|>]},
2122                {template3[<|"speed" -> Round[QuantityMagnitude[Now -
       startTime, "ms"]/(numiter-freebies), 0.01]|>]},
2123                {ProgressIndicator[Dynamic[numiter], {1, totalIter}]}
2124              },
2125            Frame -> All
2126            ],
2127          Full,
2128          Alignment -> Center
2129          ]
2130        ]
2131      ];
2132    ];
2133    Do[
2134      (
2135        exportFname = FileNameJoin[{moduleDir, "data", "
      CrystalFieldTable_f"<>ToString[numE]<>".m"}];
2136        If[And[FileExistsQ[exportFname],Not[OptionValue["Overwrite"
      ]]],
2137          Print["File exists, skipping ..."];
2138          numiter+=  TotalCFIters[numE, numE];
2139          freebies+= TotalCFIters[numE, numE];
2140          Continue[];
2141        ];
```

```
        CrystalFieldTable = <||>;
        Do[
          (
            numiter+=1;
            {S,L}   = FindSL[NKSL];
            {Sp,Lp} = FindSL[NKSLp];
            CrystalFieldTable[{numE,NKSL,J,M,NKSLp,Jp,Mp}] =
              Which[
                Abs[M-Mp]>6,
                0,
                Abs[S-Sp]!=0,
                0,
                True,
                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}
    ]
  )

  ParseBenelli2015::usage="ParseBenelli2015[] parses the data from
    file /data/benelli_and_gatteschi_table3p3.csv which corresponds to
     Table 3.3 of Benelli and Gatteshi, Introduction to Molecular.
    This data provides the form that the crystal field has under
    different point group symmetries. This function parses that data
    into an association with keys equal to strings representing any of
     the 32 crystallographic point groups.";
  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 describe 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 ###### *)
```

```
2270
2271  CasimirSO3::usage = "CasimirSO3[{SL, SpLp}] returns LS reduced
        matrix element of the configuration interaction term corresponding
         to the Casimir operator of R3.";
2272  CasimirSO3[{SL_, SpLp_}] := (
2273    {S, L} = FindSL[SL];
2274    If[SL == SpLp,
2275      α * L * (L + 1),
2276      0
2277    ]
2278  )
2279
2280  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.
2281  Reference: Wybourne, \"Spectroscopic Properties of Rare Earths\",
        table 2-6.";
2282  GG2U = Association[{
2283      "00" -> 0,
2284      "10" -> 6/12 ,
2285      "11" -> 12/12,
2286      "20" -> 14/12,
2287      "21" -> 21/12,
2288      "22" -> 30/12,
2289      "30" -> 24/12,
2290      "31" -> 32/12,
2291      "40" -> 36/12}
2292    ];
2293
2294  CasimirG2::usage = "CasimirG2[{SL, SpLp}] returns LS reduced matrix
         element of the configuration interaction term corresponding to
        the Casimir operator of G2.";
2295  CasimirG2[{SL_, SpLp_}] := (
2296    Ulabel = FindNKLSTerm[SL][[1]][[4]];
2297    If[SL==SpLp,
2298      β * GG2U[Ulabel],
2299      0
2300    ]
2301  )
2302
2303  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.
2304  Reference: Wybourne, \"Spectroscopic Properties of Rare Earths\",
        table 2-7.";
2305  GSO7W := Association[
2306    {
2307      "000" -> 0,
2308      "100" -> 3/5,
2309      "110" -> 5/5,
2310      "111" -> 6/5,
2311      "200" -> 7/5,
2312      "210" -> 9/5,
2313      "211" -> 10/5,
2314      "220" -> 12/5,
2315      "221" -> 13/5,
2316      "222" -> 15/5
2317    }
2318  ];
2319
2320  CasimirSO7::usage = "CasimirSO7[{SL, SpLp}] returns the LS reduced
        matrix element of the configuration interaction term corresponding
         to the Casimir operator of R7.";
2321  CasimirSO7[{SL_, SpLp_}] := (
2322    Wlabel = FindNKLSTerm[SL][[1]][[3]];
2323    If[SL==SpLp,
2324      γ * GSO7W[Wlabel],
2325      0
2326    ]
2327  )
2328
2329  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];
  )
];

(* #### 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]
  )
];
```

```
2397
2398  EnergyStates::usage = "Alias for AllowedNKSLJMforJTerms. At some
        point may be used to redefine states used in basis.";
2399  EnergyStates[numE_, J_]:= AllowedNKSLJMforJTerms[numE, J];
2400
2401  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.";
2402  Options[JJBlockMatrixFileName] = {"FilenameAppendix" -> ""};
2403  JJBlockMatrixFileName[numE_Integer, OptionsPattern[]] := (
2404    fileApp = OptionValue["FilenameAppendix"];
2405    fname = FileNameJoin[{moduleDir,
2406        "hams",
2407        StringJoin[{"f", ToString[numE], "_JJBlockMatrixTable",
        fileApp ,".m"}]}];
2408    Return[fname];
2409  );
2410
2411  TabulateJJBlockMatrixTable::usage = "TabulateJJBlockMatrixTable[
        numE, CFTable] returns an association JJBlockMatrixTable with keys
         equal to lists of the form {numE, J, Jp} and values equal to JJ
        blocks of the semi-empirical Hamiltonian. The function admits an
        optional parameter \"Sparse\" which can be used to control whether
         the matrix is returned as a SparseArray or not. The default is
        True. Another admitted option is \"ChenDeltas\" which 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
        LoadChenDeltas[].";
2412  Options[TabulateJJBlockMatrixTable] = {"Sparse"->True, "ChenDeltas"
        ->False};
2413  TabulateJJBlockMatrixTable[numE_, CFTable_, OptionsPattern[]] := (
2414    JJBlockMatrixTable = <||>;
2415    totalIterations = Length[AllowedJ[numE]]^2;
2416    template1 = StringTemplate["Iteration `numiter` of `totaliter`"];
2417    template2 = StringTemplate["`remtime` min remaining"];
2418    template4 = StringTemplate["Time elapsed = `runtime` min"];
2419    numiter   = 0;
2420    startTime = Now;
2421    If[$FrontEnd =!= Null,
2422      (
2423        temp = PrintTemporary[
2424          Dynamic[
2425            Grid[
2426              {
2427                {template1[<|"numiter"->numiter, "totaliter"->
        totalIterations|>]},
2428                {template2[<|"remtime"->Round[QuantityMagnitude[
        UnitConvert[(Now-startTime)/(Max[1,numiter])*(totalIterations-
        numiter), "min"]], 0.1]|>]},
2429                {template4[<|"runtime"->Round[QuantityMagnitude[
        UnitConvert[(Now-startTime), "min"]], 0.1]|>]},
2430                {ProgressIndicator[numiter, {1, totalIterations}]}
2431              }
2432            ]
2433        ]
2434        ];
2435      )
2436    ];
2437    Do[
2438      (
2439        JJBlockMatrixTable[{numE, J, Jp}] = JJBlockMatrix[numE, J, Jp
        , CFTable, "Sparse"->OptionValue["Sparse"], "ChenDeltas" ->
        OptionValue["ChenDeltas"]];
2440        numiter += 1;
2441      ),
2442    {Jp, AllowedJ[numE]},
2443    {J, AllowedJ[numE]}
2444    ];
2445    If[$FrontEnd =!= Null,
2446      NotebookDelete[temp]
2447    ];
2448    Return[JJBlockMatrixTable];
2449  );
2450
```

```
2451  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'|>.";
2452  Options[TabulateManyJJBlockMatrixTables] = {"Overwrite"->False, "
       Sparse"->True, "ChenDeltas"->False, "FilenameAppendix"-> "", "
       Compressed" -> False};
2453  TabulateManyJJBlockMatrixTables[ns_, OptionsPattern[]] := (
2454    overwrite = OptionValue["Overwrite"];
2455    fNames = <||>;
2456    fileApp = OptionValue["FilenameAppendix"];
2457    ExportFun = If[OptionValue["Compressed"], ExportMZip, Export];
2458    Do[
2459      (
2460        CFdataFilename = FileNameJoin[{moduleDir, "data", "
       CrystalFieldTable_f"<>ToString[numE]<>".zip"}];
2461        PrintTemporary["Importing CrystalFieldTable from ",
       CFdataFilename, " ..."];
2462        CrystalFieldTable = ImportMZip[CFdataFilename];
2463
2464        PrintTemporary["#------- numE = ", numE, " -------#"];
2465        exportFname = JJBlockMatrixFileName[numE, "FilenameAppendix"
       -> fileApp];
2466        fNames[numE] = exportFname;
2467        If[FileExistsQ[exportFname] && Not[overwrite],
2468          Continue[]
2469        ];
2470        JJBlockMatrixTable = TabulateJJBlockMatrixTable[numE,
       CrystalFieldTable, "Sparse"->OptionValue["Sparse"], "ChenDeltas"
       -> OptionValue["ChenDeltas"]];
2471        If[FileExistsQ[exportFname] && overwrite,
2472          DeleteFile[exportFname]
2473        ];
2474        ExportFun[exportFname, JJBlockMatrixTable];
2475
2476        ClearAll[CrystalFieldTable];
2477      ),
2478    {numE, ns}
2479    ];
2480    Return[fNames];
2481  );
2482
2483  HamMatrixAssembly::usage = "HamMatrixAssembly[numE] returns the
       Hamiltonian matrix for the f^numE configuration. The matrix is
       returned as a SparseArray.
2484  The function admits an optional parameter \"FilenameAppendix\"
       which can be used to control which variant of the JJBlocks is used
        to assemble the matrix.
2485  It also admits an optional parameter \"IncludeZeeman\" which can be
        used to include the Zeeman interaction. The default is False.
2486  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.
2487  The option \"ReturnInBlocks\" can be used to return the matrix in
       block or flattened form. The default is to return it in flattened
       form.";
2488  Options[HamMatrixAssembly] = {
2489        "FilenameAppendix" -> "",
2490        "IncludeZeeman"    -> False,
2491        "Set t2Switch"     -> True,
2492        "ReturnInBlocks"   -> False,
2493        "OperatorBasis"    -> "Legacy"};
2494  HamMatrixAssembly[nf_, OptionsPattern[]] := Module[
2495    {numE, ii, jj, howManyJs, Js, blockHam, opBasis},
2496    (
2497      (*####################################*)
2498      ImportFun = ImportMZip;
2499      opBasis = OptionValue["OperatorBasis"];
2500      If[Not[MemberQ[{"Legacy", "MostlyOrthogonal", "Orthogonal"},
       opBasis]],
2501        Print["Operator basis ", opBasis, " not recognized, using \"
```

```
    Legacy\" basis."];
2502        opBasis = "Legacy";
2503      ];
2504      If[opBasis == "Orthogonal",
2505        Print["Operator basis \"Orthogonal\" not implemented yet,
    aborting ..."];
2506        Return[Null];
2507      ];
2508      (*************************************)
2509      If[opBasis == "MostlyOrthogonal",
2510        (
2511          blockHam = HamMatrixAssembly[nf,
2512          "OperatorBasis" -> "Legacy",
2513          "FilenameAppendix" -> OptionValue["FilenameAppendix"],
2514          "IncludeZeeman" -> OptionValue["IncludeZeeman"],
2515          "Set t2Switch" -> OptionValue["Set t2Switch"],
2516          "ReturnInBlocks" -> OptionValue["ReturnInBlocks"]];
2517          paramChanger = Which[
2518            nf < 7,
2519            <|
2520              F0 -> 1/91 (54 E1p+91 E0p+78 γp),
2521              F2 -> (15/392 *
2522                (
2523                  140   E1p  +
2524                  20020 E2p +
2525                  1540  E3p  +
2526                  770 αp -
2527                  70 γp  +
2528                  22 Sqrt[2] T2p -
2529                  11 Sqrt[2] nf T2p t2Switch -
2530                  11 Sqrt[2] (14-nf) T2p (1 - t2Switch)
2531                )
2532              ),
2533              F4 -> (99/490 *
2534                (
2535                  70   E1p -
2536                  9100 E2p +
2537                  280  E3p +
2538                  140 αp -
2539                  35 γp  +
2540                  4 Sqrt[2] T2p -
2541                  2 Sqrt[2] nf T2p t2Switch -
2542                  2 Sqrt[2] (14-nf) T2p (1-t2Switch)
2543                )
2544                ),
2545              F6 -> (5577/7000 *
2546                (
2547                  20  E1p +
2548                  700 E2p -
2549                  140 E3p -
2550                  70 αp -
2551                  10 γp -
2552                  2 Sqrt[2] T2p +
2553                  Sqrt[2] nf T2p t2Switch +
2554                  Sqrt[2] (14-nf) T2p (1-t2Switch)
2555                )
2556                ),
2557              ζ  -> ζ,
2558              α -> (5 αp)/4,
2559              β  -> -6  (5 αp + βp),
2560              γ -> 5/2 (2 βp + 5 γp),
2561              T2 -> 0
2562            |>,
2563          nf >= 7,
2564            <|
2565              F0 -> 1/91 (54 E1p+91 E0p+78 γp),
2566              F2 -> (15/392 *
2567                (
2568                  140   E1p  +
2569                  20020 E2p +
2570                  1540  E3p  +
2571                  770 αp -
2572                  70 γp  +
2573                  22 Sqrt[2] T2p -
2574                  11 Sqrt[2] nf T2p
2575                )
```

```
2576              ),
2577              F4 -> (99/490 *
2578                (
2579                  70   E1p -
2580                  9100 E2p +
2581                  280  E3p +
2582                  140 αp -
2583                  35 γp  +
2584                  4 Sqrt[2] T2p -
2585                  2 Sqrt[2] nf T2p
2586                )
2587                ),
2588              F6 -> (5577/7000 *
2589                (
2590                  20  E1p +
2591                  700 E2p -
2592                  140 E3p -
2593                  70 αp -
2594                  10 γp -
2595                  2 Sqrt[2] T2p +
2596                  Sqrt[2] nf T2p
2597                )
2598                ),
2599              ζ  -> ζ,
2600              α -> (5 αp)/4,
2601              β  -> -6  (5 αp + βp),
2602              γ -> 5/2 (2 βp + 5 γp),
2603              T2 -> 0
2604            |>
2605          ];
2606          blockHamM0 = Which[
2607            OptionValue["ReturnInBlocks"] == False,
2608            ReplaceInSparseArray[blockHam, paramChanger],
2609            OptionValue["ReturnInBlocks"] == True,
2610            Map[ReplaceInSparseArray[#, paramChanger]&, blockHam,
    {2}]
2611          ];
2612          Return[blockHamM0];
2613        )
2614      ];
2615      (*######################################*)
2616
2617      (*hole-particle equivalence enforcement*)
2618      numE = nf;
2619      allVars = {E0, E1, E2, E3, ζ, F0, F2, F4, F6, M0, M2, M4, T2,
    T2p,
2620        T3, T4, T6, T7, T8, P0, P2, P4, P6, gs,
2621        α, β, γ, B02, B04, B06, B12, B14, B16,
2622        B22, B24, B26, B34, B36, B44, B46, B56, B66, S12, S14, S16,
    S22,
2623        S24, S26, S34, S36, S44, S46, S56, S66, T11p, T12, T14, T15,
    T16,
2624        T17, T18, T19, Bx, By, Bz};
2625      params0 = AssociationThread[allVars, allVars];
2626      If[nf > 7,
2627        (
2628          numE = 14 - nf;
2629          params = HoleElectronConjugation[params0];
2630          If[OptionValue["Set t2Switch"], params[t2Switch] = 0];
2631        ),
2632        params = params0;
2633        If[OptionValue["Set t2Switch"], params[t2Switch] = 1];
2634      ];
2635      (* Load symbolic expressions for LS,J,J' energy sub-matrices.
    *)
2636      emFname = JJBlockMatrixFileName[numE, "FilenameAppendix" ->
    OptionValue["FilenameAppendix"]];
2637      JJBlockMatrixTable = ImportFun[emFname];
2638      (*Patch together the entire matrix representation using J,J'
    blocks.*)
2639      PrintTemporary["Patching JJ blocks ..."];
2640      Js        = AllowedJ[numE];
2641      howManyJs = Length[Js];
2642      blockHam  = ConstantArray[0, {howManyJs, howManyJs}];
2643      Do[
2644        blockHam[[jj, ii]] = JJBlockMatrixTable[{numE, Js[[ii]], Js[[
```

```
    jj]]}];,
2645      {ii, 1, howManyJs},
2646      {jj, 1, howManyJs}
2647      ];
2648      (* Once the block form is created flatten it *)
2649      If[Not[OptionValue["ReturnInBlocks"]],
2650        (blockHam = ArrayFlatten[blockHam];
2651        blockHam  = ReplaceInSparseArray[blockHam, params];
2652        ),
2653        (blockHam  = Map[ReplaceInSparseArray[#, params]&,blockHam
    ,{2}];)
2654      ];
2655
2656      If[OptionValue["IncludeZeeman"],
2657        (
2658          PrintTemporary["Including Zeeman terms ..."];
2659          {magx, magy, magz} = MagDipoleMatrixAssembly[numE, "
    ReturnInBlocks" -> OptionValue["ReturnInBlocks"]];
2660          blockHam += - teslaToKayser * (Bx * magx + By * magy + Bz *
     magz);
2661        )
2662      ];
2663      Return[blockHam];
2664    )
2665  ];
2666
2667  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.
2668  If the option \"Export\" is set to True, then the function also
      exports the resulting sparse array to the ./hams/ folder.
2669  The option \"PrependToFilename\" can be used to prepend a string to
       the filename to which the function may export to.
2670  The option \"Return\" can be used to choose whether the function
      returns the matrix or not.
2671  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.
2672  The option \"IncludeZeeman\" can be used to toggle the inclusion of
       the Zeeman interaction with an external magnetic field.
2673  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.";
2674  Options[SimplerSymbolicHamMatrix] = {
2675    "Export"->True,
2676    "PrependToFilename"->"",
2677    "EorF"->"F",
2678    "Overwrite" -> False,
2679    "Return" -> True,
2680    "Set t2Switch" -> False,
2681    "IncludeZeeman" -> False,
2682    "OperatorBasis" -> "Legacy"
2683    };
2684  SimplerSymbolicHamMatrix[numE_, simplifier_, OptionsPattern[]] :=
      Module[
2685    {thisHam,fname, fnamemx},
2686    (
2687      If[Not[ValueQ[ElectrostaticTable]],
2688        LoadElectrostatic[]
2689        ];
2690      If[Not[ValueQ[SOOandECSOTable]],
2691        LoadSOOandECSO[]
2692      ];
2693      If[Not[ValueQ[SpinOrbitTable]],
2694        LoadSpinOrbit[]
2695      ];
```

```
2696      If[Not[ValueQ[SpinSpinTable]],
2697        LoadSpinSpin[]
2698      ];
2699      If[Not[ValueQ[ThreeBodyTable]],
2700        LoadThreeBody[]
2701      ];
2702
2703      opBasis = OptionValue["OperatorBasis"];
2704      If[Not[MemberQ[{"Legacy", "MostlyOrthogonal", "Orthogonal"},
    opBasis]],
2705        Print["Operator basis ", opBasis, " not recognized, using \"
    Legacy\" basis."];
2706        opBasis = "Legacy";
2707      ];
2708      If[opBasis == "Orthogonal",
2709        Print["Operator basis \"Orthogonal\" not implemented yet,
    aborting ..."];
2710        Return[Null];
2711      ];
2712
2713      fnamePrefix = Which[
2714        opBasis == "Legacy",
2715        "SymbolicMatrix-f",
2716        opBasis == "MostlyOrthogonal",
2717        "SymbolicMatrix-mostly-orthogonal-f",
2718        opBasis == "Orthogonal",
2719        "SymbolicMatrix-orthogonal-f"
2720      ];
2721      fname   = FileNameJoin[{moduleDir, "hams",
2722        OptionValue["PrependToFilename"] <> fnamePrefix <> ToString[
    numE] <> ".m"}];
2723      fnamemx = FileNameJoin[{moduleDir, "hams",
2724        OptionValue["PrependToFilename"] <> fnamePrefix <> ToString[
    numE] <> ".mx"}];
2725      fnamezip = FileNameJoin[{moduleDir, "hams",
2726        OptionValue["PrependToFilename"] <> fnamePrefix <> ToString[
    numE] <> ".zip"}];
2727      If[Or[FileExistsQ[fname],
2728            FileExistsQ[fnamemx],
2729            FileExistsQ[fnamezip]]
2730        && Not[OptionValue["Overwrite"]],
2731        (
2732          If[OptionValue["Return"],
2733            (
2734              Which[
2735                FileExistsQ[fnamezip],
2736                (
2737                  Print["File ", fnamezip, " already exists, and
    option \"Overwrite\" is set to False, loading file ..."];
2738                  thisHam = ImportMZip[fnamezip];
2739                  Return[thisHam];
2740                ),
2741                FileExistsQ[fnamemx],
2742                (
2743                  Print["File ", fnamemx, " already exists, and
    option \"Overwrite\" is set to False, loading file ..."];
2744                  thisHam = Import[fnamemx];
2745                  Return[thisHam];
2746                ),
2747                FileExistsQ[fname],
2748                (
2749                  Print["File ", fname, " already exists, and option
    \"Overwrite\" is set to False, loading file ..."];
2750                  thisHam = Import[fname];
2751                  Print["Exporting to file ", fnamemx, " for quicker
    loading."];
2752                  Export[fnamemx, thisHam];
2753                  Return[thisHam];
2754                )
2755              ]
2756            ),
2757            (
2758              Print["File ", fname, " already exists, skipping ..."];
2759              Return[Null];
2760            )
2761          ]
```

