

EFFECTIVE NUCLEAR OPERATORS WITH THE
COUPLED-CLUSTER METHOD

By

Samuel John Novario

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PUBLIC ABSTRACT

EFFECTIVE NUCLEAR OPERATORS WITH THE COUPLED-CLUSTER METHOD

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ABSTRACT

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KEY TO SYMBOLS AND ABBREVIATIONS

Ap-Bh *A*-particle, *B*-hole excitation or de-excitation from the reference state

$|0\rangle$ vacuum state

$|\Phi_0\rangle$ reference state

$|\Psi\rangle$ correlated ground state

$|\Phi_{i_1 \dots i_B}^{a_1 \dots a_A}\rangle$ specific *Ap-Bh* state

$\{\dots\}$ normal-ordered with respect to the reference state

$\hbar\omega$ harmonic oscillator energy scale

\hat{H} Hamiltonian

\bar{H} similarity-transformed Hamiltonian

\hat{H}_N normal-ordered Hamiltonian

\bar{H}_N normal-ordered similarity-transformed Hamiltonian

\hat{T} cluster operator

$\varepsilon_{i_1 \dots i_B}^{a_1 \dots a_A}$ energy denominator, $f_{i_1}^{i_1} + \dots + f_{i_B}^{i_B} - f_{a_1}^{a_1} - \dots - f_{a_A}^{a_A}$

CC coupled cluster

CCD coupled cluster with doubles

CCSD coupled cluster with singles and doubles

CCSDT coupled cluster with singles, doubles, and triples

CCSD(T) coupled cluster with singles, doubles, and triples approximation

Λ -CCSD(T) coupled cluster with singles, doubles, and Λ -triples approximation

CI configuration interaction

FCI full configuration interaction

COM center of mass

EOM equations-of-motion

PA particle-attached

PR particle-removed
EOM equations-of-motion
EOM-CC equations-of-motion coupled cluster
EOM-CCSD equations-of-motion coupled cluster with singles and doubles
HF Hartree-Fock
IM-SRG in-medium similarity renormalization group
HO harmonic oscillator
LECs low-energy constants
MBPT many-body perturbation theory
WS Woods-Saxon
DIIS direct-inversion of the iterative subspace
QCD quantum chromodynamics
EFT effective field theory
NN nucleon-nucleon
3N three-nucleon
NLO next-to leading order
N²LO next-to-next-to leading order
N³LO next-to-next-to-next-to leading order

Chapter 1

Introduction

Steady progress in any modern scientific endeavor requires a strong, dynamic relationship between experimental data to paint an accurate picture of some natural phenomena and theoretical models to interpret those phenomena with respect to the growing network of other scientific models. Conversely, the predictive capability of theoretical models can highlight blurry or unfinished areas of that picture which can be clarified or completed by new or improved experimental techniques. In the pursuit to understand and describe the atomic nucleus and the corresponding implications from quarks to neutron stars, this push-and-pull coordination between theory and experiment makes progress in modern nuclear physics robust and persistent.

An integral component of modern nuclear physics is describing the structure and emergent properties of self-bound systems of protons and neutrons. The systems in question can be stable nuclei, rare isotopes far from stability, and even infinite nuclear matter which can be used to model neutron stars. Relevant properties to nuclear structure include ground-state energies—for determining nuclear masses, excited-state energies—for identification in gamma or neutron spectroscopy, and transition or decay amplitudes—for calculating the respective rates for those processes. This wide array of emergent properties inserts both nuclear structure theory and experiment into a prominent role within every other subfield of modern nuclear physics, from lattice quantum chromodynamics (QCD) to nuclear astrophysics, and beyond, to questions about fundamental symmetries and dark matter. However, two inextricable characteristics of a comprehensive model of nuclear structure—the increasingly large

size of many-body nuclear systems and the complexity and strength of the nucleon-nucleon interactions—have been imposing hurdles for theorists to overcome.

1.1 A Brief History of Nuclear Structure Theory

The project to solve the correlation problem in many-fermion systems began with the work of Brueckner, Bethe, and Goldstone [22, 11, 1] with the reformulation of the nuclear interaction by accounting for two-body correlations from the nuclear medium. This work continued with the work of Coester [30, 31] and Kummel [76], amongst many other, with a further resummation of nuclear correlations in the form of an exponential ansatz into what would become coupled-cluster (CC) theory. However, there were two major obstacles that hindered the progress in this area for decades. First, while these methods were systematically improvable by including progressively higher-level correlations, the highly nonperturbative nature of the nuclear force required computationally infeasible summations. Second, there wasn't a reliable and consistent theory to model nucleon-nucleon interactions.