```
2762          )
2763        ];
2764
2765        thisHam = HamMatrixAssembly[numE, "Set t2Switch" -> OptionValue
      ["Set t2Switch"], "IncludeZeeman" -> OptionValue["IncludeZeeman"],
       "OperatorBasis" -> opBasis];
2766        If[Length[simplifier] > 0,
2767          thisHam = ReplaceInSparseArray[thisHam, simplifier];
2768        ];
2769        (* This removes zero entries from being included in the sparse
      array *)
2770        thisHam = SparseArray[thisHam];
2771        If[OptionValue["Export"],
2772        (
2773          If[Not@FileExistsQ[fname],
2774          (
2775            Print["Exporting to file ", fname];
2776            Export[fname,   thisHam];
2777          )
2778          ];
2779          If[Not@FileExistsQ[fnamemx],
2780          (
2781            Print["Exporting to file ", fnamemx];
2782            Export[fnamemx, thisHam];
2783          )
2784          ];
2785        )
2786        ];
2787        If[OptionValue["Return"],
2788          Return[thisHam],
2789          Return[Null]
2790        ];
2791      )
2792    ];
2793
2794    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.";
2795    ScalarLSJMFromLS[numE_, LSReducedMatrixElements_] := Module[
2796      {jjBlocktable, NKSLJMs, NKSLJMps, J, Jp, eMatrix, SLterm,
      SpLpterm,
2797      MJ, MJp, subKron, matValue, Js, howManyJs, blockHam, ii, jj},
2798      (
2799      SparseDiagonalArray[diagonalElements_] := SparseArray[
2800        Table[{i, i} -> diagonalElements[[i]],
2801          {i, 1, Length[diagonalElements]}]
2802        ];
2803      SparseZeroArray[width_, height_] := (
2804        SparseArray[
2805          Join[
2806          Table[{1, i} -> 0, {i, 1, width}],
2807          Table[{i, 1} -> 0, {i, 1, height}]
2808          ]
2809          ]
2810        );
2811      jjBlockTable = <||>;
2812      Do[
2813        NKSLJMs  = AllowedNKSLJMforJTerms[numE, J];
2814        NKSLJMps = AllowedNKSLJMforJTerms[numE, Jp];
2815        If[J != Jp,
2816          jjBlockTable[{numE, J, Jp}] = SparseZeroArray[Length[NKSLJMs
      ], Length[NKSLJMps]];
2817        Continue[];
2818        ];
2819        eMatrix = Table[
2820          (* Condition for a scalar matrix op *)
2821          SLterm = NKSLJM[[1]];
2822          SpLpterm = NKSLJMp[[1]];
2823          MJ = NKSLJM[[3]];
2824          MJp = NKSLJMp[[3]];
2825          subKron = (KroneckerDelta[MJ, MJp]);
2826          matValue = If[subKron == 0,
2827          0,
2828            (
```

```
            Which[MemberQ[Keys[LSReducedMatrixElements], {numE, SLterm, SpLpterm}],
              LSReducedMatrixElements[{numE, SLterm, SpLpterm}],
              MemberQ[Keys[LSReducedMatrixElements], {numE, SpLpterm, SLterm}],
              LSReducedMatrixElements[{numE, SpLpterm, SLterm}],
              True,
              0
            ]
          )
        ];
        matValue,
        {NKSLJMp, NKSLJMps},
        {NKSLJM, NKSLJMs}
        ];
      jjBlockTable[{numE, J, Jp}] = SparseArray[eMatrix],
      {J, AllowedJ[numE]},
      {Jp, AllowedJ[numE]}
      ];

    Js = AllowedJ[numE];
    howManyJs = Length[Js];
    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[]];
```

```
2950      If[Not[ValueQ[SOOandECSOTable]],LoadSOOandECSO[]];
2951      If[Not[ValueQ[SpinOrbitTable]],LoadSpinOrbit[]];
2952      If[Not[ValueQ[SpinSpinTable]],LoadSpinSpin[]];
2953      If[Not[ValueQ[ThreeBodyTable]],LoadThreeBody[]];
2954      fname   = FileNameJoin[{moduleDir,"hams",OptionValue["
          PrependToFilename"]<>"Level-SymbolicMatrix-f"<>ToString[numE]<>"-"
          <>ToString[hash]<>".m"}];
2955      fnamemx = FileNameJoin[{moduleDir,"hams",OptionValue["
          PrependToFilename"]<>"Level-SymbolicMatrix-f"<>ToString[numE]<>"-"
          <>ToString[hash]<>".mx"}];
2956      If[Or[FileExistsQ[fname],FileExistsQ[fnamemx]]&&Not[OptionValue
          ["Overwrite"]],
2957        (
2958          If[OptionValue["Return"],
2959          (
2960            Which[FileExistsQ[fnamemx],
2961            (
2962              Print["File ",fnamemx," already exists, and option \"
                  Overwrite\" is set to False, loading file ..."];
2963              thisHamAssoc=Import[fnamemx];
2964              Return[thisHamAssoc];
2965            ),
2966            FileExistsQ[fname],
2967            (
2968              Print["File ",fname," already exists, and option \"
                  Overwrite\" is set to False, loading file ..."];
2969              thisHamAssoc=Import[fname];
2970              Print["Exporting to file ",fnamemx," for quicker loading.
                  "];
2971              Export[fnamemx,thisHamAssoc];
2972              Return[thisHamAssoc];
2973            )
2974            ]
2975          ),
2976          (
2977            Print["File ",fname," already exists, skipping ..."];
2978            Return[Null];
2979          )
2980          ]
2981        )
2982        ];
2983        Js           = AllowedJ[numE];
2984        thisHamAssoc = HamMatrixAssembly[numE,
2985          "Set t2Switch"->True,
2986          "IncludeZeeman"->False,
2987          "ReturnInBlocks"->True
2988        ];
2989        thisHamAssoc = Diagonal[thisHamAssoc];
2990        thisHamAssoc = Map[SparseArray[ReplaceInSparseArray[#,
          simplifier]]&,thisHamAssoc,{1}];
2991        thisHamAssoc = FreeHam[thisHamAssoc,numE];
2992        thisHamAssoc = AssociationThread[Js->thisHamAssoc];
2993        If[OptionValue["Export"],
2994          (
2995            Print["Exporting to file ",fname," and to ",fnamemx];
2996            Export[fname,thisHamAssoc];
2997            Export[fnamemx,thisHamAssoc];
2998          )
2999        ];
3000        If[OptionValue["Return"],
3001          Return[thisHamAssoc],
3002          Return[Null]
3003        ];
3004      )
3005    ];
3006
3007    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.
3008    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
        subspace, 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.
3009  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.
3010  The following options are admitted:
3011    - \"Overwrite Hamiltonian\", if set to True the function will
    overwrite the symbolic Hamiltonian. Default is False.
3012    - \"Return as states\", see description above. Default is True.
3013    - \"Simplifier\", this is a list with symbols that are set to
    zero for defining the parameters kept in the level  description.
3014  ";
3015  Options[LevelSolver] = {
3016    "Overwrite Hamiltonian" -> False,
3017    "Return as states" -> True,
3018    "Simplifier" -> Join[
3019      cfSymbols,
3020      TSymbols,
3021      casimirSymbols,
3022      pseudoMagneticSymbols,
3023      marvinSymbols,
3024      DeleteCases[magneticSymbols,ζ]
3025    ],
3026    "PrintFun" -> PrintTemporary
3027  };
3028  LevelSolver[numE_Integer, params0_Association, OptionsPattern[]] :=
     Module[
3029    {ln, simplifier, simpleHam, basis,
3030    numHam, eigensys, startTime, endTime,
3031    diagonalTime, params=params0, globalBasis,
3032    eigenVectors, eigenEnergies, eigenJs,
3033    states, groundEnergy, allEnergies, PrintFun},
3034    (
3035      ln          = theLanthanides[[numE]];
3036      basis       = BasisLSJ[numE, "AsAssociation"->True];
3037      simplifier  = OptionValue["Simplifier"];
3038      PrintFun    = OptionValue["PrintFun"];
3039      PrintFun["> LevelSolver for ",ln," with ",numE," f-electrons."
    ];
3040      PrintFun["> Loading the symbolic level Hamiltonian ..."];
3041      simpleHam   = LevelSimplerSymbolicHamMatrix[numE,
3042        "Simplifier" -> simplifier,
3043        "Overwrite"  -> OptionValue["Overwrite Hamiltonian"]
3044      ];
3045      (* Everything that is not given is set to zero *)
3046      PrintFun["> Setting to zero every parameter not given ..."];
3047      params     = ParamPad[params, "PrintFun" -> PrintFun];
3048      PrintFun[params];
3049      (* Create the numeric hamiltonian *)
3050      PrintFun["> Replacing parameters in the J-blocks of the
    Hamiltonian to produce numeric arrays ..."];
3051      numHam     = N /@ Map[ReplaceInSparseArray[#, params]&,
    simpleHam];
3052      Clear[simpleHam];
3053      (* Eigensolver *)
3054      PrintFun["> Diagonalizing the numerical Hamiltonian within each
     separate J-subspace ..."];
3055      startTime     = Now;
3056      eigensys      = Eigensystem /@ numHam;
3057      endTime       = Now;
3058      diagonalTime  = QuantityMagnitude[endTime-startTime,"Seconds"];
3059      allEnergies   = Flatten[First/@Values[eigensys]];
3060      groundEnergy  = Min[allEnergies];
3061      eigensys      = Map[Chop[{#[[1]]-groundEnergy,#[[2]]}]&,eigensys
    ];
3062      eigensys      = Association@KeyValueMap[#1->{basis[#1],#2} &,
    eigensys];
```

```
3063        PrintFun[">> Diagonalization took ",diagonalTime," seconds."];
3064        If[OptionValue["Return as states"],
3065          (
3066            PrintFun["> Padding the eigenvectors to correspond to the
      level basis ..."];
3067            eigenVectors  = SparseArray @ BlockDiagonalMatrix[Values
      [#[[2, 2]] & /@ eigensys]];
3068            globalBasis   = Flatten[Values[basis], 1];
3069            eigenEnergies = Flatten[Values[#[[2, 1]] & /@ eigensys]];
3070            eigenJs       = Flatten[KeyValueMap[ConstantArray[#1,
      Length[#2[[2, 2]]]] &, eigensys]];
3071            states        = Transpose[{eigenEnergies, eigenJs,
      eigenVectors}];
3072            states        = SortBy[states, First];
3073            eigenVectors  = Last /@ states;
3074            LSJmultiplets = (RemoveTrailingDigits[#[[1]]] <> ToString[
      InputForm[#[[2]]]]) & /@ globalBasis;
3075            majorComponentIndices = Ordering[Abs[#]][[-1]] & /@
      eigenVectors;
3076            levelLabels           = LSJmultiplets[[
      majorComponentIndices]];
3077            Return[{globalBasis, levelLabels, states}];
3078          ),
3079          Return[{basis, eigensys}]
3080        ];
3081      )
3082    ];
3083
3084
3085    (* ##################### Level Description ##################### *)
3086    (* ############################################################ *)
3087
3088    (* ############################################################ *)
3089    (* ##################### Optical Operators ##################### *)
3090
3091    magOp = <||>;
3092
3093    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.
3094    See eqn 15.7 in TASS.
3095    Here it is provided in atomic units in which the Bohr magneton is
        1/2.
3096    \[Mu] = -(1/2) (L + gs S)
3097    We are using the Racah convention for the reduced matrix elements
        in the Wigner-Eckart theorem. See TASS eqn 11.15.
3098    ";
3099    Options[JJBlockMagDip]={"Sparse"->True};
3100    JJBlockMagDip[numE_, braJ_, ketJ_, OptionsPattern[]] := Module[
3101      {braSLJs, ketSLJs,
3102      braSLJ,   ketSLJ,
3103      braSL,    ketSL,
3104      braS,     braL,
3105      ketS,     ketL,
3106      braMJ,    ketMJ,
3107      matValue, magMatrix,
3108      summand1, summand2,
3109      threejays},
3110      (
3111        braSLJs  = AllowedNKSLJMforJTerms[numE,braJ];
3112        ketSLJs  = AllowedNKSLJMforJTerms[numE,ketJ];
3113        magMatrix  = Table[
3114          braSL     = braSLJ[[1]];
3115          ketSL     = ketSLJ[[1]];
3116          {braS, braL}   = FindSL[braSL];
3117          {ketS, ketL}   = FindSL[ketSL];
3118          braMJ       = braSLJ[[3]];
3119          ketMJ       = ketSLJ[[3]];
3120          summand1    = If[Or[braJ  != ketJ,
3121                              braSL != ketSL],
3122            0,
3123            Sqrt[braJ*(braJ+1)*TPO[braJ]]
3124          ];
3125          (* looking at the string includes checking L=L', S=S', and \
```

```
    alpha=\alpha'*)
3126        summand2 = If[braSL!= ketSL,
3127          0,
3128          (gs-1) *
3129            Phaser[braS+braL+ketJ+1] *
3130              Sqrt[TPO[braJ]*TPO[ketJ]] *
3131                SixJay[{braJ,1,ketJ},{braS,braL,braS}] *
3132                  Sqrt[braS(braS+1)TPO[braS]]
3133        ];
3134        matValue = summand1 + summand2;
3135        (* We are using the Racah convention for red matrix elements
    in Wigner-Eckart *)
3136        threejays = (ThreeJay[{braJ, -braMJ}, {1, #}, {ketJ, ketMJ}]
    &) /@ {-1,0,1};
3137        threejays *= Phaser[braJ-braMJ];
3138        matValue  = - 1/2 * threejays * matValue;
3139        matValue,
3140      {braSLJ, braSLJs},
3141      {ketSLJ, ketSLJs}
3142      ];
3143      If[OptionValue["Sparse"],
3144        magMatrix = SparseArray[magMatrix]
3145      ];
3146      Return[magMatrix];
3147    )
3148  ];
3149
3150  Options[TabulateJJBlockMagDipTable]={"Sparse"->True};
3151  TabulateJJBlockMagDipTable[numE_,OptionsPattern[]] := (
3152    JJBlockMagDipTable=<||>;
3153    Js=AllowedJ[numE];
3154    Do[
3155      (
3156        JJBlockMagDipTable[{numE,braJ,ketJ}]=
3157          JJBlockMagDip[numE,braJ,ketJ,"Sparse"->OptionValue["Sparse"
    ]]
3158      ),
3159    {braJ, Js},
3160    {ketJ, Js}
3161    ];
3162    Return[JJBlockMagDipTable];
3163  );
3164
3165  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'|>.";
3166  Options[TabulateManyJJBlockMagDipTables]={"FilenameAppendix"->"","
     Overwrite"->False,"Compressed"->True};
3167  TabulateManyJJBlockMagDipTables[ns_,OptionsPattern[]] := (
3168    fnames=<||>;
3169    Do[
3170    (
3171      ExportFun=If[OptionValue["Compressed"],ExportMZip,Export];
3172      PrintTemporary["#------- numE = ",numE," -------#"];
3173      appendTo     = (OptionValue["FilenameAppendix"]<>"-magDip");
3174      exportFname  = JJBlockMatrixFileName[numE,"FilenameAppendix"->
    appendTo];
3175      fnames[numE] = exportFname;
3176      If[FileExistsQ[exportFname]&&Not[OptionValue["Overwrite"]],
3177        Continue[]
3178      ];
3179      JJBlockMatrixTable = TabulateJJBlockMagDipTable[numE];
3180      If[FileExistsQ[exportFname] && OptionValue["Overwrite"],
3181        DeleteFile[exportFname]
3182      ];
3183      ExportFun[exportFname,  JJBlockMatrixTable];
3184    ),
3185    {numE,ns}
3186    ];
3187    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;
    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}];
  )
];

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>.";
3242 Options[MagDipLineStrength]={"Reload MagOp" -> False, "Units"->"SI"
    , "States" -> All};
3243 MagDipLineStrength[theEigensys_List, numE0_Integer, OptionsPattern
    []] := Module[
3244   {numE, allEigenvecs, Sx, Sy, Sz, Stot, factor},
3245   (
3246     numE = Min[14-numE0, numE0];
3247     (*If not loaded then load it, *)
3248     If[Or[
3249         Not[MemberQ[Keys[magOp], numE]],
3250         OptionValue["Reload MagOp"]],
3251     (
3252       magOp[numE] = ReplaceInSparseArray[#, {gs->2}]& /@
    MagDipoleMatrixAssembly[numE];
3253     )
3254     ];
3255     allEigenvecs = Transpose[Last /@ theEigensys];
3256     Which[OptionValue["States"] === All,
3257       (
3258         {Sx, Sy, Sz} = (ConjugateTranspose[allEigenvecs].#.
    allEigenvecs) & /@ magOp[numE];
3259         Stot         = Abs[Sx]^2+Abs[Sy]^2+Abs[Sz]^2;
3260       ),
3261       IntegerQ[OptionValue["States"]],
3262       (
3263         singleState  = theEigensys[[OptionValue["States"],2]];
3264         {Sx, Sy, Sz} = (ConjugateTranspose[allEigenvecs].#.
    singleState) & /@ magOp[numE];
3265         Stot         = Abs[Sx]^2+Abs[Sy]^2+Abs[Sz]^2;
3266       )
3267     ];
3268     Which[
3269       OptionValue["Units"] == "SI",
3270         Return[4 \[Mu]B^2 * Stot],
3271       OptionValue["Units"] == "Hartree",
3272         Return[Stot],
3273       True,
3274       (
3275         Print["Invalid option for \"Units\". Options are \"SI\" and
     \"Hartree\"."];
3276         Abort[];
3277       )
3278     ];
3279   )
3280 ];
3281
3282 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.
3283 Based on table 7.3 of Thorne 1999, using g2=1.
3284 The energy unit assumed in eigenSys is kayser.
3285 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>.
3286 By default this assumes that the refractive index is unity, this
    may be changed by setting the option \"RefractiveIndex\" to the
    desired value.
3287 The option \"Lifetime\" can be used to return the reciprocal of the
     transition rates. The default is to return the transition rates."
    ;
3288 Options[MagDipoleRates]={"Units"->"SI", "Lifetime"->False, "
    RefractiveIndex"->1};
3289 MagDipoleRates[eigenSys_List, numE0_Integer,OptionsPattern[]] :=
    Module[
3290   {AMD, Stot, eigenEnergies,
3291   transitionWaveLengthsInMeters, nRefractive},
3292   (
3293     nRefractive   = OptionValue["RefractiveIndex"];
3294     numE          = Min[14-numE0, numE0];
```

```
3295      Stot          = MagDipLineStrength[eigenSys, numE, "Units" ->
    OptionValue["Units"]];
3296      eigenEnergies = Chop[First/@eigenSys];
3297      energyDiffs   = Outer[Subtract, eigenEnergies, eigenEnergies];
3298      energyDiffs   = ReplaceDiagonal[energyDiffs, Indeterminate];
3299      (* Energies assumed in kayser.*)
3300      transitionWaveLengthsInMeters = 0.01/energyDiffs;
3301
3302      unitFactor    = Which[
3303      OptionValue["Units"]=="Hartree",
3304      (
3305        (* The bohrRadius factor in SI needed to convert the
    wavelengths which are assumed in m*)
3306        16 \[Pi]^3 (\[Mu]0Hartree /(3 hPlanckHartree)) * bohrRadius^3
3307      ),
3308      OptionValue["Units"]=="SI",
3309      (
3310        16 \[Pi]^3 \[Mu]0/(3 hPlanck)
3311      ),
3312      True,
3313      (
3314        Print["Invalid option for \"Units\". Options are \"SI\" and \
    "Hartree\"."];
3315        Abort[];
3316      )
3317      ];
3318      AMD = unitFactor / transitionWaveLengthsInMeters^3 * Stot *
    nRefractive^3;
3319      Which[OptionValue["Lifetime"],
3320        Return[1/AMD],
3321        True,
3322        Return[AMD]
3323      ]
3324    )
3325  ];
3326
3327  GroundMagDipoleOscillatorStrength::usage = "
      GroundMagDipoleOscillatorStrength[eigenSys, numE] calculates the
      magnetic dipole oscillator strengths between the ground state and
      the excited states as given by eigenSys.
3328  Based on equation 8 of Carnall 1965, removing the 2J+1 factor since
       this degeneracy has been removed by the crystal field.
3329  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.
3330  The energy unit assumed in eigenSys is Kayser.
3331  The oscillator strengths are dimensionless.
3332  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>.
3333  By default this assumes that the refractive index is unity, this
      may be changed by setting the option \"RefractiveIndex\" to the
      desired value.";
3334  Options[GroundMagDipoleOscillatorStrength]={"RefractiveIndex"->1};
3335  GroundMagDipoleOscillatorStrength[eigenSys_List, numE_Integer,
      OptionsPattern[]] := Module[
3336    {eigenEnergies, SMDGS, GSEnergy, energyDiffs,
3337    transitionWaveLengthsInMeters, unitFactor, nRefractive},
3338    (
3339      eigenEnergies    = First/@eigenSys;
3340      nRefractive      = OptionValue["RefractiveIndex"];
3341      SMDGS            = MagDipLineStrength[eigenSys,numE, "Units"->"
    SI", "States"->1];
3342      GSEnergy         = eigenSys[[1,1]];
3343      energyDiffs      = eigenEnergies-GSEnergy;
3344      energyDiffs[[1]] = Indeterminate;
3345      transitionWaveLengthsInMeters = 0.01/energyDiffs;
3346      unitFactor       = (8\[Pi]^2 me)/(3 hPlanck eCharge^2 cLight);
3347      fMDGS            = unitFactor / transitionWaveLengthsInMeters *
     SMDGS * nRefractive;
3348      Return[fMDGS];
3349    )
3350  ];
3351
3352  (* ################### Optical Operators ################### *)
3353  (* ########################################################## *)
```

```
3354
3355  (* ############################################################ *)
3356  (* ################### Printers and Labels ################### *)
3357
3358  PrintL::usage = "PrintL[L] give the string representation of a
        given angular momentum.";
3359  PrintL[L_] := If[StringQ[L], L, StringTake[specAlphabet, {L + 1}]]
3360
3361  FindSL::usage = "FindSL[LS] gives the spin and orbital angular
        momentum that corresponds to the provided string LS.";
3362  FindSL[SL_] := (
3363    FindSL[SL] =
3364    If[StringQ[SL],
3365      {
3366        (ToExpression[StringTake[SL, 1]]-1)/2,
3367        StringPosition[specAlphabet, StringTake[SL, {2}]][[1, 1]]-1
3368      },
3369      SL
3370    ]
3371  );
3372
3373  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.";
3374  PrintSLJ[SLJ_] := (
3375    RowBox[{
3376      SuperscriptBox[" ", 2 SLJ[[1]] + 1],
3377      SubscriptBox[PrintL[SLJ[[2]]], SLJ[[3]]]
3378      }
3379      ] // DisplayForm
3380    );
3381
3382  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."
        ;
3383  PrintSLJM[SLJM_] := (
3384    RowBox[{
3385      SuperscriptBox[" ", 2 SLJM[[1]] + 1],
3386      SubscriptBox[PrintL[SLJM[[2]]], {SLJM[[3]], SLJM[[4]]}]
3387      }
3388      ] // DisplayForm
3389    );
3390
3391  (* ################### Printers and Labels ################### *)
3392  (* ############################################################ *)
3393
3394  (* ############################################################ *)
3395  (* ###################### Term management ###################### *)
3396
3397  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.";
3398  AllowedSLTerms[numE_] := Map[FindSL[First[#]] &, CFPTerms[Min[numE,
        14-numE]]];
3399
3400  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.";
3401  AllowedNKSLTerms[numE_] := Map[First, CFPTerms[Min[numE, 14-numE
        ]]];
3402  AllowedNKSLTerms[0]  = {"1S"};
3403  AllowedNKSLTerms[14] = {"1S"};
3404
3405  MaxJ::usage = "MaxJ[numE] gives the maximum J = S+L that
        corresponds to the configuration f^numE.";
3406  MaxJ[numE_] := Max[Map[Total, AllowedSLTerms[Min[numE, 14-numE]]]];
3407
```

```
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]}
        ];
      allowedSLJ = Join[allowedSLJ, termsSLJ];
      idx1++
    ];
    SortBy[allowedSLJ, Last]
  )
];

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.";
AllowedNKSLJTerms[numE_] := Module[
  {allowedSL, allowedNKSL, allowedSLJ, nn},
  (
    allowedNKSL = AllowedNKSLTerms[numE];
    allowedSL = AllowedSLTerms[numE];
    allowedSLJ = {};
    For[
      nn = 1,
      nn <= Length[allowedSL],
      (
        termSL = allowedSL[[nn]];
        termNKSL = allowedNKSL[[nn]];
        termsSLJ =
          Table[{termNKSL, J},
          {J, Abs[termSL[[1]] - termSL[[2]]], Total[termSL]}
          ];
        allowedSLJ = Join[allowedSLJ, termsSLJ];
        nn++
      )
    ];
    SortBy[allowedSLJ, Last]
  )
];

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.";
AllowedNKSLforJTerms[numE_, J_] := Module[
  {allowedSL, allowedNKSL, allowedSLJ,
  nn, termSL, termNKSL, termsSLJ},
  (
    allowedNKSL = AllowedNKSLTerms[numE];
    allowedSL = AllowedSLTerms[numE];
    allowedSLJ = {};
    For[
      nn = 1,
      nn <= Length[allowedSL],
      (
        termSL = allowedSL[[nn]];
```

```
3471            termNKSL = allowedNKSL[[nn]];
3472            termsSLJ = If[Abs[termSL[[1]] - termSL[[2]]] <= J <= Total[
    termSL],
3473              {{termNKSL, J}},
3474              {}
3475              ];
3476            allowedSLJ = Join[allowedSLJ, termsSLJ];
3477            nn++
3478          )
3479        ];
3480        Return[allowedSLJ]
3481      )
3482    ];
3483
3484    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
      }.";
3485    AllowedSLJMTerms[numE_] := Module[
3486      {allowedSLJ, allowedSLJM,
3487      termSLJ, termsSLJM, nn},
3488      (
3489      allowedSLJ = AllowedSLJTerms[numE];
3490      allowedSLJM = {};
3491      For[
3492        nn = 1,
3493        nn <= Length[allowedSLJ],
3494        nn++,
3495        (
3496          termSLJ = allowedSLJ[[nn]];
3497          termsSLJM =
3498            Table[{termSLJ[[1]], termSLJ[[2]], termSLJ[[3]], M},
3499            {M, - termSLJ[[3]], termSLJ[[3]]}
3500            ];
3501          allowedSLJM = Join[allowedSLJM, termsSLJM];
3502        )
3503      ];
3504      Return[SortBy[allowedSLJM, Last]];
3505      )
3506    ];
3507
3508    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}.";
3509    AllowedNKSLJMforJMTerms[numE_, J_, MJ_] := Module[
3510      {allowedSL, allowedNKSL,
3511      allowedSLJM, nn},
3512      (
3513        allowedNKSL = AllowedNKSLTerms[numE];
3514        allowedSL   = AllowedSLTerms[numE];
3515        allowedSLJM = {};
3516        For[
3517          nn = 1,
3518          nn <= Length[allowedSL],
3519          termSL = allowedSL[[nn]];
3520          termNKSL = allowedNKSL[[nn]];
3521          termsSLJ = If[(Abs[termSL[[1]] - termSL[[2]]]
3522                         <= J
3523                         <= Total[termSL]
3524                         && (Abs[MJ] <= J)
3525                         ),
3526                         {{termNKSL, J, MJ}},
3527                         {}];
3528          allowedSLJM = Join[allowedSLJM, termsSLJ];
3529          nn++
3530        ];
3531        Return[allowedSLJM];
3532      )
3533    ];
3534
3535    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}.";
```

```
AllowedNKSLJMforJTerms[numE_, J_] := Module[
  {MJs, labelsAndMomenta, termsWithJ},
  (
    MJs = AllowedMforJ[J];
    (* Pair LS labels and their {S,L} momenta *)
    labelsAndMomenta = ({#, FindSL[#]}) & /@ AllowedNKSLTerms[numE
  ];
    (* A given term will contain J if |L-S|<=J<=L+S *)
    ContainsJ[{SL_String, {S_, L_}}] := (Abs[S - L] <= J <= (S + L)
  );
    (* Keep just the terms that satisfy this condition *)
    termsWithJ = Select[labelsAndMomenta, ContainsJ];
    (* We don't want to keep the {S,L} *)
    termsWithJ = {#[[1]], J} & /@ termsWithJ;
    (* This is just a quick way of including up all the MJ values
  *)
    Return[Flatten /@ Tuples[{termsWithJ, MJs}]]
  )
];

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 ~~ ")"]],
3597            {2, 4}
3598          ] &,
3599        nielsonKosterLabels];
3600      racahU =
3601        Map[
3602          StringTake[
3603            Flatten[StringCases[Part[# , {3}],
3604              "(" ~~ DigitCharacter ~~ DigitCharacter ~~ ")"]],
3605            {2, 3}
3606          ] &,
3607        nielsonKosterLabels];
3608      fnTermLabels = Join[termLabels, seniorities, racahW, racahU,
    2];
3609      fnTermLabels = Sort[fnTermLabels];
3610      If[OptionValue["Export"],
3611        (
3612          broadFname = FileNameJoin[{moduleDir,"data","fnTerms.m"}];
3613          Export[broadFname, fnTermLabels];
3614        )
3615      ];
3616      Return[fnTermLabels];
3617    )
3618  ];
3619
3620  (* ##################### Term management ##################### *)
3621  (* ############################################################ *)
3622
3623  LoadLaF3Parameters::usage="LoadLaF4Parameters[ln] gives the models
      for the semi-empirical Hamiltonian for the trivalent lanthanide
      ion with symbol ln.";
3624  Options[LoadLaF3Parameters] = {
3625    "Vintage"  -> "Standard",
3626    "With Uncertainties"->False
3627  };
3628  LoadLaF3Parameters[Ln_String, OptionsPattern[]]:= Module[
3629    {paramData, params},
3630    (
3631      paramData = Which[
3632        OptionValue["Vintage"] == "Carnall",
3633        Import[FileNameJoin[{moduleDir, "data", "carnallParams.m"}]],
3634        OptionValue["Vintage"] == "Standard",
3635        Import[FileNameJoin[{moduleDir, "data", "standardParams.m"}]]
3636      ];
3637    params = If[OptionValue["With Uncertainties"],
3638      paramData[Ln],
3639      If[Head[#] === Around, #["Value"], #] & /@ paramData[Ln]
3640    ];
3641    Return[params];
3642    )
3643  ];
3644
3645  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 complementary 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 of T2 the
      relationship is more complex and is controlled by the value of the
       t2Switch variable.";
3646  HoleElectronConjugation[params_] := Module[
3647    {newparams = params},
3648    (
3649      flipSignsOf = Join[{ζ} , cfSymbols, TSymbols];
3650      flipped = Table[
3651        (
3652          flipper -> - newparams[flipper]
3653        ),
3654        {flipper, flipSignsOf}
3655        ];
3656      nonflipped = Table[
3657        (
3658          flipper -> newparams[flipper]
3659        ),
3660        {flipper, Complement[Keys[newparams], flipSignsOf]}
3661        ];
```

```
3662        flippedParams = Association[Join[nonflipped, flipped]];
3663        flippedParams = Select[flippedParams, FreeQ[#, Missing]&];
3664        Return[flippedParams];
3665      )
3666    ];
3667
3668    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.
3669    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.
3670    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\".
3671    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.
3672    The ordered basis in which these eigenvectors are to be interpreted
          is the one corresponding to BasisLSJMJ[numE].
3673    The function admits the following options:
3674    \"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.
3675    \"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.
3676    \"Zeroes\" (list) : A list with symbols assumed to be zero.
3677    ";
3678    Options[IonSolver] = {
3679      "Include Spin-Spin" -> True,
3680      "Overwrite Hamiltonian" -> False,
3681      "Zeroes" -> {}
3682    };
3683    IonSolver[numE_Integer, params0_Association, host_String:"Ln",
          OptionsPattern[]] := Module[
3684      {ln, simplifier, simpleHam, numHam, eigensys,
3685      startTime, endTime, diagonalTime,
3686      params=params0, zeroSymbols},
3687      (
3688        ln = theLanthanides[[numE]];
3689
3690        (* This could be done when replacing values, but this produces
          smaller saved arrays. *)
3691        simplifier = (#-> 0) & /@ OptionValue["Zeroes"];
3692        simpleHam = SimplerSymbolicHamMatrix[numE,
3693          simplifier,
3694          "PrependToFilename" -> host,
3695          "Overwrite" -> OptionValue["Overwrite Hamiltonian"]
3696        ];
3697
3698        (* Note that we don't have to flip signs of parameters for fn
          beyond f7 since the matrix produced
3699        by SimplerSymbolicHamMatrix has already accounted for this. *)
3700
3701        (* Everything that is not given is set to zero *)
3702        params = ParamPad[params];
3703        PrintFun[params];
3704
3705        (* Enforce the override to the spin-spin contribution to the
          magnetic interactions *)
3706        params[\[Sigma]SS] = If[OptionValue["Include Spin-Spin"], 1,
          0];
3707
3708        (* Create the numeric hamiltonian *)
3709        numHam = ReplaceInSparseArray[simpleHam, params];
3710        Clear[simpleHam];
3711
3712        (* Eigensolver *)
3713        PrintFun["> Diagonalizing the numerical Hamiltonian ..."];
3714        startTime = Now;
3715        eigensys  = Eigensystem[numHam];
```

```
3716        endTime   = Now;
3717        diagonalTime = QuantityMagnitude[endTime - startTime, "Seconds"
      ];
3718        PrintFun[">> Diagonalization took ", diagonalTime, " seconds."
      ];
3719        eigensys = Chop[eigensys];
3720        eigensys = Transpose[eigensys];
3721
3722        (* Shift the baseline energy *)
3723        eigensys = ShiftedLevels[eigensys];
3724        (* Sort according to energy *)
3725        eigensys = SortBy[eigensys, First];
3726        Return[eigensys];
3727      )
3728    ];
3729
3730    ShiftedLevels::usage = "ShiftedLevels[eigenSys] takes a list of
       levels of the form
3731    {{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.";
3732    ShiftedLevels[originalLevels_] := Module[
3733      {groundEnergy, shifted},
3734      (
3735        groundEnergy = Sort[originalLevels][[1,1]];
3736        shifted      = Map[{#[[1]] - groundEnergy, #[[2]]} &,
      originalLevels];
3737        Return[shifted];
3738      )
3739    ];
3740
3741
3742    (* ############################################################ *)
3743    (* ############## Optical Transitions for Levels ############## *)
3744
3745    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.
3746    The function returns a list with the following elements:
3747      - basis : A list with the allowed {SL, J} terms in the f^numE
       configuration. Equal to BasisLSJ[numE].
3748      - eigenSys : A list with the eigensystem of the Hamiltonian for
       the f^n configuration.
3749      - levelLabels : A list with the labels of the major components of
        the level eigenstates.
3750      - 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.
3751      The function admits the following options:
3752      \"PrintFun\" : A function that will be used to print the progress
        of the calculations. The default is PrintTemporary.";
3753    Options[JuddOfeltUkSquared] = {"PrintFun" -> PrintTemporary};
3754    JuddOfeltUkSquared[numE_, params_, OptionsPattern[]] := Module[
3755      {eigenChanger, numEH, basis, eigenSys,
3756      Js, Ukmat, LevelUkSquared, kRank,
3757      S, L, Sp, Lp, J, Jp, phase,
3758      braTerm, ketTerm, levelLabels,
3759      eigenVecs, majorComponentIndices},
3760      (
3761        If[Not[ValueQ[ReducedUkTable]],
3762          LoadUk[]
3763        ];
3764        numEH = Min[numE, 14-numE];
3765        PrintFun = OptionValue["PrintFun"];
3766        PrintFun["> Calculating the levels for the given parameters ...
      "];
3767        {basis, majorComponents, eigenSys} = LevelSolver[numE, params];
3768        (* The change of basis matrix to the eigenstate basis *)
3769        eigenChanger = Transpose[Last /@ eigenSys];
3770        PrintFun["Calculating the matrix elements of Uk in the physical
       coupling basis ..."];
```

```
3771      LevelUkSquared = <||>;
3772      Do[(
3773        Ukmat = Table[(
3774            {S, L}   = FindSL[braTerm[[1]]];
3775            J        = braTerm[[2]];
3776            Jp       = ketTerm[[2]];
3777            {Sp, Lp} = FindSL[ketTerm[[1]]];
3778            phase    = Phaser[S + Lp + J + kRank];
3779            Simplify @ (
3780              phase *
3781              Sqrt[TPO[J]*TPO[Jp]] *
3782              SixJay[{J, Jp, kRank}, {Lp, L, S}] *
3783              ReducedUkTable[{numEH, 3, braTerm[[1]], ketTerm[[1]],
    kRank}]
3784              )
3785            ),
3786          {braTerm, basis},
3787          {ketTerm, basis}
3788        ];
3789        Ukmat = (Transpose[eigenChanger] . Ukmat . eigenChanger)^2;
3790        Ukmat = Chop@Ukmat;
3791        LevelUkSquared[kRank] = SymmetrizedArray[Ukmat, Dimensions[
    eigenChanger], Symmetric[{1, 2}]];
3792        ),
3793        {kRank, {2, 4, 6}}
3794      ];
3795      LSJmultiplets = (RemoveTrailingDigits[#[[1]]] <> ToString[
    InputForm[#[[2]]]]) & /@ basis;
3796      eigenVecs = Last /@ eigenSys;
3797      majorComponentIndices = Ordering[Abs[#]][[-1]] & /@ eigenVecs;
3798      levelLabels = LSJmultiplets[[majorComponentIndices]];
3799      Return[{basis, eigenSys, levelLabels, LevelUkSquared}];
3800    )
3801  ];
3802
3803  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.
3804  The third parameter juddOfeltParams is an association with keys
      equal to the three Judd-Ofelt intensity parameters {\[CapitalOmega
      ]2, \[CapitalOmega]4, \[CapitalOmega]6} and corresponding values
      in cm^2.
3805  The local field correction implemented here corresponds to the one
      given by the virtual cavity model of Lorentz.
3806  The function returns a list with the following elements:
3807    - basis : A list with the allowed {SL, J} terms in the f^numE
      configuration. Equal to BasisLSJ[numE].
3808    - eigenSys : A list with the eigensystem of the Hamiltonian for
      the f^n configuration in the level description.
3809    - levelLabels : A list with the labels of the major components of
       the calculated levels.
3810    - 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.
3811  The function admits the following three options:
3812    \"PrintFun\" : A function that will be used to print the progress
       of the calculations. The default is PrintTemporary.
3813    \"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.
3814    \"LocalFieldCorrection\" : The local field correction to be used.
       The default is \"VirtualCavity\". The options are: \"
      VirtualCavity\" and \"EmptyCavity\".
3815  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}];
    )
  ];

  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 levels are
  described by eigenSys in configuration f^numE. 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.
    - 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.
3996  The function admits the following option:
3997    \"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.
3998  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.";
3999  Options[LevelMagDipoleOscillatorStrength]={
4000    "RefractiveIndex" -> 1
4001  };
4002  LevelMagDipoleOscillatorStrength[eigenSys_, numE_, OptionsPattern
      []] := Module[
4003    {eigenEnergies, eigenVecs, levelJs,
4004    energyDiffs, magDipoleOstrength, nRef,
4005    wavelengthsInNM, nRefs, degenDivisor},
4006    (
4007      basis          = BasisLSJ[numE];
4008      eigenEnergies  = First/@eigenSys;
4009      nRef           = OptionValue["RefractiveIndex"];
4010      eigenVecs      = Last/@eigenSys;
4011      levelJs        = #[[2]]&/@eigenSys;
4012      energyDiffs    = -Outer[Subtract,eigenEnergies,eigenEnergies];
4013      energyDiffs  *= kayserToHartree;
4014      magDipoleOstrength = LevelMagDipoleLineStrength[eigenSys, numE,
       "Units"->"Hartree"];
4015      magDipoleOstrength = 2/3 * \[Alpha]Fine^2 * energyDiffs *
      magDipoleOstrength;
4016      degenDivisor       = #1 / ( 2 * levelJs[[#2[[1]]]] + 1 ) &;
4017      magDipoleOstrength = MapIndexed[degenDivisor,
      magDipoleOstrength, {2}];
4018      Which[nRef===1,
4019        True,
4020        NumberQ[nRef],
4021          (
4022            magDipoleOstrength = nRef * magDipoleOstrength;
4023          ),
4024        True,
4025          (
4026            wavelengthsInNM    = Abs[kayserToHartree / energyDiffs] *
       10^7;
4027            nRefs              = Map[nRef, wavelengthsInNM];
4028            magDipoleOstrength = nRefs * magDipoleOstrength;
4029          )
4030      ];
4031      Return[{basis, eigenSys, magDipoleOstrength}];
4032    )
4033  ];
4034
4035  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.
4036  The function admits two optional arguments:
4037    + \"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.
4038   + \"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."
  ;
4039 Options[LevelMagDipoleSpontaneousDecayRates] = {
4040   "Units" -> "SI",
4041   "RefractiveIndex" -> 1};
4042 LevelMagDipoleSpontaneousDecayRates[eigenSys_List, numE_Integer,
  OptionsPattern[]] := Module[
4043   {levMDlineStrength, eigenEnergies, energyDiffs,
4044   levelJs, spontaneousRatesInHartree, spontaneousRatesInSI,
4045   degenDivisor, units, nRef, nRefs, wavelengthsInNM},
4046   (
4047     nRef              = OptionValue["RefractiveIndex"];
4048     units             = OptionValue["Units"];
4049     levMDlineStrength =
  LowerTriangularize@LevelMagDipoleLineStrength[eigenSys,numE,"Units
  "->"Hartree"];
4050     levMDlineStrength = SparseArray[levMDlineStrength];
4051     eigenEnergies     = First /@ eigenSys;
4052     energyDiffs       = Outer[Subtract, eigenEnergies,
  eigenEnergies];
4053     energyDiffs       = kayserToHartree * energyDiffs;
4054     energyDiffs       = SparseArray[LowerTriangularize[energyDiffs
  ]];
4055     levelJs           = #[[2]]& /@ eigenSys;
4056     spontaneousRatesInHartree = 4/3 αFine^5 * energyDiffs^3 *
  levMDlineStrength;
4057     degenDivisor              = #1 / (2*levelJs[[#2[[1]]]] + 1)&;
4058     spontaneousRatesInHartree = MapIndexed[degenDivisor,
  spontaneousRatesInHartree, {2}];
4059     Which[nRef===1,
4060       True,
4061       NumberQ[nRef],
4062         (
4063           spontaneousRatesInHartree = nRef^3 *
  spontaneousRatesInHartree;
4064         ),
4065       True,
4066         (
4067           wavelengthsInNM    = Abs[kayserToHartree / energyDiffs] *
   10^7;
4068           nRefs              = Map[nRef, wavelengthsInNM];
4069           spontaneousRatesInHartree = nRefs^3 *
  spontaneousRatesInHartree;
4070         )
4071     ];
4072     If[units == "SI",
4073       (
4074         spontaneousRatesInSI = 1/hartreeTime *
  spontaneousRatesInHartree;
4075         Return[SparseArray@spontaneousRatesInSI];
4076       ),
4077       Return[SparseArray@spontaneousRatesInHartree];
4078     ];
4079   )
4080 ];
4081
4082 (* ############# Optical Transitions for Levels ############## *)
4083 (* ############################################################ *)
4084
4085 (* ############################################################ *)
4086 (* ################## Eigensystem analysis ################### *)
4087
4088 PrettySaundersSL::usage = "PrettySaundersSL[SL] produces a human-
  readeable 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\
  ".";
4089 Options[PrettySaundersSL] = {"Representation" -> "Ket"};
```