However, with the well-known and highly-perturbative Coulomb force, which underlies the many-electron systems in atoms and molecules, the field of quantum chemistry made consistent advances in ab-initio quantum chemistry possible since the 1950s. Along with the quasi-exact method of configuration interaction (CI) theory [110, 32, 4, 129] which have been utilized since the formulation of quantum mechanics, chemists successfully employed approximate methods like many-body perturbation theory (MBPT) [66, 67, 105, 108] and coupled-cluster theory [28, 26, 27, 96, 108].

Fortunately, within the past decade, two breakthroughs have allowed ab-initio nuclear structure to resurface and thrive the way that quantum chemistry had done in the previous decades. First was the invention of chiral effective field theory (EFT) [41, 83] which gave

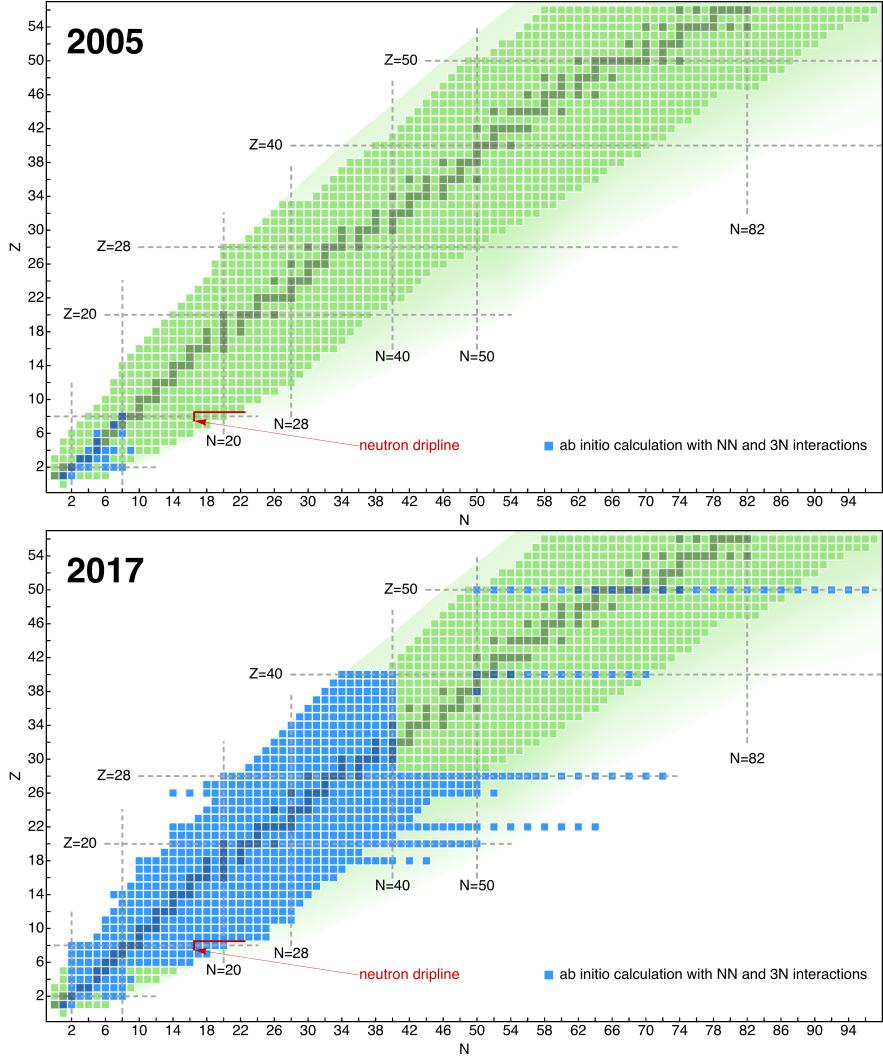


Figure 1.1: Nuclear chart of nuclei with ground-state energies which have been calculated with ab-initio methods and NN+3N interactions. Figure taken from [64].

theorists the ability to construct nucleon-nucleon interactions consistent with the underlying QCD of the strong nuclear force. Second was the application of renormalization group (RG) methods to the nuclear force [17, 104]. This procedure can “soften” the NN interaction, to decouple the high- and low-momentum components of the nuclear force and generate less-correlated systems that can be calculated at a reasonable computational cost. These major changes to nuclear structure theory made it possible to merge the field with the progress of quantum chemistry and open a new area for additional developments in *ab initio* descriptions

of many-fermion systems, see Fig. 1.1.