```
PrettySaundersSL[SL_, OptionsPattern[]] := (
  If[StringQ[SL],
  (
    {S,L} = FindSL[SL];
    L     = StringTake[SL,{2,-1}];
  ),
    {S,L}=SL
  ];
  pretty = RowBox[{
      AdjustmentBox[Style[2*S+1,Smaller], BoxBaselineShift->-1,
 BoxMargins->0],
      AdjustmentBox[PrintL[L]]
    }
  ];
  pretty = DisplayForm[pretty];
  pretty = Which[
    OptionValue["Representation"]=="Ket",
      Ket[pretty],
    OptionValue["Representation"]=="Symbol",
      pretty
  ];
  Return[pretty];
);

PrettySaundersSLJmJ::usage = "PrettySaundersSLJmJ[{SL, J, mJ}]
 produces a formatted symbol for the given basis vector {SL, J, mJ
 }.";
Options[PrettySaundersSLJmJ] = {"Representation" -> "Ket"};
PrettySaundersSLJmJ[{SL_, J_, mJ_}, OptionsPattern[]] := (If[
  StringQ[SL],
  ({S, L} = FindSL[SL];
    L = StringTake[SL, {2, -1}];
    ),
  {S, L} = SL];
  pretty = RowBox[{AdjustmentBox[Style[2*S + 1, Smaller],
      BoxBaselineShift -> -1, BoxMargins -> 0],
      AdjustmentBox[PrintL[L], BoxMargins -> -0.2],
      AdjustmentBox[
      Style[Row[{InputForm[J], ",", mJ}], Small],
      BoxBaselineShift -> 1,
      BoxMargins -> {{0.7, 0}, {0.4, 0.4}}]}];
  pretty = DisplayForm[pretty];
  If[OptionValue["Representation"] == "Ket",
    pretty = Ket[pretty]
  ];
  Return[pretty];
);

PrettySaundersSLJ::usage = "PrettySaundersSLJ[{SL, J}] produces a
 formatted symbol for the given level {SL, J}. SL can be either a
 list of two numbers representing S and L or a string representing
 the spin multiplicity in spectroscopic notation. J is the total
 angular momentum to which S and L have coupled to. The option \"
 Representation\" can be used to specify whether the output is
 given as a symbol or as a ket. The default is \"Ket\".";
Options[PrettySaundersSLJ] = {"Representation"->"Ket"};
PrettySaundersSLJ[{SL_,J_},OptionsPattern[]] := (
  If[StringQ[SL],
  (
    {S,L}=FindSL[SL];
    L=StringTake[SL,{2,-1}];
  ),
    {S,L}=SL
  ];
  pretty = RowBox[{
      AdjustmentBox[Style[2*S+1,Smaller],BoxBaselineShift->-1,
 BoxMargins->0],
      AdjustmentBox[PrintL[L],BoxMargins->-0.2],
      AdjustmentBox[Style[InputForm[J],Small,FontTracking->"Narrow"
 ],BoxBaselineShift->1,BoxMargins->{{0.7,0},{0.4,0.4}}]
    }
  ];
  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 formatted 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 formatted 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]]
    )
  ];

  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.";
```

```
ParseStatesByProbabilitySum[eigensys_, basis_, probSum_, roundTo_ : 0.01, maxParts_: 20] := Module[
  {parsedByProb, numStates, state, energy,
  eigenVec, amplitudes, probs, ordering,
  orderedProbs, truncationIndex, accProb,
  thresholdIndex, chosenIndices, majorComponents,
  majorAmplitudes, absMajorAmplitudes, notnullAmplitudes, majorRep
  },
  (
    numStates    = Length[eigensys];
    parsedByProb = Table[(
      state              = eigensys[[idx]];
      {energy, eigenVec} = state;
      (*Round them up*)
      amplitudes         = Round[eigenVec, roundTo];
      probs              = Round[Abs[eigenVec^2], roundTo];
      ordering           = Reverse[Ordering[probs]];
      (*Order the probabilities from high to low*)
      orderedProbs       = probs[[ordering]];
      (*To speed up Accumulate, assume that only as much as maxParts will be needed*)
      truncationIndex    = Min[maxParts, Length[orderedProbs]];
      orderedProbs       = orderedProbs[[;;truncationIndex]];
      (*Accumulate the probabilities*)
      accProb            = Accumulate[orderedProbs];
      (*Find the index of the first element in accProb that is greater than probSum*)
      thresholdIndex     = Min[Length[accProb], FindThresholdPosition[accProb, probSum]];
      (*Grab all the indicees up till that one*)
      chosenIndices      = ordering[[;; thresholdIndex]];
      (*Select the corresponding elements from the basis*)
      majorComponents    = basis[[chosenIndices]];
      (*Select the corresponding amplitudes*)
      majorAmplitudes    = amplitudes[[chosenIndices]];
      (*Take their absolute value*)
      absMajorAmplitudes = Abs[majorAmplitudes];
      (*Make sure that there are no effectively zero contributions*)
      notnullAmplitudes  = Flatten[Position[absMajorAmplitudes, x_ /; x != 0]];
      (* majorComponents    = PrettySaundersSLJmJ [{#[[1]],#[[2]],#[[3]]}] & /@ majorComponents; *)
      majorComponents    = PrettySaundersSLJmJ /@ majorComponents;
      majorAmplitudes    = majorAmplitudes[[notnullAmplitudes]];
      majorComponents    = majorComponents[[notnullAmplitudes]];
      (*Multiply and add to build the final Ket*)
      majorRep           = majorAmplitudes . majorComponents;
      {energy, majorRep}
      ), {idx, numStates}];
    Return[parsedByProb];
  )
];

(* ################## Eigensystem analysis ################### *)
(* ############################################################ *)

(* ############################################################ *)
(* ########################## Misc ########################### *)

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.";
SymbToNum[expr_, replacementAssociation_] := (
  includedKeys = Keys[replacementAssociation];
  (*If a key is not defined, make its value zero.*)
  fullAssociation = Table[(
    If[MemberQ[includedKeys, key],
      ToExpression[key]->replacementAssociation[key],
      ToExpression[key]->0
    ]
  ),
  {key, paramSymbols}];
  Return[expr/.fullAssociation];
```

```
);

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.";
SimpleConjugate[expr_] := expr /. Complex[a_, b_] :> a - I b;

ExportMZip::usage = "ExportMZip[\"dest.[zip,m]\", expr] 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, mxImportName, 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 ..."];
      imported = Import[mImportName];
      If[OptionValue["Leave Uncompressed"],
        (
          Export[mxImportName, imported];
        )
      ];
      Return[Import[mImportName]];
    )
    ];
    imported = Import[zipImportName, importKey];
    If[OptionValue["Leave Uncompressed"],
      (
        Export[mImportName, imported];
        Export[mxImportName, imported];
      )
    ];
    Return[imported];
  )
];
```

```
4396
4397  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.";
4398  ReplaceInSparseArray[sparseA_SparseArray, rules_] := (
4399    SparseArray[Automatic,
4400      sparseA["Dimensions"],
4401      sparseA["Background"] /. rules,
4402      {
4403        1,
4404        {sparseA["RowPointers"], sparseA["ColumnIndices"]},
4405        sparseA["NonzeroValues"] /. rules
4406      }
4407      ]
4408  );
4409
4410  MapToSparseArray::usage = "MapToSparseArray[sparseArray, function]
        takes a sparse array and returns a sparse array after the function
         has been applied to it.";
4411  MapToSparseArray[sparseA_SparseArray, func_] := Module[
4412    {nonZ, backg, mapped},
4413    (
4414      nonZ   = func /@ sparseA["NonzeroValues"];
4415      backg  = func[sparseA["Background"]];
4416      mapped = SparseArray[Automatic,
4417        sparseA["Dimensions"],
4418        backg,
4419        {
4420          1,
4421          {sparseA["RowPointers"], sparseA["ColumnIndices"]},
4422          nonZ
4423        }
4424      ];
4425      Return[mapped];
4426    )
4427  ];
4428
4429  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.";
4430  Options[ParseTeXLikeSymbol] = {"Form" -> "List"};
4431  ParseTeXLikeSymbol[bigString_, OptionsPattern[]] := Module[
4432    {form, mainSymbol, symbols},
4433    (
4434      form = OptionValue["Form"];
4435      (* parse greek *)
4436      symbols = Table[(
4437          str = StringReplace[string, {"\\alpha" -> "α",
4438            "\\beta" -> "β",
4439            "\\gamma" -> "γ",
4440            "\\psi" -> "\[Psi]"}];
4441          symbol = Which[
4442            StringContainsQ[str, "_"] && Not[StringContainsQ[str, "^"
        ]],
4443            (
4444              (*yes sub no sup*)
4445              mainSymbol = StringSplit[str, "_"][[1]];
4446              mainSymbol = ToExpression[mainSymbol];
4447
4448              subPart =
4449                StringCases[str,
4450                  RegularExpression@"\\{(.*?)\\}" -> "$1"][[1]];
4451              Subscript[mainSymbol, subPart]
4452            ),
4453            Not[StringContainsQ[str, "_"]] && StringContainsQ[str, "^
        "],
4454            (
4455              (*no sub yes sup*)
4456              mainSymbol = StringSplit[str, "^"][[1]];
4457              mainSymbol = ToExpression[mainSymbol];
4458
```

```
              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.
    - The reciprocal of the 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,
  "Energy Unit" -> "cm^-1"
  };
EnergyLevelDiagram[states_, OptionsPattern[]] := Module[
  {energies, epi, explora},
  (
    energies = If[Length[Dimensions[states]]==1,
      states,
      First/@states
    ];
    epi = OptionValue["Epilog"];
    explora = If[OptionValue["Explorer"],
      ExploreGraphics,
      Identity
    ];
    frameLabel = "E (" <> OptionValue["Energy Unit"] <> ")";
    plotTips = Which[
      OptionValue["Energy Unit"] == "cm^-1",
      Tooltip[{{#, 0}, {#, 1}}, {Quantity[#/8065.54429, "eV"],
  Quantity[#, 1/"Centimeters"]}] &/@ energies,
      OptionValue["Energy Unit"] == "eV",
      Tooltip[{{#, 0}, {#, 1}}, {Quantity[# * 8065.54429, 1/"
  Centimeters"], Quantity[#, "eV"]}] &/@ energies,
      True,
      Tooltip[{{#, 0}, {#, 1}}, Quantity[# , OptionValue["Energy
  Unit"]]] &/@ energies
    ];
    explora@ListPlot[plotTips,
      Joined       -> True,
      PlotStyle    -> Black,
      AspectRatio  -> OptionValue["AspectRatio"],
      ImageSize    -> OptionValue["ImageSize"],
      Frame        -> True,
      PlotRange    -> {All, {0, 1}},
      FrameTicks   -> {{None, None}, {Automatic, Automatic}},
      FrameStyle   -> Directive[15, Dashed, Thin],
      PlotLabel    -> Style[OptionValue["Title"], 15, Bold],
      Background   -> OptionValue["Background"],
      FrameLabel   -> {frameLabel},
      Epilog       -> epi]
  )
];

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.
Based on ZeitPolizei @ https://mathematica.stackexchange.com/
  questions/7142/how-to-manipulate-2d-plots.
The option \"OptAxesRedraw\" can be used to specify whether the
  axes should be redrawn. The default is False.";
Options[ExploreGraphics] = {OptAxesRedraw -> False};
ExploreGraphics[graph_Graphics, opts : OptionsPattern[]] := With[
  {
    gr  = First[graph],
    opt = DeleteCases[Options[graph],
          PlotRange -> PlotRange | AspectRatio | AxesOrigin -> _],
    plr = PlotRange /. AbsoluteOptions[graph, PlotRange],
    ar  = AspectRatio /. AbsoluteOptions[graph, AspectRatio],
    ao  = AbsoluteOptions[AxesOrigin],
    rectangle = {Dashing[Small],
      Line[{#1,
```

```
4589                {First[#2], Last[#1]},
4590                #2,
4591                {First[#1], Last[#2]},
4592                #1}]} &,
4593      optAxesRedraw = OptionValue[OptAxesRedraw]
4594    },
4595    DynamicModule[
4596        {dragging=False, first, second, rx1, rx2, ry1, ry2,
4597        range = plr},
4598        {{rx1, rx2}, {ry1, ry2}} = plr;
4599      Panel@
4600      EventHandler[
4601        Dynamic@Graphics[
4602          If[dragging, {gr, rectangle[first, second]}, gr],
4603          PlotRange -> Dynamic@range,
4604          AspectRatio -> ar,
4605          AxesOrigin -> If[optAxesRedraw,
4606            Dynamic@Mean[range\[Transpose]], ao],
4607          Sequence @@ opt],
4608        {{"MouseDown", 1} :> (
4609          first = MousePosition["Graphics"]
4610          ),
4611        {"MouseDragged", 1} :> (
4612          dragging = True;
4613          second = MousePosition["Graphics"]
4614          ),
4615        "MouseClicked" :> (
4616          If[CurrentValue@"MouseClickCount"==2,
4617            range = plr];
4618          ),
4619        {"MouseUp", 1} :> If[dragging,
4620          dragging = False;
4621
4622          range = {{rx1, rx2}, {ry1, ry2}} =
4623            Transpose@{first, second};
4624          range[[2]] = {0, 1}],
4625        {"MouseDown", 2} :> (
4626          first = {sx1, sy1} = MousePosition["Graphics"]
4627          ),
4628        {"MouseDragged", 2} :> (
4629          second = {sx2, sy2} = MousePosition["Graphics"];
4630          rx1 = rx1 - (sx2 - sx1);
4631          rx2 = rx2 - (sx2 - sx1);
4632          ry1 = ry1 - (sy2 - sy1);
4633          ry2 = ry2 - (sy2 - sy1);
4634          range = {{rx1, rx2}, {ry1, ry2}};
4635          range[[2]] = {0, 1};
4636          )}]]];
4637
4638  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.";
4639  Options[LabeledGrid]={
4640      ItemSize->Automatic,
4641      Alignment->Center,
4642      Frame->All,
4643      "Separator"->",",
4644      "Pivot"->""
4645  };
4646  LabeledGrid[data_,rowHeaders_,columnHeaders_,OptionsPattern[]] :=
       Module[
4647    {gridList=data, rowHeads=rowHeaders, colHeads=columnHeaders},
4648    (
4649      separator=OptionValue["Separator"];
4650      pivot=OptionValue["Pivot"];
4651      gridList=Table[
4652              Tooltip[
4653                data[[rowIdx,colIdx]],
4654                DisplayForm[
4655                  RowBox[{rowHeads[[rowIdx]],
4656                          separator,
4657                          colHeads[[colIdx]]}
4658                        ]
```

```
                        ]
                      ],
          {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"]]
     )

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 \"OverlayValues\"
   can be used to specify whether the matrix elements should be
  displayed on top of the matrix plot.";
Options[HamiltonianMatrixPlot] = Join[Options[MatrixPlot], {"Hover"
   -> True, "OverlayValues" -> True}];
HamiltonianMatrixPlot[hamMatrix_, basisLabels_, opts :
  OptionsPattern[]] := (
   braLabels = DisplayForm[RowBox[{"\[LeftAngleBracket]", #, "\[
  RightBracketingBar]"}]] & /@ basisLabels;
   ketLabels = DisplayForm[Rotate[RowBox[{"\[LeftBracketingBar]", #,
    "\[RightAngleBracket]"}],\[Pi]/2]] & /@ basisLabels;
   ketLabelsUpright = DisplayForm[RowBox[{"\[LeftBracketingBar]", #,
    "\[RightAngleBracket]"}]] & /@ basisLabels;
   numRows = Length[hamMatrix];
   numCols = Length[hamMatrix[[1]]];
   epiThings = Which[
     And[OptionValue["Hover"], Not[OptionValue["OverlayValues"]]],
     Flatten[
       Table[
         Tooltip[
           {
             Transparent,
             Rectangle[
             {j - 1, numRows - i},
             {j - 1, numRows - i} + {1, 1}
             ]
           },
         Row[{braLabels[[i]],ketLabelsUpright[[j]],"=",hamMatrix[[i,
    j]]}]
         ],
       {i, 1, numRows},
       {j, 1, numCols}
       ]
     ],
     And[OptionValue["Hover"], OptionValue["OverlayValues"]],
     Flatten[
       Table[
         Tooltip[
```

```
            {
              Transparent,
              Rectangle[
              {j - 1, numRows - i},
              {j - 1, numRows - i} + {1, 1}
              ]
            },
          DisplayForm[RowBox[{"\[LeftAngleBracket]", basisLabels[[i
  ]], "\[LeftBracketingBar]", basisLabels[[j]], "\[RightAngleBracket
  ]"}]]
          ],
        {i, numRows},
        {j, numCols}
        ]
      ],
      True,
      {}
      ];
    textOverlay = If[OptionValue["OverlayValues"],
      (
        Flatten[
        Table[
          Text[hamMatrix[[i, j]],
            {j - 1/2, numRows - i + 1/2}
          ],
        {i, 1, numRows},
        {j, 1, numCols}
        ]
        ]
      ),
      {}
      ];
    epiThings = Join[epiThings, textOverlay];
    MatrixPlot[hamMatrix,
      FrameTicks -> {
        {Transpose[{Range[Length[braLabels]], braLabels}], None},
        {None, Transpose[{Range[Length[ketLabels]], ketLabels}]}
        },
      Evaluate[FilterRules[{opts}, Options[MatrixPlot]]],
      Epilog -> epiThings
    ]
  );

  (* ################## Some Plotting Routines ################## *)
  (* ############################################################ *)

  (* ############################################################ *)
  (* ###################### Load Functions ###################### *)

  LoadAll::usage = "LoadAll[] executes most Load* functions.";
  LoadAll[] := (
    LoadTermLabels[];
    LoadCFP[];
    LoadUk[];
    LoadV1k[];
    LoadT22[];
    LoadSOOandECSOLS[];

    LoadElectrostatic[];
    LoadSpinOrbit[];
    LoadSOOandECSO[];
    LoadSpinSpin[];
    LoadThreeBody[];
    LoadChenDeltas[];
    LoadCarnall[];
  );

  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.";

  LoadTermLabels::usage = "LoadTermLabels[] loads into the session
    the labels for the terms in the f^n configurations.";
  LoadTermLabels[] := (
```

```
4782      If[ValueQ[fnTermLabels], Return[]];
4783      PrintTemporary["Loading data for state labels in the f^n
          configurations..."];
4784      fnTermsFname = FileNameJoin[{moduleDir, "data", "fnTerms.m"}];
4785
4786      If[!FileExistsQ[fnTermsFname],
4787        (PrintTemporary[">> fnTerms.m not found, generating ..."];
4788          fnTermLabels = ParseTermLabels["Export"->True];
4789        ),
4790        fnTermLabels = Import[fnTermsFname];
4791      ];
4792    );
4793
4794    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}";
4795
4796    LoadCarnall::usage = "LoadCarnall[] loads data for trivalent
          lanthanides in LaF3 using the data from Bill Carnall's 1989 paper.
          ";
4797    LoadCarnall[] := (
4798      If[ValueQ[Carnall], Return[]];
4799      carnallFname = FileNameJoin[{moduleDir, "data", "Carnall.m"}];
4800      If[!FileExistsQ[carnallFname],
4801        (PrintTemporary[">> Carnall.m not found, generating ..."];
4802          Carnall = ParseCarnall[];
4803        ),
4804        Carnall = Import[carnallFname];
4805      ];
4806    );
4807
4808    LoadChenDeltas::usage = "LoadChenDeltas[] loads the differences
          noted by Chen.";
4809    LoadChenDeltas[] := (
4810      If[ValueQ[chenDeltas], Return[]];
4811      PrintTemporary["Loading the association of discrepancies found by
           Chen ..."];
4812      chenDeltasFname = FileNameJoin[{moduleDir, "data", "chenDeltas.m"
          }];
4813      If[!FileExistsQ[chenDeltasFname],
4814        (PrintTemporary[">> chenDeltas.m not found, generating ..."];
4815          chenDeltas = ParseChenDeltas[];
4816        ),
4817        chenDeltas = Import[chenDeltasFname];
4818      ];
4819    );
4820
4821    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";
4822    Options[ParseChenDeltas] = {"Export" -> True};
4823    ParseChenDeltas[OptionsPattern[]] := (
4824      chenDeltasRaw = Import[FileNameJoin[{moduleDir, "data", "the-chen
          -deltas-A.csv"}]];
4825      chenDeltasRaw = chenDeltasRaw[[2 ;;]];
4826      chenDeltas = <||>;
4827      chenDeltasA = <||>;
4828      Off[Power::infy];
4829      Do[
4830        ({right, wrong} = {chenDeltasRaw[[row]][[4 ;;]],
4831          chenDeltasRaw[[row + 1]][[4 ;;]]};
4832        key = chenDeltasRaw[[row]][[1 ;; 3]];
4833        repRule = (#[[1]] -> #[[2]]*#[[1]]) & /@
4834          Transpose[{{M0, M2, M4, P2, P4, P6}, right/wrong}];
4835        chenDeltasA[key] = <|"right" -> right, "wrong" -> wrong,
4836          "repRule" -> repRule|>;
4837        chenDeltasA[{key[[1]], key[[3]], key[[2]]}] = <|"right" ->
          right,
4838          "wrong" -> wrong, "repRule" -> repRule|>;
4839        ),
4840        {row, 1, Length[chenDeltasRaw], 2}];
4841      chenDeltas["A"] = chenDeltasA;
4842
```

```
    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];
  );
```

```
4969
4970  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.
4971  These are according to the tables from Nielson & Koster.";
4972
4973  CFPAssoc::usage = "CFPAssoc is an association where keys are of
        lists of the form {num_electrons, daughterTerm, 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[].
4974  These are according to the tables from Nielson & Koster.";
4975
4976  LoadCFP::usage = "LoadCFP[] loads CFP, CFPAssoc, and CFPTable into
        the session.";
4977  LoadCFP[] := (
4978    If[And[ValueQ[CFP], ValueQ[CFPTable], ValueQ[CFPAssoc]],Return
      []];
4979
4980    PrintTemporary["Loading CFPtable ..."];
4981    CFPTablefname = FileNameJoin[{moduleDir, "data", "CFPTable.m"}];
4982    If[!FileExistsQ[CFPTablefname],
4983      (PrintTemporary[">> CFPTable.m not found, generating ..."];
4984        CFPTable = GenerateCFPTable["Export"->True];
4985      ),
4986      CFPTable = Import[CFPTablefname];
4987    ];
4988
4989    PrintTemporary["Loading CFPs.m ..."];
4990    CFPfname = FileNameJoin[{moduleDir, "data", "CFPs.m"}];
4991    If[!FileExistsQ[CFPfname],
4992      (PrintTemporary[">> CFPs.m not found, generating ..."];
4993        CFP = GenerateCFP["Export"->True];
4994      ),
4995      CFP = Import[CFPfname];
4996    ];
4997
4998    PrintTemporary["Loading CFPAssoc.m ..."];
4999    CFPAfname = FileNameJoin[{moduleDir, "data", "CFPAssoc.m"}];
5000    If[!FileExistsQ[CFPAfname],
5001      (PrintTemporary[">> CFPAssoc.m not found, generating ..."];
5002        CFPAssoc = GenerateCFPAssoc["Export"->True];
5003      ),
5004      CFPAssoc = Import[CFPAfname];
5005    ];
5006  );
5007
5008  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[].";
5009
5010  LoadUk::usage = "LoadUk[] loads into session the reduced matrix
        elements for unit tensor operators.";
5011  LoadUk[] := (
5012    If[ValueQ[ReducedUkTable], Return[]];
5013    PrintTemporary["Loading the association of reduced matrix
      elements for unit tensor operators ..."];
5014    ReducedUkTableFname = FileNameJoin[{moduleDir, "data", "
      ReducedUkTable.m"}];
5015    If[!FileExistsQ[ReducedUkTableFname],
5016      (PrintTemporary[">> ReducedUkTable.m not found, generating ..."
      ];
5017        ReducedUkTable = GenerateReducedUkTable[7];
5018      ),
5019      ReducedUkTable = Import[ReducedUkTableFname];
5020    ];
5021  );
5022
5023  ElectrostaticTable::usage = "ElectrostaticTable[{n, SL, SpLp}]
        provides the calculated result of Electrostatic[{n, SL, SpLp}].
        Load using LoadElectrostatic[].";
5024
```

```
LoadElectrostatic::usage = "LoadElectrostatic[] loads the reduced matrix elements for the electrostatic interaction.";
LoadElectrostatic[] := (
  If[ValueQ[ElectrostaticTable], Return[]];
  PrintTemporary["Loading the association of matrix elements for the electrostatic interaction ..."];
  ElectrostaticTablefname = FileNameJoin[{moduleDir, "data", "ElectrostaticTable.m"}];
  If[!FileExistsQ[ElectrostaticTablefname],
    (PrintTemporary[">> ElectrostaticTable.m not found, generating ..."];
      ElectrostaticTable = GenerateElectrostaticTable[7];
    ),
    ElectrostaticTable = Import[ElectrostaticTablefname];
  ];
);

LoadV1k::usage = "LoadV1k[] loads into session the matrix elements of V1k.";
LoadV1k[] := (
  If[ValueQ[ReducedV1kTable], Return[]];
  PrintTemporary["Loading the association of matrix elements for V1k ..."];
  ReducedV1kTableFname = FileNameJoin[{moduleDir, "data", "ReducedV1kTable.m"}];
  If[!FileExistsQ[ReducedV1kTableFname],
    (PrintTemporary[">> ReducedV1kTable.m not found, generating ..."];
      ReducedV1kTable = GenerateReducedV1kTable[7];
    ),
    ReducedV1kTable = Import[ReducedV1kTableFname];
  ]
);

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 ..."
```

```
    ];
5082     SOOandECSOTableFname = FileNameJoin[{moduleDir, "data", "
    SOOandECSOTable.m"}];
5083     If[!FileExistsQ[SOOandECSOTableFname],
5084       (PrintTemporary[">> SOOandECSOTable.m not found, generating ...
    "];
5085         SOOandECSOTable = GenerateSOOandECSOTable[7, "Export"->True];
5086       ),
5087       SOOandECSOTable = Import[SOOandECSOTableFname];
5088     ];
5089   );
5090
5091   LoadT22::usage = "LoadT22[] loads into session the matrix elements
      of the double tensor operator T22.";
5092   LoadT22[] := (
5093     If[ValueQ[T22Table], Return[]];
5094     PrintTemporary["Loading the association of reduced T22 matrix
    elements ..."];
5095     T22TableFname = FileNameJoin[{moduleDir, "data", "ReducedT22Table
    .m"}];
5096     If[!FileExistsQ[T22TableFname],
5097       (PrintTemporary[">> ReducedT22Table.m not found, generating ...
    "];
5098         T22Table = GenerateT22Table[7];
5099       ),
5100       T22Table = Import[T22TableFname];
5101     ];
5102   );
5103
5104   LoadSpinSpin::usage = "LoadSpinSpin[] loads into session the matrix
       elements of spin-spin.";
5105   LoadSpinSpin[] := (
5106     If[ValueQ[SpinSpinTable], Return[]];
5107     PrintTemporary["Loading the association of matrix elements for
    spin-spin ..."];
5108     SpinSpinTableFname = FileNameJoin[{moduleDir, "data", "
    SpinSpinTable.m"}];
5109     If[!FileExistsQ[SpinSpinTableFname],
5110       (PrintTemporary[">> SpinSpinTable.m not found, generating ..."
    ];
5111         SpinSpinTable = GenerateSpinSpinTable[7, "Export" -> True];
5112       ),
5113       SpinSpinTable = Import[SpinSpinTableFname];
5114     ];
5115   );
5116
5117   LoadThreeBody::usage = "LoadThreeBody[] loads into session the
      matrix elements of three-body configuration-interaction effects.";
5118   LoadThreeBody[] := (
5119     If[ValueQ[ThreeBodyTable], Return[]];
5120     PrintTemporary["Loading the association of matrix elements for
    three-body configuration-interaction effects ..."];
5121     ThreeBodyFname   = FileNameJoin[{moduleDir, "data", "
    ThreeBodyTable.m"}];
5122     ThreeBodiesFname = FileNameJoin[{moduleDir, "data", "
    ThreeBodyTables.m"}];
5123     If[!FileExistsQ[ThreeBodyFname],
5124       (PrintTemporary[">> ThreeBodyTable.m not found, generating ..."
    ];
5125         {ThreeBodyTable, ThreeBodyTables} = GenerateThreeBodyTables["
    Export" -> True];
5126       ),
5127       ThreeBodyTable  = Import[ThreeBodyFname];
5128       ThreeBodyTables = Import[ThreeBodiesFname];
5129     ];
5130   );
5131
5132   (* ###################### Load Functions ###################### *)
5133   (* ########################################################### *)
5134
5135 End[];
5136
5137 LoadTermLabels[];
5138 LoadCFP[];
5139
5140 EndPackage[];
```

18.2 fittings.m

This file has code useful for fitting the Hamiltonian.

```
(*-------------------------------------------------------------------*)
(*~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~*)
(*~~~+-----------------------------------------------------------+~~~*)
(*~~~|                                                           |~~~*)
(*~~~|                 _____ __  __  _                           |~~~*)
(*~~~|               / __(_) /_/ /_(_)___  ____ ______          |~~~*)
(*~~~|              / /_/ / __/ __/ / __ \/ __ `/ ___/          |~~~*)
(*~~~|             / __/ / /_/ /_/ / / / / /_/ (__  )           |~~~*)
(*~~~|            /_/ /_/\__/\__/_/_/ /_/\__, /____/            |~~~*)
(*~~~|                                   /____/                  |~~~*)
(*~~~|                                                           |~~~*)
(*~~~+-----------------------------------------------------------+~~~*)
(*~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~*)
(*~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~*)
(*~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~*)
(*~~~~+---------------------------------------------------------+~~~~*)
(*~~~~|                                                         |~~~~*)
(*~~~~|                                                         |~~~~*)
(*~~~~| This script puts together some code useful for fitting  |~~~~*)
(*~~~~|            the model Hamiltonian to data.               |~~~~*)
(*~~~~|                                                         |~~~~*)
(*~~~~|                                                         |~~~~*)
(*~~~~+---------------------------------------------------------+~~~~*)
(*~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~*)
(*~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~*)
+-------------------------------------------------------------------*)

BeginPackage["DavidLizarazo`fittings`"];

Get[FileNameJoin[{DirectoryName[$InputFileName],"qlanth.m"}]];
Get[FileNameJoin[{DirectoryName[$InputFileName],"qonstants.m"}]];
Get[FileNameJoin[{DirectoryName[$InputFileName],"misc.m"}]];

LoadCarnall[];
LoadFreeIon[];

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.
    }
  |>;

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[(
      {
        "",
        Dynamic[Framed[progressMessage],
          UpdateInterval -> OptionValue["UpdateInterval"]],
        Dynamic[
        GraphicsColumn[
         {ListPlot[rmsHistory,
           PlotMarkers -> "OpenMarkers",
           Frame -> True,
           FrameLabel -> {"Iteration", "RMS"},
           ImageSize -> 800,
           AspectRatio -> 1/3,
           FrameStyle -> Directive[Thick, 15],
           PlotLabel -> If[Length[rmsHistory] != 0, rmsHistory[[-1]],
    ""]
```

```
            ],
           ListPlot[(#/#[[1]]) & /@ Transpose[paramSols],
            Joined -> True,
            PlotRange -> {All, {-5, 5}},
            Frame -> True,
            ImageSize -> 800,
            AspectRatio -> 1,
            FrameStyle -> Directive[Thick, 15],
            FrameLabel -> {"Iteration", "Params"}
            ]
           }
          ],
         TrackedSymbols :> {rmsHistory, paramSols},
         UpdateInterval -> OptionValue["UpdateInterval"]
         ],
        Dynamic[
         TextCell[
          If[
           Length[paramSols] > 0,
           TableForm[Transpose[{stringPartialVars, paramSols[[-1]]}]],
           ""
           ],
          "Output"
          ],
         TrackedSymbols :> {paramSols, stringPartialVars}
         ]
        }
       ),
      WindowSize -> {600, 1000},
      WindowSelected -> True,
      TextAlignment -> Center,
      WindowTitle -> "Solver Progress"
      ]
    ];
    Return[nb];
    );

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`";
energyCostFunTemplate = StringTemplate["
TheRightEnergyPath[`varPatterns`]:= (
  {eigenEnergies, eigenVecs} = Eigensystem[compHam[`vars`]];
  ordering      = Ordering[eigenEnergies];
  eigenEnergies = eigenEnergies - Min[eigenEnergies];
  states        = Transpose[{eigenEnergies, eigenVecs}];
  states        = states[[ordering]];
  coarseStates  = ParseStates[states, basis];
  coarseStates  = {#[[1]],#[[-1]]}& /@ coarseStates;
  (* The eigenvectors need to be simplified in order to compare labels to labels *)
  missingLevels = Length[coarseStates]-Length[expData];
  (* The energies are in the first element of the tuples. *)
  energyDiffFun = (Abs[#1[[1]]-#2[[1]]])&;
  (* match disregarding 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]];
  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];
  )
]"];

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, 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];
  );

ClassicalFit::usage="ClassicalFit[numE, expData, excludeDataIndices,
   problemVars, startValues, constraints] 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 a 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 option \"Experimental Uncertainty in K\" 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.
```

```
939 - \"expData\" which is the experimental data used for fitting.
940 - \"problemVars\" which are the symbols considered for fitting
941
942 - \"maxIterations\" which is the maximum number of iterations used by
      NMinimize.
943 - \"hamDim\" which is the dimension of the full Hamiltonian.
944 - \"allVars\" which are all the symbols defining the Hamiltonian
    under the aggregate simplifications.
945 - \"freeBies\" which are the free-ion parameters used to define the
    intermediate coupling basis.
946 - \"truncatedDim\" which is the dimension of the truncated
    Hamiltonian.
947 - \"compiledIntermediateFname\" the file name of the compiled
    function used for the truncated Hamiltonian.
948
949 - \"fittedLevels\" which is the number of levels fitted for.
950 - \"actualSteps\" the number of steps that FindMiniminum actually
    took.
951 - \"solWithUncertainty\" which is a list of replacement rules of the
    form (paramSymbol -> {bestEstimate, uncertainty}).
952 - \"rmsHistory\" which is a list of the \[Sigma] values found during
    the fitting.
953 - \"Appendix\" which is an association appended to the log file under
      the key \"Appendix\".
954 - \"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.
955
956 - \"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.
957 - \"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.
958 - \"degreesOfFreedom\" which is equal to the nunber of fitted state
    energies minus the number of parameters used in fitting.
959
960 The function admits the following options with corresponding default
    values:
961 - \"MaxHistory\" : determines how long the logs for the solver can be
    .
962 - \"MaxIterations\": determines the maximum number of iterations used
      by NMinimize.
963 - \"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\".
964 - \"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.
965
966 - \"AccuracyGoal\": the accuracy goal used by NMinimize, default of
    5.
967 - \"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.
968 - \"PrintFun\": the function used to print progress messages, the
    default is PrintTemporary.
969 - \"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\".
970
971 - \"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
     startValues.
- \"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,
  "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,
    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, solCompendium,
    openNotebooks, ordering, otherSimplifier, p0,
    paramBest, perHam,
    presentDataIndices, PrintFun, problemVarsPositions,
    problemVarsQ, problemVarsQString, problemVarsVec,
    problemVarsWithStartValues, reducedModelSymbols,
    roundedTruncationEnergy,
    runningInteractive, shiftToggle, simplifier,
    sol, solWithUncertainty,
    sortedTruncationIndex, sqdiff, standardValues,
    startTime,
    states, steps, symmetrySimplifier,
    theIntermediateEigensystems, TheIntermediateEigensystems,
    TheTruncatedAndSignedPathGenerator, timeTaken,
    truncatedIntermediateBasis, truncatedIntermediateHam,
    truncationEnergy, truncationIndices, RefParams,
    truncationUmbral, varHash,
    varsWithConstants, \[Lambda]0Vec,
    \[Lambda]exp
    },
  (
    \[Sigma]exp   = OptionValue["Energy Uncertainty in K"];
```

```
1099    solCompendium = <||>;
1100    refParamsVintage = OptionValue["RefParamsVintage"];
1101    RefParams = Which[
1102      refParamsVintage === "LaF3",
1103      LoadLaF3Parameters,
1104      refParamsVintage === "LiYF4",
1105      LoadLiYF4Parameters,
1106      True,
1107      refParamsVintage
1108    ];
1109    hamDim         = Binomial[14, numE];
1110    addShift       = OptionValue["AddConstantShift"];
1111    ln             = theLanthanides[[numE]];
1112    maxHistory     = OptionValue["MaxHistory"];
1113    maxIterations  = OptionValue["MaxIterations"];
1114    logFilePrefix  = If[OptionValue["FilePrefix"] == "",
1115                        ToString[theLanthanides[[numE]]],
1116                        OptionValue["FilePrefix"]
1117                      ];
1118    accuracyGoal  = OptionValue["AccuracyGoal"];
1119    PrintFun      = OptionValue["PrintFun"];
1120    freeIonSymbols       = OptionValue["FreeIonSymbols"];
1121    runningInteractive   = (Head[$ParentLink] === LinkObject);
1122    magneticSimplifier   = OptionValue["MagneticSimplifier"];
1123    magFieldSimplifier   = OptionValue["MagFieldSimplifier"];
1124    symmetrySimplifier   = OptionValue["SymmetrySimplifier"];
1125    otherSimplifier      = OptionValue["OtherSimplifier"];
1126    threeBodySimplifier  = If[Head[OptionValue["ThreeBodySimplifier"]]
       == Association,
1127                            OptionValue["ThreeBodySimplifier"][numE],
1128                            OptionValue["ThreeBodySimplifier"]
1129                          ];
1130
1131    truncationEnergy = If[OptionValue["TruncationEnergy"] ===
      Automatic,
1132      (
1133        PrintFun["Truncation energy set to Automatic, using the
      maximum energy (+20%) in the data ..."];
1134        Round[1.2 * Max[Select[First /@ expData, NumericQ[#] &]]]
1135      ),
1136      OptionValue["TruncationEnergy"]
1137      ];
1138    truncationEnergy = Max[50000, truncationEnergy];
1139    PrintFun["Using a truncation energy of ", truncationEnergy, " K"
      ];
1140
1141    simplifier = Join[magneticSimplifier,
1142                      magFieldSimplifier,
1143                      symmetrySimplifier,
1144                      threeBodySimplifier,
1145                      otherSimplifier];
1146
1147    PrintFun["Determining gaps in the data ..."];
1148    (* whatever is non-numeric is assumed as a known gap *)
1149    presentDataIndices = Flatten[Position[expData, {_?(NumericQ[#] &)
      , ___}]];
1150    (* some indices omitted here based on the excludeDataIndices
      argument *)
1151    presentDataIndices = Complement[presentDataIndices,
      excludeDataIndices];
1152
1153    solCompendium["simplifier"]          = simplifier;
1154    solCompendium["excludeDataIndices"] = excludeDataIndices;
1155    solCompendium["startValues"]         = startValues;
1156    solCompendium["freeIonSymbols"]      = freeIonSymbols;
1157    solCompendium["truncationEnergy"]    = truncationEnergy;
1158    solCompendium["numE"]                = numE;
1159    solCompendium["expData"]             = expData;
1160    solCompendium["problemVars"]         = problemVars;
1161    solCompendium["maxIterations"]       = maxIterations;
1162    solCompendium["hamDim"]              = hamDim;
1163    solCompendium["constraints"]         = constraints;
1164
1165    modelSymbols  = Sort[Select[paramSymbols, Not[MemberQ[Join[
      racahSymbols, juddOfeltIntensitySymbols, chenSymbols,{t2Switch, \[
      Epsilon], gs, nE}],#]]&]];
```

```
1166    (* remove the symbols that will be removed by the simplifier, no
        symbol should remain here that is not in the symbolic Hamiltonian
        *)
1167    reducedModelSymbols = Select[modelSymbols, Not[MemberQ[Keys[
        simplifier],#]]&];
1168
1169    (* this is useful to understand what are the arguments of the
        truncated compiled Hamiltonian *)
1170    If[OptionValue["SignatureCheck"],
1171      (
1172        PrintFun["Given the model parameters and the simplifying
        assumptions, the resultant model parameters are:"];
1173        PrintFun[{reducedModelSymbols}];
1174        PrintFun["Exiting ..."];
1175        Return[""];
1176      )
1177    ];
1178
1179    (* calculate the basis *)
1180    PrintFun["Retrieving the LSJMJ basis for f^", numE, " ..."];
1181    basis    = BasisLSJMJ[numE];
1182
1183    Which[refParamsVintage === Automatic,
1184      (
1185        PrintFun["Using the automatic vintage with freshly fitted
        free-ion parameters and others as in LaF3 ..."];
1186        lnParams      = LoadLaF3Parameters[ln];
1187        freeIonSol    = FreeIonSolver[expData, numE];
1188        freeIonParams = freeIonSol["bestParams"];
1189        lnParams      = Join[lnParams, freeIonParams];
1190      ),
1191      MemberQ[{List, Association}, Head[RefParams]],
1192      (
1193        RefParams = Association[RefParams];
1194        PrintFun["Using the given parameters as a starting point ..."
        ];
1195        lnParams    = RefParams;
1196        extraParams = LoadLaF3Parameters[ln];
1197        lnParams    = Join[extraParams, lnParams];
1198      ),
1199      True,
1200      (
1201        (* get the reference parameters from the given vintage *)
1202        PrintFun["Getting reference free-ion parameters for ", ln, "
        using ", refParamsVintage, " ..."];
1203        lnParams = ParamPad[RefParams[ln], "PrintFun" -> PrintFun];
1204      )
1205    ];
1206    freeBies = Prepend[Values[(#->(#/.lnParams)) &/@ freeIonSymbols],
          numE];
1207    (* a more explicit alias *)
1208    allVars            = reducedModelSymbols;
1209    numericConstraints = Association@Select[constraints, NumericQ
        [#[[2]]] &];
1210    standardValues     = allVars /. Join[lnParams, numericConstraints
        ];
1211    solCompendium["allVars"]  = allVars;
1212    solCompendium["freeBies"] = freeBies;
1213
1214    (* reload compiled version if found *)
1215    varHash                   = Hash[{numE, allVars, freeBies,
        truncationEnergy, simplifier}];
1216    compiledIntermediateFname = ln <> "-compiled-intermediate-
        truncated-ham-" <> ToString[varHash] <> ".mx";
1217    compiledIntermediateFname = FileNameJoin[{moduleDir, "compiled",
        compiledIntermediateFname}];
1218    solCompendium["compiledIntermediateFname"] =
        compiledIntermediateFname;
1219
1220    If[FileExistsQ[compiledIntermediateFname],
1221      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 ..."];
1222      PrintFun["Using : ", compiledIntermediateFname];
1223      {compileIntermediateTruncatedHam, truncatedIntermediateBasis} =
```

```
 Import[compiledIntermediateFname];,
(
  If[truncationEnergy == Infinity,
  (
      ham = HamMatrixAssembly[numE, "ReturnInBlocks" -> False];
      theSimplifier = simplifier;
      ham = Normal@ReplaceInSparseArray[ham, simplifier];
      PrintFun["Compiling a function for the Hamiltonian with no truncation ..."];
      (* 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[ham]];
      truncatedIntermediateBasis = SparseArray@IdentityMatrix[Binomial[14, numE]];
      (* save the compiled function *)
      PrintFun["Saving the compiled function for the Hamiltonian with no truncation and a placeholder intermediate basis ..."];
      Export[compiledIntermediateFname, {compileIntermediateTruncatedHam, truncatedIntermediateBasis}];
  ),
  (
    (* grab the Hamiltonian preserving the 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 aggregate set of simplification rules ..."];
    ham           = Map[ReplaceInSparseArray[#, simplifier]&, ham, {2}];
    PrintFun["Zeroing out every symbol in the Hamiltonian that is not a free-ion parameter ..."];
    (* Get the free ion symbols *)
    freeIonSimplifier = (#->0) & /@ Complement[reducedModelSymbols, freeIonSymbols];
    (* Take the diagonal blocks for the intermediate analysis *)
    PrintFun["Grabbing the diagonal blocks of the Hamiltonian ..."];
    diagonalBlocks       = Diagonal[ham];
    (* simplify them to only keep the free ion symbols *)
    PrintFun["Simplifying the diagonal blocks to only keep the free ion symbols ..."];
    diagonalScalarBlocks = ReplaceInSparseArray[#, freeIonSimplifier]&/@diagonalBlocks;
    (* these include the MJ quantum numbers, remove that *)
    PrintFun["Contracting the basis vectors by removing the MJ quantum numbers from the diagonal blocks ..."];
    diagonalScalarBlocks = FreeHam[diagonalScalarBlocks, numE];

    argsOfTheIntermediateEigensystems         = StringJoin[Riffle[Prepend[(ToString[#]<>"v_") & /@ freeIonSymbols,"numE_"],", "]];
    argsForEvalInsideOfTheIntermediateSystems = StringJoin[Riffle[(ToString[#]<>"v") & /@ freeIonSymbols,", "]];
    PrintFun["argsOfTheIntermediateEigensystems = ", argsOfTheIntermediateEigensystems];
    PrintFun["argsForEvalInsideOfTheIntermediateSystems = ", argsForEvalInsideOfTheIntermediateSystems];
    PrintFun["(if the following fails, it might help to see if the arguments of TheIntermediateEigensystems match the ones shown above)"];

    (* 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 *)
    (* compile one function for each of the blocks *)
    PrintFun["Compiling functions for the diagonal blocks of the Hamiltonian ..."];
    compiledDiagonal = Compile[Evaluate[freeIonSymbols], Evaluate[N[Normal[#]]]]&/@diagonalScalarBlocks;
    (* use that to create a function that will calculate the free-ion eigensystem *)
    TheIntermediateEigensystems[numEv_, F0v_, F2v_, F4v_, F6v_, ζv_] := (
```

```
1270        theNumericBlocks = (#[F0v, F2v, F4v, F6v, ζv]&) /@
    compiledDiagonal;
1271        theIntermediateEigensystems = Eigensystem /@
    theNumericBlocks;
1272        Js      = AllowedJ[numEv];
1273        basisJ = BasisLSJMJ[numEv,"AsAssociation"->True];
1274        (* having calculated the eigensystems with the removed
    degeneracies, put the degeneracies back in explicitly *)
1275        elevatedIntermediateEigensystems = MapIndexed[EigenLever
    [#1, 2Js[[#2[[1]]]]+1 ]&, theIntermediateEigensystems];
1276        (* Identify a single MJ to keep *)
1277        pivot          = If[EvenQ[numEv],0,-1/2];
1278        LSJmultiplets = (#[[1]]<>ToString[InputForm[#[[2]]]])&/
    @Select[BasisLSJMJ[numEv],#[[-1]]== pivot &];
1279        (* calculate the multiplet assignments that the
    intermediate basis eigenvectors have *)
1280        needlePosition = 0;
1281        multipletAssignments = Table[
1282          (
1283            J          = Js[[idx]];
1284            eigenVecs = theIntermediateEigensystems[[idx]][[2]];
1285            majorComponentIndices         = Ordering[Abs[#]][[-1]]&/
    @eigenVecs;
1286            majorComponentIndices        += needlePosition;
1287            needlePosition               += Length[
    majorComponentIndices];
1288            majorComponentAssignments     = LSJmultiplets[[#]]&/
    @majorComponentIndices;
1289            (* All of the degenerate eigenvectors belong to the
    same multiplet*)
1290            elevatedMultipletAssignments = ListRepeater[
    majorComponentAssignments, 2J+1];
1291            elevatedMultipletAssignments
1292          ),
1293        {idx, 1, Length[Js]}
1294        ];
1295        (* put together the multiplet assignments and the energies
    *)
1296        freeIenergiesAndMultiplets = Transpose/@Transpose[{First/
    @elevatedIntermediateEigensystems, multipletAssignments}];
1297        freeIenergiesAndMultiplets = Flatten[
    freeIenergiesAndMultiplets, 1];
1298        (* calculate the change of basis matrix using the
    intermediate coupling eigenvectors *)
1299        basisChanger = BlockDiagonalMatrix[Transpose/@Last/
    @elevatedIntermediateEigensystems];
1300        basisChanger = SparseArray[basisChanger];
1301        Return[{theIntermediateEigensystems, multipletAssignments,
    elevatedIntermediateEigensystems, freeIenergiesAndMultiplets,
    basisChanger}]
1302      );
1303
1304      PrintFun["Calculating the intermediate eigensystems for ",ln,
    " using free-ion params from LaF3 ..."];
1305      (* calculate intermediate coupling basis using the free-ion
    params for LaF3 *)
1306      {theIntermediateEigensystems, multipletAssignments,
    elevatedIntermediateEigensystems, freeIenergiesAndMultiplets,
    basisChanger} = TheIntermediateEigensystems@@freeBies;
1307
1308      (* use that intermediate coupling basis to compile a function
     for the full Hamiltonian *)
1309      allFreeEnergies = Flatten[First/
    @elevatedIntermediateEigensystems];
1310      (* important that the intermediate coupling basis have
    attached energies, which make possible the truncation *)
1311      ordering = Ordering[allFreeEnergies];
1312      (* sort the free ion energies and determine which indices
    should be included in the truncation *)
1313      allFreeEnergiesSorted          = Sort[allFreeEnergies];
1314      {minFreeEnergy, maxFreeEnergy} = MinMax[allFreeEnergies];
1315      (* determine the index at which the energy is equal or larger
     than the truncation energy *)
1316      sortedTruncationIndex = Which[
1317        truncationEnergy > (maxFreeEnergy-minFreeEnergy),
1318        hamDim,
```

```
1319            True,
1320            FirstPosition[allFreeEnergiesSorted - Min[
    allFreeEnergiesSorted],x_/;x>truncationEnergy,{0},1][[1]]
1321        ];
1322        (* the actual energy at which the truncation is made *)
1323        roundedTruncationEnergy = allFreeEnergiesSorted[[
    sortedTruncationIndex]];
1324
1325        (* the indices that participate in the truncation *)
1326        truncationIndices = ordering[[;;sortedTruncationIndex]];
1327        PrintFun["Computing the block structure of the change of
    basis array ..."];
1328        blockSizes          = BlockArrayDimensionsArray[ham];
1329        basisChangerBlocks = ArrayBlocker[basisChanger, blockSizes];
1330        blockShifts         = First /@ Diagonal[blockSizes];
1331        numBlocks           = Length[blockSizes];
1332        (* using the ham (with all the symbols) change the basis to
    the computed one *)
1333        PrintFun["Changing the basis of the Hamiltonian to the
    intermediate coupling basis ..."];
1334        intermediateHam = BlockMatrixMultiply[ham, basisChangerBlocks
    ];
1335        PrintFun["Distributing products inside of symbolic matrix
    elements to keep complexity in check ..."];
1336        Do[
1337          intermediateHam[[rowIdx, colIdx]] = MapToSparseArray[
    intermediateHam[[rowIdx, colIdx]], Distribute /@ # &];,
1338          {rowIdx, 1, numBlocks},
1339          {colIdx, 1, numBlocks}
1340        ];
1341        intermediateHam = BlockMatrixMultiply[BlockTranspose[
    basisChangerBlocks],intermediateHam];
1342        PrintFun["Distributing products inside of symbolic matrix
    elements to keep complexity in check ..."];
1343        Do[
1344          intermediateHam[[rowIdx, colIdx]] = MapToSparseArray[
    intermediateHam[[rowIdx, colIdx]], Distribute /@ # &];,
1345          {rowIdx, 1, numBlocks},
1346          {colIdx, 1, numBlocks}
1347        ];
1348        (* using the truncation indices truncate that one *)
1349        PrintFun["Truncating the Hamiltonian ..."];
1350        truncatedIntermediateHam = TruncateBlockArray[intermediateHam
    , truncationIndices, blockShifts];
1351        (* these are the basis vectors for the truncated hamiltonian
    *)
1352        PrintFun["Saving the truncated intermediate basis ..."];
1353        truncatedIntermediateBasis = basisChanger[[All,
    truncationIndices]];
1354
1355        PrintFun["Compiling a function for the truncated Hamiltonian
    ..."];
1356        (* compile a function that will calculate the truncated
    Hamiltonian given the parameters in allVars, this is the function
    to be use in fitting *)
1357        compileIntermediateTruncatedHam = Compile[Evaluate[allVars],
    Evaluate[truncatedIntermediateHam]];
1358        (* save the compiled function *)
1359        PrintFun["Saving the compiled function for the truncated
    Hamiltonian and the truncated intermediate basis ..."];
1360        Export[compiledIntermediateFname, {
    compileIntermediateTruncatedHam, truncatedIntermediateBasis}];
1361      )
1362    ];
1363    )
1364    ];
1365
1366    truncationUmbral = Dimensions[truncatedIntermediateBasis][[2]];
1367    PrintFun["The truncated Hamiltonian has a dimension of ",
    truncationUmbral, "x", truncationUmbral, " ..."];
1368    presentDataIndices = Select[presentDataIndices, # <=
    truncationUmbral &];
1369    solCompendium["presentDataIndices"] = presentDataIndices;
1370
1371    (* the problemVars are the symbols that will be fitted for *)
1372
```

```
1373    PrintFun["Starting up the fitting process using the Levenberg-
        Marquardt method ..."];
1374    (* using the problemVars I need to create the argument list
        including _?NumericQ *)
1375    problemVarsQ       = (ToString[#] <> "_?NumericQ") & /@
        problemVars;
1376    problemVarsQString = StringJoin[Riffle[problemVarsQ, ", "]];
1377    (* we also need to have the padded arguments with the variables
        in the right position and the fixed values in the remaining ones
        *)
1378    problemVarsPositions = Position[allVars, #][[1, 1]] & /@
        problemVars;
1379    problemVarsString    = StringJoin[Riffle[ToString /@ problemVars,
         ", "]];
1380    (* 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 *)
1381    varsWithConstants                          = standardValues;
1382    varsWithConstants[[problemVarsPositions]] = problemVars;
1383    varsWithConstantsString                    = ToString[
        varsWithConstants];
1384
1385    (* this following function serves eigenvalues from the
        Hamiltonian, has memoization so it might grow to use a lot of RAM
        *)
1386    Clear[HamSortedEigenvalues];
1387    hamEigenvaluesTemplate = StringTemplate["
1388    HamSortedEigenvalues[`problemVarsQ`]:=(
1389      ham          = compileIntermediateTruncatedHam@@`
        varsWithConstants`;
1390      eigenValues = Chop[Sort@Eigenvalues@ham];
1391      eigenValues = eigenValues - Min[eigenValues];
1392      HamSortedEigenvalues[`problemVarsString`] = eigenValues;
1393      Return[eigenValues]
1394    )"];
1395    hamString = hamEigenvaluesTemplate[<|
1396        "problemVarsQ"      -> problemVarsQString,
1397        "varsWithConstants" -> varsWithConstantsString,
1398        "problemVarsString" -> problemVarsString
1399        |>];
1400    ToExpression[hamString];
1401
1402    (* 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 *)
1403    eigenvalueDispenserTemplate = StringTemplate["
1404    PartialHamEigenvalues[`problemVarsQ`][i_]:=(
1405      eigenVals = HamSortedEigenvalues[`problemVarsString`];
1406      eigenVals[[i]]
1407    )
1408    "];
1409    eigenValueDispenserString = eigenvalueDispenserTemplate[<|
1410        "problemVarsQ"      -> problemVarsQString,
1411        "problemVarsString" -> problemVarsString
1412        |>];
1413    ToExpression[eigenValueDispenserString];
1414
1415    PrintFun["Determining the free variables after constraints ..."];
1416    constrainedProblemVars     = (problemVars /. constraints);
1417    constrainedProblemVarsList = Variables[constrainedProblemVars];
1418    If[addShift,
1419      PrintFun["Adding a constant shift to the fitting parameters ...
        "];
1420      constrainedProblemVarsList = Append[constrainedProblemVarsList,
         \[Epsilon]]
1421    ];
1422
1423    indepVars = Complement[problemVars, #[[1]] & /@ constraints];
1424    stringPartialVars = ToString/@constrainedProblemVarsList;
1425
1426    paramSols  = {};
1427    rmsHistory = {};
1428    steps      = 0;
1429    problemVarsWithStartValues = KeyValueMap[{#1,#2} &, startValues];
1430    If[addShift,
1431      problemVarsWithStartValues = Append[problemVarsWithStartValues,
```

```
    {\[Epsilon] ,0}];
];
openNotebooks     = If[runningInteractive,
          ("WindowTitle"/.NotebookInformation[#]) & /@ Notebooks[],
          {}];
If[Not[MemberQ[openNotebooks,"Solver Progress"]] && OptionValue["ProgressView"],
  ProgressNotebook["Basic"->False]
];
degressOfFreedom = Length[presentDataIndices] - Length[problemVars] - 1;
PrintFun["Fitting for ", Length[presentDataIndices], " data points with ", Length[problemVars], " free parameters.", " The effective degrees of freedom are ", degressOfFreedom, " ..."];

PrintFun["Fitting model to data ..."];
startTime   = Now;
shiftToggle = If[addShift, 1, 0];
sol = FindMinimum[
  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"];
1560    solCompendium["timeTaken/s"]          = timeTaken;
1561    solCompendium["bestParams"]           = bestParams;
1562    solCompendium["bestParamsWithConstraints"] = bestWithConstraints;
1563
1564    If[OptionValue["SaveEigenvectors"],
1565        solCompendium["states"] = {#[[1]] + \[Epsilon]Best, #[[2]]}
    &/@ (Chop /@ ShiftedLevels[states]),
1566        (
1567          finalEnergies = Sort[First /@ states];
1568          finalEnergies = finalEnergies - finalEnergies[[1]];
1569          finalEnergies = finalEnergies + \[Epsilon]Best;
1570          finalEnergies  = Chop /@ finalEnergies;
1571          solCompendium["energies"] = finalEnergies;
1572      )
1573    ];
1574    If[OptionValue["SaveToLog"],
1575      PrintFun["Saving the solution to the log file ..."];
1576      LogSol[solCompendium, logFilePrefix];
1577    ];
1578    PrintFun["Finished ..."];
1579    Return[solCompendium];
1580  )
1581 ];
1582
1583
1584 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.
1585
1586 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.
1587
1588 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.
1589
1590 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\"
    .
1591
1592 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.
1593
1594 The list startValues is a list with all of the parameters needed to
    define the Hamiltonian (including the initial values for
    problemVars).
1595
1596 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.
1597
1598 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.
1599
1600 The function returns an association with the following keys:
1601
1602 - \"bestRMS\" which is the best \[Sigma] value found.
1603 - \"bestParams\" which is the best set of parameters found for the
    variables that were not constrained.
1604 - \"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.
1605 - \"paramSols\" which is a list of the parameters trajectories during
     the stepping of the fitting algorithm.
1606 - \"timeTaken/s\" which is the time taken to find the best fit.
1607 - \"simplifier\" which is the simplifier used to simplify the
    Hamiltonian.
1608 - \"excludeDataIndices\" as given in the input.
1609 - \"startValues\" as given in the input.
1610
1611 - \"freeIonSymbols\" which are the symbols used in the intermediate
    coupling basis.
1612 - \"truncationEnergy\" which is the energy used to truncate the
    Hamiltonian, if it was set to Automatic, the value here is the
    actual energy used.
1613 - \"numE\" which is the number of electrons in the f^numE
    configuration.
1614 - \"expData\" which is the experimental data used for fitting.
1615 - \"problemVars\" which are the symbols considered for fitting
1616
1617 - \"maxIterations\" which is the maximum number of iterations used by
     NMinimize.
1618 - \"hamDim\" which is the dimension of the full Hamiltonian.
1619 - \"allVars\" which are all the symbols defining the Hamiltonian
    under the aggregate simplifications.
1620 - \"freeBies\" which are the free-ion parameters used to define the
    intermediate coupling basis.
1621 - \"truncatedDim\" which is the dimension of the truncated
    Hamiltonian.
1622 - \"compiledIntermediateFname\" the file name of the compiled
    function used for the truncated Hamiltonian.
1623
1624 - \"fittedLevels\" which is the number of levels fitted for.
1625 - \"actualSteps\" the number of steps that FindMiniminum actually
    took.
1626 - \"solWithUncertainty\" which is a list of replacement rules of the
    form (paramSymbol -> {bestEstimate, uncertainty}).
1627 - \"rmsHistory\" which is a list of the \[Sigma] values found during
    the fitting.
1628 - \"Appendix\" which is an association appended to the log file under
     the key \"Appendix\".
1629 - \"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.
1630
1631 - \"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.
1632 - \"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.
1633 - \"degreesOfFreedom\" which is equal to the nunber of fitted state
    energies minus the number of parameters used in fitting.
1634
1635 The function admits the following options with corresponding default
    values:
1636 - \"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.

- \"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[MostlyOrthogonalFit] = {
  "MaxHistory"       -> 200,
  "MaxIterations"    -> 100,
  "FilePrefix"       -> "calcs",
  "ProgressView"     -> True,
  "TruncationEnergy" -> Automatic,
  "AccuracyGoal"     -> 5,
  "PrintFun"         -> PrintTemporary,
  "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" -> {
    E0p->0,
    P0->0,
    \[Sigma]SS->0,
    T11p->0,
    T12->0,
    T14->0,
    T15->0,
    T16->0,
    T18->0,
    T17->0,
    T19->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  -> {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,
```

```
1967        slaterSymbols,
1968        {α, β, γ},
1969        {T2},
1970        chenSymbols,
1971        {t2Switch, \[Epsilon], gs, nE}],#]]&
1972      ];
1973      modelSymbols = Sort[modelSymbols];
1974      (* remove the symbols that will be removed by the simplifier, no
          symbol should remain here that is not in the symbolic Hamiltonian
          *)
1975      reducedModelSymbols = Select[modelSymbols, Not[MemberQ[Keys[
          simplifier],#]]&];
1976
1977      (* this is useful to understand what are the arguments of the
          truncated compiled Hamiltonian *)
1978      If[OptionValue["SignatureCheck"],
1979        (
1980          PrintFun["Given the model parameters and the simplifying
          assumptions, the resultant model parameters are:"];
1981          PrintFun[{reducedModelSymbols}];
1982          PrintFun["Exiting ..."];
1983          Return[""];
1984        )
1985      ];
1986
1987      (* calculate the basis *)
1988      PrintFun["Retrieving the LSJMJ basis for f^", numE, " ..."];
1989      basis    = BasisLSJMJ[numE];
1990
1991      Which[refParamsVintage === Automatic,
1992        (
1993          PrintFun["This is not currently supported, please provide a
          vintage for the reference parameters."];
1994          Return[$Failed];
1995        ),
1996        MemberQ[{List, Association}, Head[RefParams]],
1997        (
1998          RefParams = Association[RefParams];
1999          PrintFun["Using the given parameters as a starting point ..."
          ];
2000          lnParams    = RefParams;
2001          extraParams = FromNonOrthogonalToMostlyOrthogonal[
          LoadLaF3Parameters[ln], numE];
2002          lnParams    = Join[extraParams, lnParams];
2003        ),
2004        True,
2005        (
2006          (* get the reference parameters from the given vintage *)
2007          PrintFun["Getting reference free-ion parameters for ", ln, "
          using ", refParamsVintage, " ..."];
2008          lnParams = ParamPad[FromNonOrthogonalToMostlyOrthogonal[
          RefParams[ln], numE],
2009            "PrintFun" -> PrintFun];
2010        )
2011      ];
2012
2013      freeBies = Prepend[Values[(# -> (# /. lnParams)) &/@
          freeIonSymbols],  numE];
2014
2015      (* a more explicit alias *)
2016      allVars            = reducedModelSymbols;
2017      numericConstraints = Association@Select[constraints, NumericQ
          [#[[2]]] &];
2018      standardValues     = allVars /. Join[lnParams, numericConstraints
          ];
2019      solCompendium["allVars"]  = allVars;
2020      solCompendium["freeBies"] = freeBies;
2021
2022      (* reload compiled version if found *)
2023      varHash                   = Hash[{numE, allVars, freeBies,
          truncationEnergy, simplifier}];
2024      compiledIntermediateFname = ln <> "-compiled-intermediate-
          truncated-ham-" <> ToString[varHash] <> ".mx";
2025      compiledIntermediateFname = FileNameJoin[{moduleDir, "compiled",
          compiledIntermediateFname}];
2026      solCompendium["compiledIntermediateFname"] =
```

```
    compiledIntermediateFname;

    If[FileExistsQ[compiledIntermediateFname],
    (
      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 ..."];
      PrintFun["Using : ", compiledIntermediateFname];
      {compileIntermediateTruncatedHam, truncatedIntermediateBasis} = Import[compiledIntermediateFname];
    ),
    (
      If[truncationEnergy == Infinity,
      (
          ham = HamMatrixAssembly[numE,
                "ReturnInBlocks" -> False,
                "OperatorBasis" -> "MostlyOrthogonal"];
          theSimplifier = simplifier;
          ham = Normal @ ReplaceInSparseArray[ham, simplifier];
          PrintFun["Compiling a function for the Hamiltonian with no truncation ..."];
          (* 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[ham]];
          truncatedIntermediateBasis      = SparseArray@IdentityMatrix[Binomial[14, numE]];
          (* save the compiled function *)
          PrintFun["Saving the compiled function for the Hamiltonian with no truncation and a placeholder intermediate basis ..."];
          Export[compiledIntermediateFname, {compileIntermediateTruncatedHam, truncatedIntermediateBasis}];
      ),
      (
        (* grab the Hamiltonian preserving the block structure *)
        PrintFun["Assembling the Hamiltonian for f^", numE, " keeping the block structure ..."];
        ham           = HamMatrixAssembly[numE,
                          "ReturnInBlocks"->True,
                          "OperatorBasis"->"MostlyOrthogonal"];
        (* apply the simplifier *)
        PrintFun["Simplifying using the aggregate set of simplification rules ..."];
        ham           = Map[ReplaceInSparseArray[#, simplifier]&, ham, {2}];
        PrintFun["Zeroing out every symbol in the Hamiltonian that is not a free-ion parameter ..."];

        (* Get the free ion symbols *)
        freeIonSimplifier = (#->0) & /@ Complement[reducedModelSymbols, freeIonSymbols];

        (* Take the diagonal blocks for the intermediate analysis *)
        PrintFun["Grabbing the diagonal blocks of the Hamiltonian ..."];
        diagonalBlocks       = Diagonal[ham];

        (* simplify them to only keep the free ion symbols *)
        PrintFun["Simplifying the diagonal blocks to only keep the free ion symbols ..."];
        diagonalScalarBlocks = ReplaceInSparseArray[#, freeIonSimplifier] &/@ diagonalBlocks;

        (* these include the MJ quantum numbers, remove that *)
        PrintFun["Contracting the basis vectors by removing the MJ quantum numbers from the diagonal blocks ..."];
        diagonalScalarBlocks = FreeHam[diagonalScalarBlocks, numE];

        argsOfTheIntermediateEigensystems         = StringJoin[Riffle[Prepend[(ToString[#]<>"v_") & /@ freeIonSymbols,"numE_"],", "]];
        argsForEvalInsideOfTheIntermediateSystems = StringJoin[Riffle[(ToString[#]<>"v") & /@ freeIonSymbols,", "]];
        PrintFun["argsOfTheIntermediateEigensystems = ", argsOfTheIntermediateEigensystems];
```

```
2079        PrintFun["argsForEvalInsideOfTheIntermediateSystems = ",
    argsForEvalInsideOfTheIntermediateSystems];
2080        PrintFun["(if the following fails, it might help to see if
    the arguments of TheIntermediateEigensystems match the ones shown
    above)"];
2081
2082        (* 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 *)
2083        (* compile one function for each of the blocks *)
2084        PrintFun["Compiling functions for the diagonal blocks of the
    Hamiltonian ..."];
2085        compiledDiagonal = Compile[Evaluate[freeIonSymbols], Evaluate
    [N[Normal[#]]]]& /@ diagonalScalarBlocks;
2086        (* use that to create a function that will calculate the free
    -ion eigensystem *)
2087        TheIntermediateEigensystems[numEv_, E0pv_, E1pv_, E2pv_,
    E3pv_, ζv_] := (
2088          theNumericBlocks = (#[E0pv, E1pv, E2pv, E3pv, ζv]&) /@
    compiledDiagonal;
2089          theIntermediateEigensystems = Eigensystem /@
    theNumericBlocks;
2090          Js      = AllowedJ[numEv];
2091          basisJ = BasisLSJMJ[numEv, "AsAssociation"->True];
2092          (* having calculated the eigensystems with the removed
    degeneracies, put the degeneracies back in explicitly *)
2093          elevatedIntermediateEigensystems = MapIndexed[EigenLever
    [#1, 2Js[[#2[[1]]]]+1 ]&, theIntermediateEigensystems];
2094          (* Identify a single MJ to keep *)
2095          pivot          = If[EvenQ[numEv],0,-1/2];
2096          LSJmultiplets = (#[[1]]<>ToString[InputForm[#[[2]]]])&/
    @Select[BasisLSJMJ[numEv],#[[-1]]== pivot &];
2097          (* calculate the multiplet assignments that the
    intermediate basis eigenvectors have *)
2098          needlePosition = 0;
2099          multipletAssignments = Table[
2100            (
2101              J          = Js[[idx]];
2102              eigenVecs = theIntermediateEigensystems[[idx]][[2]];
2103              majorComponentIndices         = Ordering[Abs[#]][[-1]]&/
    @eigenVecs;
2104              majorComponentIndices        += needlePosition;
2105              needlePosition               += Length[
    majorComponentIndices];
2106              majorComponentAssignments     = LSJmultiplets[[#]]&/
    @majorComponentIndices;
2107              (* All of the degenerate eigenvectors belong to the
    same multiplet*)
2108              elevatedMultipletAssignments = ListRepeater[
    majorComponentAssignments, 2J+1];
2109              elevatedMultipletAssignments
2110            ),
2111          {idx, 1, Length[Js]}
2112          ];
2113
2114          (* put together the multiplet assignments and the energies
    *)
2115          freeIenergiesAndMultiplets = Transpose/@Transpose[{First/
    @elevatedIntermediateEigensystems, multipletAssignments}];
2116          freeIenergiesAndMultiplets = Flatten[
    freeIenergiesAndMultiplets, 1];
2117          (* calculate the change of basis matrix using the
    intermediate coupling eigenvectors *)
2118          basisChanger = BlockDiagonalMatrix[Transpose/@Last/
    @elevatedIntermediateEigensystems];
2119          basisChanger = SparseArray[basisChanger];
2120          Return[{theIntermediateEigensystems,
2121            multipletAssignments,
2122            elevatedIntermediateEigensystems,
2123            freeIenergiesAndMultiplets,
2124            basisChanger}]
2125        );
2126
2127        PrintFun["Calculating the intermediate eigensystems for ",ln,
    " using free-ion params from LaF3 ..."];
2128        (* calculate intermediate coupling basis using the free-ion
```

```
    params for LaF3 *)
2129        {theIntermediateEigensystems, multipletAssignments,
    elevatedIntermediateEigensystems, freeIenergiesAndMultiplets,
    basisChanger} = TheIntermediateEigensystems@@freeBies;
2130
2131        (* use that intermediate coupling basis to compile a function
     for the full Hamiltonian *)
2132        allFreeEnergies = Flatten[First/
    @elevatedIntermediateEigensystems];
2133        (* important that the intermediate coupling basis have
    attached energies, which make possible the truncation *)
2134        ordering = Ordering[allFreeEnergies];
2135        (* sort the free ion energies and determine which indices
    should be included in the truncation *)
2136        allFreeEnergiesSorted          = Sort[allFreeEnergies];
2137        {minFreeEnergy, maxFreeEnergy} = MinMax[allFreeEnergies];
2138        (* determine the index at which the energy is equal or larger
     than the truncation energy *)
2139        sortedTruncationIndex = Which[
2140          truncationEnergy > (maxFreeEnergy-minFreeEnergy),
2141          hamDim,
2142          True,
2143          FirstPosition[allFreeEnergiesSorted - Min[
    allFreeEnergiesSorted],x_/;x>truncationEnergy,{0},1][[1]]
2144        ];
2145        (* the actual energy at which the truncation is made *)
2146        roundedTruncationEnergy = allFreeEnergiesSorted[[
    sortedTruncationIndex]];
2147
2148        (* the indices that participate in the truncation *)
2149        truncationIndices = ordering[[;;sortedTruncationIndex]];
2150        PrintFun["Computing the block structure of the change of
    basis array ..."];
2151        blockSizes         = BlockArrayDimensionsArray[ham];
2152        basisChangerBlocks = ArrayBlocker[basisChanger, blockSizes];
2153        blockShifts        = First /@ Diagonal[blockSizes];
2154        numBlocks          = Length[blockSizes];
2155
2156        (* using the ham (with all the symbols) change the basis to
    the computed one *)
2157        PrintFun["Changing the basis of the Hamiltonian to the
    intermediate coupling basis ..."];
2158        intermediateHam = BlockMatrixMultiply[ham, basisChangerBlocks
    ];
2159        PrintFun["Distributing products inside of symbolic matrix
    elements to keep complexity in check ..."];
2160        Do[
2161          intermediateHam[[rowIdx, colIdx]] = MapToSparseArray[
    intermediateHam[[rowIdx, colIdx]], Distribute /@ # &];,
2162          {rowIdx, 1, numBlocks},
2163          {colIdx, 1, numBlocks}
2164        ];
2165        intermediateHam = BlockMatrixMultiply[BlockTranspose[
    basisChangerBlocks],intermediateHam];
2166        PrintFun["Distributing products inside of symbolic matrix
    elements to keep complexity in check ..."];
2167        Do[
2168          intermediateHam[[rowIdx, colIdx]] = MapToSparseArray[
    intermediateHam[[rowIdx, colIdx]], Distribute /@ # &];,
2169          {rowIdx, 1, numBlocks},
2170          {colIdx, 1, numBlocks}
2171        ];
2172        (* using the truncation indices truncate that one *)
2173        PrintFun["Truncating the Hamiltonian ..."];
2174        truncatedIntermediateHam = TruncateBlockArray[intermediateHam
    , truncationIndices, blockShifts];
2175        (* these are the basis vectors for the truncated hamiltonian
    *)
2176        PrintFun["Saving the truncated intermediate basis ..."];
2177        truncatedIntermediateBasis = basisChanger[[All,
    truncationIndices]];
2178
2179        PrintFun["Compiling a function for the truncated Hamiltonian
    ..."];
2180        (* compile a function that will calculate the truncated
    Hamiltonian given the parameters in allVars, this is the function
```

```
    to be use in fitting *)
2181        compileIntermediateTruncatedHam = Compile[Evaluate[allVars],
    Evaluate[truncatedIntermediateHam]];
2182        (* save the compiled function *)
2183        PrintFun["Saving the compiled function for the truncated
    Hamiltonian and the truncated intermediate basis ..."];
2184        Export[compiledIntermediateFname, {
    compileIntermediateTruncatedHam, truncatedIntermediateBasis}];
2185      )
2186    ];
2187    )
2188    ];
2189
2190    truncationUmbral = Dimensions[truncatedIntermediateBasis][[2]];
2191    PrintFun["The truncated Hamiltonian has a dimension of ",
    truncationUmbral, "x", truncationUmbral, " ..."];
2192    presentDataIndices = Select[presentDataIndices, # <=
    truncationUmbral &];
2193    solCompendium["presentDataIndices"] = presentDataIndices;
2194
2195    (* the problemVars are the symbols that will be fitted for *)
2196
2197    PrintFun["Starting up the fitting process using the Levenberg-
    Marquardt method ..."];
2198    (* using the problemVars I need to create the argument list
    including _?NumericQ *)
2199    problemVarsQ       = (ToString[#] <> "_?NumericQ") & /@
    problemVars;
2200    problemVarsQString = StringJoin[Riffle[problemVarsQ, ", "]];
2201    (* we also need to have the padded arguments with the variables
    in the right position and the fixed values in the remaining ones
    *)
2202    problemVarsPositions = Position[allVars, #][[1, 1]] & /@
    problemVars;
2203    problemVarsString    = StringJoin[Riffle[ToString /@ problemVars,
     ", "]];
2204    (* 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 *)
2205    varsWithConstants                          = standardValues;
2206    varsWithConstants[[problemVarsPositions]] = problemVars;
2207    varsWithConstantsString                    = ToString[
    varsWithConstants];
2208
2209    (* this following function serves eigenvalues from the
    Hamiltonian, has memoization so it might grow to use a lot of RAM
    *)
2210    Clear[HamSortedEigenvalues];
2211    hamEigenvaluesTemplate = StringTemplate["
2212    HamSortedEigenvalues[`problemVarsQ`]:=(
2213      ham          = compileIntermediateTruncatedHam@@`
    varsWithConstants`;
2214      eigenValues = Chop[Sort@Eigenvalues@ham];
2215      eigenValues = eigenValues - Min[eigenValues];
2216      HamSortedEigenvalues[`problemVarsString`] = eigenValues;
2217      Return[eigenValues]
2218    )"];
2219    hamString = hamEigenvaluesTemplate[<|
2220        "problemVarsQ"      -> problemVarsQString,
2221        "varsWithConstants" -> varsWithConstantsString,
2222        "problemVarsString" -> problemVarsString
2223        |>];
2224    ToExpression[hamString];
2225
2226    (* 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 *)
2227    eigenvalueDispenserTemplate = StringTemplate["
2228    PartialHamEigenvalues[`problemVarsQ`][i_]:=(
2229      eigenVals = HamSortedEigenvalues[`problemVarsString`];
2230      eigenVals[[i]]
2231    )
2232    "];
2233    eigenValueDispenserString = eigenvalueDispenserTemplate[<|
2234        "problemVarsQ"      -> problemVarsQString,
2235        "problemVarsString" -> problemVarsString
```

```
2236        |>];
2237    ToExpression[eigenValueDispenserString];
2238
2239    PrintFun["Determining the free variables after constraints ..."];
2240    constrainedProblemVars     = (problemVars /. constraints);
2241    constrainedProblemVarsList = Variables[constrainedProblemVars];
2242    If[addShift,
2243      PrintFun["Adding a constant shift to the fitting parameters ..."];
2244      constrainedProblemVarsList = Append[constrainedProblemVarsList, \[Epsilon]]
2245    ];
2246
2247    indepVars = Complement[problemVars, #[[1]] & /@ constraints];
2248    stringPartialVars = ToString/@constrainedProblemVarsList;
2249
2250    paramSols  = {};
2251    rmsHistory = {};
2252    steps      = 0;
2253    problemVarsWithStartValues = KeyValueMap[{#1,#2} &, startValues];
2254    If[addShift,
2255      problemVarsWithStartValues = Append[problemVarsWithStartValues, {\[Epsilon] ,0}];
2256    ];
2257    openNotebooks     = If[runningInteractive,
2258              ("WindowTitle"/.NotebookInformation[#]) & /@ Notebooks[],
2259              {}];
2260    If[Not[MemberQ[openNotebooks,"Solver Progress"]] && OptionValue["ProgressView"],
2261      ProgressNotebook["Basic"->False]
2262    ];
2263    degressOfFreedom = Length[presentDataIndices] - Length[problemVars] - 1;
2264    PrintFun["Fitting for ", Length[presentDataIndices], " data points with ", Length[problemVars], " free parameters.", " The effective degrees of freedom are ", degressOfFreedom, " ..."];
2265
2266    PrintFun["Fitting model to data ..."];
2267    startTime   = Now;
2268    shiftToggle = If[addShift, 1, 0];
2269    sol = FindMinimum[
2270      Sum[(expData[[j]][[1]] - (PartialHamEigenvalues @@ constrainedProblemVars)[j] - shiftToggle * \[Epsilon])^2,
2271      {j, presentDataIndices}],
2272      problemVarsWithStartValues,
2273      Method        -> "LevenbergMarquardt",
2274      MaxIterations -> OptionValue["MaxIterations"],
2275      AccuracyGoal  -> OptionValue["AccuracyGoal"],
2276      StepMonitor :> (
2277        steps      += 1;
2278        currentSqSum = Sum[(expData[[j]][[1]] - (PartialHamEigenvalues @@ constrainedProblemVars)[j] - shiftToggle * \[Epsilon])^2, {j, presentDataIndices}];
2279        currentRMS = Sqrt[currentSqSum / degressOfFreedom];
2280        paramSols  = AddToList[paramSols, constrainedProblemVarsList, maxHistory];
2281        rmsHistory = AddToList[rmsHistory, currentRMS, maxHistory];
2282        )
2283      ];
2284    endTime = Now;
2285    timeTaken = QuantityMagnitude[endTime - startTime, "Seconds"];
2286    PrintFun["Solution found in ", timeTaken, "s"];
2287
2288    solVec = constrainedProblemVars /. sol[[-1]];
2289    indepSolVec = indepVars /. sol[[-1]];
2290    If[addShift,
2291      \[Epsilon]Best = \[Epsilon]/. sol[[-1]],
2292      \[Epsilon]Best = 0
2293    ];
2294    fullSolVec = standardValues;
2295    fullSolVec[[problemVarsPositions]] = solVec;
2296    PrintFun["Calculating the truncated numerical Hamiltonian corresponding to the solution ..."];
2297    fullHam = compileIntermediateTruncatedHam @@ fullSolVec;
2298    PrintFun["Calculating energies and eigenvectors ..."];
```

```
2299    {eigenEnergies, eigenVectors} = Eigensystem[fullHam];
2300    states = Transpose[{eigenEnergies, eigenVectors}];
2301    states = SortBy[states, First];
2302    eigenEnergies = First /@ states;
2303    PrintFun["Shifting energies to make ground state zero of energy
        ..."];
2304    eigenEnergies = eigenEnergies - eigenEnergies[[1]];
2305    PrintFun["Calculating the linear approximant to each eigenvalue
        ..."];
2306    allVarsVec = Transpose[{allVars}];
2307    p0 = Transpose[{fullSolVec}];
2308    linMat = {};
2309    If[addShift,
2310      tail = -2,
2311      tail = -1];
2312    Do[
2313      (
2314        aVarPosition = Position[allVars, aVar][[1, 1]];
2315        isolationValues = ConstantArray[0, Length[allVars]];
2316        isolationValues[[aVarPosition]] = 1;
2317        dependentVars = KeyValueMap[{#1, D[#2, aVar]} &, Association[
        constraints]];
2318        Do[
2319          isolationValues[[Position[allVars, dVar[[1]]][[1, 1]]]] =
        dVar[[2]],
2320          {dVar, dependentVars}
2321        ];
2322        perHam  = compileIntermediateTruncatedHam @@ isolationValues;
2323        lin     = FirstOrderPerturbation[Last /@ states, perHam];
2324        linMat  = Append[linMat, lin];
2325      ),
2326      {aVar, constrainedProblemVarsList[[;;tail]]}
2327    ];
2328    PrintFun["Removing the gradient of the ground state ..."];
2329    linMat = (# - #[[1]] & /@ linMat);
2330    PrintFun["Transposing derivative matrices into columns ..."];
2331    linMat  = Transpose[linMat];
2332
2333    PrintFun["Calculating the eigenvalue vector at solution ..."];
2334    \[Lambda]0Vec = Transpose[{eigenEnergies[[presentDataIndices]]}];
2335    PrintFun["Putting together the experimental vector ..."];
2336    \[Lambda]exp = Transpose[{First /@ expData[[presentDataIndices
        ]]}];
2337    problemVarsVec = If[addShift,
2338      Transpose[{constrainedProblemVarsList[[;;-2]]}],
2339      Transpose[{constrainedProblemVarsList}]
2340    ];
2341    indepSolVecVec = Transpose[{indepSolVec}];
2342    PrintFun["Calculating the difference between eigenvalues at
        solution ..."];
2343    diff = If[linMat == {},
2344      (\[Lambda]0Vec - \[Lambda]exp)  + \[Epsilon]Best,
2345      (\[Lambda]0Vec - \[Lambda]exp)  + \[Epsilon]Best + linMat[[
        presentDataIndices]].(problemVarsVec - indepSolVecVec)
2346    ];
2347    PrintFun["Calculating the sum of squares of differences around
        solution ... "];
2348    sqdiff        = Expand[(Transpose[diff] . diff)[[1, 1]]];
2349    PrintFun["Calculating the minimum (which should coincide with sol
        ) ..."];
2350    minpoly       = sqdiff /. AssociationThread[problemVars -> solVec
        ];
2351    fmSolAssoc    = Association[sol[[2]]];
2352    If[\[Sigma]exp == Automatic,
2353      \[Sigma]exp = Sqrt[minpoly / degressOfFreedom];
2354    ];
2355    \[CapitalDelta]\[Chi]2 = Sqrt[degressOfFreedom];
2356    Amat = (1/\[Sigma]exp^2) * Transpose[linMat[[presentDataIndices
        ]]].linMat[[presentDataIndices]];
2357    paramIntervals = EllipsoidBoundingBox[Amat, \[CapitalDelta]\[Chi
        ]2];
2358    PrintFun["Calculating the uncertainty in the parameters ..."];
2359    solWithUncertainty = Table[
2360      (
2361        aVar        = constrainedProblemVarsList[[varIdx]];
2362        paramBest   = aVar /. fmSolAssoc;
```

```
2363          (aVar -> {paramBest, paramIntervals[[varIdx, 2]]})
2364        ),
2365      {varIdx, 1, Length[constrainedProblemVarsList]-shiftToggle}
2366      ];
2367
2368      bestRMS    = Sqrt[minpoly / degressOfFreedom];
2369      bestParams = sol[[2]];
2370      bestWithConstraints = Association@Join[constraints, bestParams];
2371      bestWithConstraints = bestWithConstraints /. bestWithConstraints;
2372      bestWithConstraints = (# + 0.) & /@ bestWithConstraints;
2373
2374      solCompendium["degreesOfFreedom"]     = degressOfFreedom;
2375      solCompendium["solWithUncertainty"]   = solWithUncertainty;
2376      solCompendium["truncatedDim"]         = truncationUmbral;
2377      solCompendium["fittedLevels"]         = Length[presentDataIndices
         ];
2378      solCompendium["actualSteps"]          = steps;
2379      solCompendium["bestRMS"]              = bestRMS;
2380      solCompendium["problemVars"]          = problemVars;
2381      solCompendium["paramSols"]            = paramSols;
2382      solCompendium["rmsHistory"]           = rmsHistory;
2383      solCompendium["Appendix"]             = OptionValue["
         AppendToLogFile"];
2384      solCompendium["timeTaken/s"]          = timeTaken;
2385      solCompendium["bestParams"]           = bestParams;
2386      solCompendium["bestParamsWithConstraints"] = bestWithConstraints;
2387
2388      If[OptionValue["SaveEigenvectors"],
2389          solCompendium["states"] = {#[[1]] + \[Epsilon]Best, #[[2]]}
         &/@ (Chop /@ ShiftedLevels[states]),
2390          (
2391            finalEnergies = Sort[First /@ states];
2392            finalEnergies = finalEnergies - finalEnergies[[1]];
2393            finalEnergies = finalEnergies + \[Epsilon]Best;
2394            finalEnergies  = Chop /@ finalEnergies;
2395            solCompendium["energies"] = finalEnergies;
2396        )
2397      ];
2398      If[OptionValue["SaveToLog"],
2399        PrintFun["Saving the solution to the log file ..."];
2400        LogSol[solCompendium, logFilePrefix];
2401      ];
2402      PrintFun["Finished ..."];
2403      Return[solCompendium];
2404    )
2405 ];
2406
2407 StringToSLJ[string_] := Module[
2408    {stringed = string, LS, J, LSindex},
2409    (
2410    If[StringContainsQ[stringed, "+"],
2411     Return["mixed"]
2412     ];
2413    LS = StringTake[stringed, {1, 2}];
2414    If[StringContainsQ[stringed, "("],
2415     (
2416       LSindex =
2417       StringCases[stringed, RegularExpression["\\(([^)]*)\\)"] :> "$1"
         ];
2418      LS = LS <> LSindex;
2419      stringed = StringSplit[stringed, ")"][[-1]];
2420      J = ToExpression[stringed];
2421      ),
2422     (
2423      J = ToExpression@StringTake[stringed, {3, -1}];
2424      )
2425     ];
2426    {LS, J}
2427    )
2428 ];
2429
2430 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:
2431    - 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, slope, intercept, 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     = Select[allEqns, FreeQ[#, Missing] &];
    allEqns     = Append[Coefficient[#[[1]], {F0, F2, F4, F6, ζ}], #[[2]]] & /@ allEqns;
    allEqns     = allEqns[[;; Min[Length[allEqns], maxMultiplets]]];
    subsetSizes = Range[minSubSetSize, Min[numMultiplets, maxSubSetSize]];
    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]];
```

```
2485    lastPosition = Position[numSubsets, lastSub][[1, 1]];
2486    chosenSubsetSizes = #[[1]] & /@ numSubsets[[;; lastPosition]];
2487    solutions    = <||>;
2488
2489    PrintFun["Selecting subsets of different lengths and fitting with
         ratio-constraints ..."];
2490    Do[
2491      subsets = Subsets[Range[1, Length[allEqns]], {subsetSize}];
2492      PrintFun["Considering ", Length[subsets], " barycenter subsets
       of size ", subsetSize, " ..."];
2493      Do[
2494      (
2495        subset    = subsets[[subsetIndex]];
2496        eqns      = allEqns[[subset]];
2497        slope     = #[[;; 5]] & /@ eqns;
2498        intercept = #[[6]] & /@ eqns;
2499        meritFun = Max[100 * Expand[Abs[(slope . freeIonParams -
       intercept)]/intercept]];
2500        sol = NMinimize[{meritFun,
2501          F0 > 0,
2502          F2 > 1000.,
2503          F4 == F4overF2 * F2,
2504          F6 == F6overF2 * F2,
2505          ζ > 0},
2506          freeIonParams,
2507          MaxIterations -> maxIterations,
2508          Method -> "Convex"];
2509        solutions[{subsetSize, subset}] = sol;
2510        )
2511      , {subsetIndex, 1, Length[subsets]}
2512      ],
2513      {subsetSize, chosenSubsetSizes}
2514    ];
2515
2516    PrintFun["Collecting solutions of different subset size ..."];
2517    goodThings = Table[
2518      (chunk =
2519        Normal[Sort[
2520          KeySelect[#[[1]] & /@ solutions, #[[1]] == subSize &]]];
2521        If[Length[chunk] >= 1,
2522        chunk[[1]],
2523        Null
2524        ]), {subSize, subsetSizes}];
2525    goodThings = Select[goodThings, Head[#] === Rule &];
2526
2527    PrintFun["Picking the solutions that are not worse than ",
       maxPercent, "% ..."];
2528    bestThings = Select[goodThings, #[[2]] <= maxPercent &];
2529    If[bestThings == {},
2530      Print["No acceptable solution found, consider increasing
       maxPercent or inspecting the given data ..."];
2531      Return[goodThings];
2532    ];
2533
2534    PrintFun["Keeping the solution with the largest number of used
       barycenters ..."];
2535    bestOfAll = bestThings[[-1]];
2536    sol       = solutions[bestOfAll[[1]]];
2537    subset    = bestOfAll[[1, 2]];
2538    eqns      = allEqns[[subset]];
2539    slope     = #[[;; 5]] & /@ eqns;
2540    intercept = #[[6]] & /@ eqns;
2541    usedMultiplets  = Keys[multipletEnergies][[subset]];
2542    usedBaricenters = {#, multipletEnergies[#]} & /@ usedMultiplets;
2543    uniqeLS = DeleteDuplicates[#[[2]] &/@ Keys[multipletEnergies]];
2544    solAssoc = Association[sol[[2]]];
2545    usedLaF3 = False;
2546    If[Length[uniqeLS] == 1,
2547      (
2548        Print["There is too little data to find Slater parameters,
       using the ones for LaF3, and keeping the fitted spin-orbit zeta
       ..."];
2549        laf3params = LoadLaF3Parameters[ln];
2550        usedLaF3   = True;
2551        solAssoc[F0] = laf3params[F0];
2552        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];
)
];

EndPackage[];
```

18.3 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,
```

```
222         FuzzyRectangle[#, lineWidth, 0, height, colorFun[#]] &/@
    lines,
223       Not[NumberQ[lineWidth]] && Length[OptionValue["Intensities"]]
    == 0,
224         MapThread[FuzzyRectangle[#1, #2, 0, height, colorFun[#1]] &,
    {lines, lineWidth}],
225       NumberQ[lineWidth] && Length[OptionValue["Intensities"]] > 0,
226         MapThread[FuzzyRectangle[#1, lineWidth, 0, height, colorFun
    [#1], #2] &, {lines, OptionValue["Intensities"]}],
227       Not[NumberQ[lineWidth]] && Length[OptionValue["Intensities"]] >
     0,
228         MapThread[FuzzyRectangle[#1, #2, 0, height, colorFun[#1], #3]
     &, {lines, lineWidth, OptionValue["Intensities"]}]
229     ];
230     comments = Which[
231       Length[OptionValue["Comments"]] > 0,
232         MapThread[StringJoin[ToString[#1]<>" nm","\n",ToString[#2]]&,
233         {lines, OptionValue["Comments"]}],
234       Length[OptionValue["Comments"]] == 0,
235         ToString[#] <>" nm" & /@ lines,
236       True,
237       {}
238     ];
239     If[OptionValue["Tooltips"],
240       fuzzyLines = MapThread[Tooltip[#1, #2] &, {fuzzyLines, comments
    }];
241     ];
242     graphicsOpts = FilterRules[{opts}, Options[Graphics]];
243     Graphics[fuzzyLines,
244         graphicsOpts,
245         Background -> Black,
246         PlotRange -> {{minWave, maxWave}, {0, height}}]
247     )
248   ];
249
250 End[];
251
252 EndPackage[];
```

18.4 misc.m

This module includes a few functions useful for data-handling.

```
1  BeginPackage["DavidLizarazo`misc`"];
2  (* Needs["MaTeX`"]; *)
3
4  ArrayBlocker;
5  BlockAndIndex;
6  BlockArrayDimensionsArray;
7  BlockMatrixMultiply;
8  BlockTranspose;
9
10 EllipsoidBoundingBox;
11 ExportToH5;
12 FirstOrderPerturbation;
13 FlattenBasis;
14
15 GetModificationDate;
16 HamTeX;
17 HelperNotebook;
18
19 RecoverBasis;
20 RemoveTrailingDigits;
21 ReplaceDiagonal;
22 RobustMissingQ;
23
24 RoundToSignificantFigures;
25 RoundValueWithUncertainty;
26 SecondOrderPerturbation;
27 SuperIdentity;
28
29 TextBasedProgressBar;
30 ToPythonSparseFunction;
31 ToPythonSymPyExpression;
32 TruncateBlockArray;
33
```

```
Begin["`Private`"];

  ExtractSymbolNames;

  RemoveTrailingDigits[s_String] := StringReplace[s,
    RegularExpression["\\d+$"] -> ""];

  BlockTranspose::usage="BlcockTranspose[anArray] takes a 2D array
    with a congruent block structure and returns the transposed array
    with the same block structure.";
  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[{w1, w2, ...}, idx] takes a
    list of block lengths wi and an index idx. The function returns in
     which block idx would be in a a list defined by {Range[1,w1],
    Range[1+w1,w1+w2], ...}. The function also returns the position
    within the bin in which the given index would be found in. 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 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. The blocks in the given array
    may be SparseArray. This is equivalent to FlattenArray[blockArray
    ][truncationIndices, truncationIndices] but may be more efficient
    if 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]],
  {fullRowIndex,truncationIndices},
  {fullColIndex,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;
   case      = Length[Dimensions[blockSizes]];
   Which[
     case == 3,
     (
       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}];
     ),
     case == 1,
     (
       expandedSizes = Table[
         {blockSizes[[idxRow]], blockSizes[[idxCol]]},
         {idxRow,1,Length[blockSizes]},
         {idxCol,1,Length[blockSizes]}
         ];
       numBlocks = Length[expandedSizes];
       blockedArray = Table[(
         blockSize = expandedSizes[[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]}]];
```

```
147
148 Options[RoundValueWithUncertainty] = {"SetPrecision" -> False};
149 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.
150 The returned value is a list with the form {roundedX, roundedDx}.
151 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.
152 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.";
153 RoundValueWithUncertainty[x_, dx_, OptionsPattern[]] := Module[
154   {xExpo, dxExpo, sigFigs, roundedX, roundedDx, returning},
155   (
156     xExpo   = RealDigits[x][[2]];
157     dxExpo  = RealDigits[dx][[2]];
158     sigFigs = (xExpo - dxExpo) + 1;
159     {roundedX, roundedDx} = If[sigFigs <= 0,
160       {0., N@RoundToSignificantFigures[dx, 1]},
161       N[
162       {
163         RoundToSignificantFigures[x, xExpo - dxExpo + 1],
164         RoundToSignificantFigures[dx, 1]}
165       ]
166       ];
167     returning = If[
168       OptionValue["SetPrecision"],
169       {SetPrecision[roundedX, Max[1, sigFigs]],
170       SetPrecision[roundedDx, 1]},
171       {roundedX, roundedDx}
172       ];
173     Return[returning]
174     )
175   ];
176
177 RoundToSignificantFigures::usage =
178   "RoundToSignificantFigures[x, sigFigs] rounds x so that it only
     has \
179 sigFigs significant figures.";
180 RoundToSignificantFigures[x_, sigFigs_] :=
181   Sign[x]*N[FromDigits[RealDigits[x, 10, sigFigs]]];
182
183 RobustMissingQ[expr_] := (FreeQ[expr, _Missing] === False);
184
185 TextBasedProgressBar[progress_, totalIterations_, prefix_:""] :=
     Module[
186     {progMessage},
187     progMessage = ToString[progress] <> "/" <> ToString[
     totalIterations];
188     If[progress < totalIterations,
189         WriteString["stdout", StringJoin[prefix, progMessage, "\r"
     ]],
190         WriteString["stdout", StringJoin[prefix, progMessage, "\n"
     ]]
191     ];
192 ];
193
194 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]_i +
     \[Delta]_i β, where the \[Delta]_i are the returned values. This
     assuming that the eigenvalues are non-degenerate.";
195 FirstOrderPerturbation[eigenVectors_,
196   perMatrix_] := (Chop@Diagonal[
197   Conjugate@eigenVectors . perMatrix . Transpose[eigenVectors]])
198
199 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.";
200 SecondOrderPerturbation[eigenValues_, eigenVectors_, perMatrix_] :=
     (
201   dim = Length[perMatrix];
202   eigenBras = Conjugate[eigenVectors];
203   eigenKets = eigenVectors;
204   matV = Abs[eigenBras . perMatrix . Transpose[eigenKets]]^2;
205   OneOver[x_, y_] := If[x == y, 0, 1/(x - y)];
206   eigenDiffs = Outer[OneOver, eigenValues, eigenValues, 1];
207   pProduct = Transpose[eigenDiffs]*matV;
208   Return[2*(Total /@ Transpose[pProduct])];
209   )
210
211 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.";
212 SuperIdentity[args___] := {args};
213
214 ExtractSymbolNames[expr_Hold] := Module[
215   {strSymbols},
216   strSymbols = ToString[expr, InputForm];
217   StringCases[strSymbols, RegularExpression["\\w+"]][[2 ;;]]
218   ]
219
220 ExportToH5::usage =
221   "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
    which mixes numbers and strings will fail, but an Association with
     mixed values exports ok. Do give it a try.
222   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.";
223 Options[ExportToH5] = {"Overwrite" -> True};
224 ExportToH5[fname_String, symbols_Hold, OptionsPattern[]] := (
225   If[And[FileExistsQ[fname], OptionValue["Overwrite"]],
226   (
227     Print["File already exists, overwriting ..."];
228     DeleteFile[fname];
229     )
230   ];
231   symbolNames = ExtractSymbolNames[symbols];
232   Do[(Print[symbolName];
233     Export[fname, ToExpression[symbolName], {"Datasets", symbolName
    },
234     OverwriteTarget -> "Append"]
235     ), {symbolName, symbolNames}]
236   )
237
238 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.";
239 HelperNotebook[nbName_:"OUT"] :=
240 Module[{screenDims, screenWidth, screenHeight, nbWidth, leftMargin,
241   PrintToOutputNb}, (
242   screenDims =
243     SystemInformation["Devices", "ScreenInformation"][[1, 2, 2]];
244   screenWidth = screenDims[[1, 2]];
245   screenHeight = screenDims[[2, 2]];
246   nbWidth = Round[screenWidth/3];
247   leftMargin = screenWidth - nbWidth;
248   outputNb = CreateDocument[{}, WindowTitle -> nbName,
249     WindowMargins -> {{leftMargin, Automatic}, {Automatic,
250         Automatic}}, WindowSize -> {nbWidth, screenHeight}];
251   PrintToOutputNb[text_] :=
252     (
253         SelectionMove[outputNb, After, Notebook];
```

```
254           NotebookWrite[outputNb, Cell[BoxData[ToBoxes[text]], "
      Output"]];
255       );
256     Return[PrintToOutputNb]
257     )
258     ]
259
260   GetModificationDate::usage="GetModificationDate[fname] returns the
      modification date of the given file.";
261   GetModificationDate[theFileName_] := FileDate[theFileName, "
      Modification"];
262
263   (*Helper function to convert Mathematica expressions to standard
      form*)
264   StandardFormExpression[expr0_] := Module[{expr=expr0}, ToString[
      expr, InputForm]];
265
266   (*Helper function to translate to Python/SymPy expressions*)
267   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."
      ;
268   ToPythonSymPyExpression[expr0_] := Module[{standardForm, expr=expr0
      },
269     standardForm = StandardFormExpression[expr];
270     StringReplace[standardForm, {
271       "Power[" -> "Pow(",
272       "Sqrt[" -> "sq(",
273       "[" -> "(",
274       "]" -> ")",
275       "\\" -> "",
276       "I" -> "1j",
277       "^" -> "**",
278       (*Remove special Mathematica backslashes*)
279       "/" -> "/" (*Ensure division is represented with a slash*)}]];
280
281   ToPythonSparseFunction::usage="ToPythonSparseFunction[array,
      funName] takes a 2D array (whose elements are linear combinations
      of a set of variables) and returns a string which defines a Python
       function that can be used to evaluate the given array in Python.
      In Python the name of the function is equal to funName. The given
      array may be a list of lists or a 2D SparseArray.";
282   ToPythonSparseFunction[arrayInput_, funName_] :=
283     Module[{
284       sparseArray = arrayInput,
285       rowPointers,
286       dimensions,
287       pyCode,
288       vars,
289       varList,
290       dataPyList,
291       colIndicesPyList
292       },
293     (
294       If[Head[sparseArray] === List,
295         sparseArray = SparseArray[sparseArray]
296         ];
297       (* Extract unique symbolic variables from the SparseArray *)
298       vars       = Union[Cases[Normal[sparseArray], _Symbol, Infinity
      ]];
299       varList    = StringRiffle[ToString /@ vars, ", "];
300       (* Convert data to SymPy compatible strings *)
301       dataPyList = StringRiffle[ToPythonSymPyExpression /@ Normal[
      sparseArray["NonzeroValues"]], ",\n        "];
302       colIndicesPyList = StringRiffle[
303         ToPythonSymPyExpression /@ (Flatten[Normal[sparseArray["
      ColumnIndices"]] - 1]), ", "];
304       (* Extract sparse array properties *)
305       rowPointers = Normal[sparseArray["RowPointers"]];
306       dimensions = Dimensions[sparseArray];
307       (*Create Python code string*)
308       pyCode = StringJoin[
309         "#!/usr/bin/env python3\n\n",
310         "from scipy.sparse import csr_matrix\n",
311         "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"];
      Return[pyCode];
    )
    ];

  Options[HamTeX] = {"T2" -> False};
  HamTeX::usage="HamTeX[numE] returns the LaTeX code for the semi-
    empirical Hamiltonian for f^numE. 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.";
  HamTeX[nE_, OptionsPattern[]] := (
    tex = Which[
      MemberQ[{1, 13}, nE],
        "\\hat{H}&=\\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+\\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"
      ];
    Return[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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