Along with exponential improvements to high-performance computing, these novel techniques have allowed modern many-body methods to extend their reach and deepen their applicability across the nuclear chart, see Fig. 1.2. The no-core shell model (NCSM), an exact method for a given model space, has been useful in calculating the radii, transition strengths, and effective interactions of light nuclei and has been extended to nuclei in the *sd* shell [90, 91, 6]. A quasi-exact technique which follows a completely different methodology than NCSM, quantum Monte Carlo (QMC), has also progressed and is now capable of calculating properties of light nuclei with modern chiral forces [98, 97, 25]. In addition to these exponentially scaling techniques' successes with lighter nuclei, polynomially scaling techniques—such as the in-medium similarity renormalization group (IMSRG) [127, 128, 62, 15, 63, 61, 116, 115], self-consistent Green's functions (SCGF) [112, 113, 114], and coupled cluster theory [136, 137, 69, 70, 53, 74, 49, 12]—have been able to reach open-shell nuclei through the *pf* shell and even up to the chain of even tin isotopes with equations-of-motion and multi-reference techniques.

1.2 Electroweak Theory and Nuclear Structure

Nuclear structure is implicated in performing and analyzing experiments to probe fundamental symmetries and physics beyond the Standard Model. One example is determining the V_{ud} component of the Cabibbo-Kobayashi-Maskawa (CKM) matrix, which relates quark eigenstates of the weak interaction to their mass eigenstates [24, 72]. This component can be determined from by measuring the half-lives of superallowed Fermi β decays [121] and applying a nucleus-dependent structure correction [122, 126, 125, 5, 71]. The value of $|V_{ud}|$ is used to test the unitarity of the CKM matrix and the conserved-vector current hypothesis,

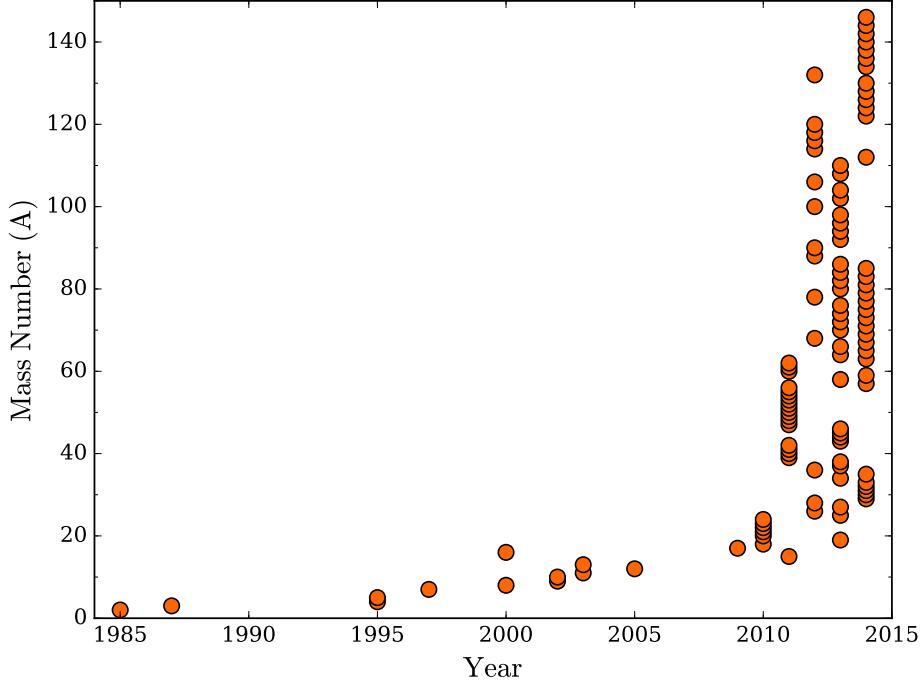


Figure 1.2: Progress of ab-initio nuclear structure from calculations of ground-state energies with NN+3N interactions. Early progress was approximately linear as the problem size scaled with Moore’s law while more recent progress has taken advantage of new algorithms which have outpaced Moore’s law. Data taken from [64].

which relates the $f\ell$ -values of superallowed Fermi β decays of different nuclei, both predicted by the standard model [56].

Another example of physics beyond the standard model is the neutrinoless double- β decay ($0\nu\beta\beta$) [117, 3]. The extremely-rare, two-neutrino double- β decay ($2\nu\beta\beta$) has been observed in many experiments [38, 86], which has motivated the search for its neutrinoless counterpart, in which two Majorana neutrinos, being their own antiparticles, annihilate one another, which is not possible in the standard electro-weak theory. The long half-lives of these theoretical decays depend on a phase-space factor, which is highly dependent on the decay Q -value, and a nuclear matrix element. The Q -value can be determined from high-precision mass measurements of the relevant nuclei [80, 51, 102, 23], while the nuclear matrix element, which contributes the largest source of uncertainty, must be calculated with

a reliable many-body theory.

The weak interaction and nuclear structure can also be exploited for supernova neutrino detection and spectroscopy. While these original detectors were based on electron-neutrino scattering [65, 13], more recent experiments utilize correlated nucleon effects of large nuclei to enhance the scattering cross section and therefore the ability to resolve energies and distinguish neutrino flavors [58, 29, 43, 77]. Supernova models predict distinct distributions for different neutrino flavors based on the temperatures at which they are emitted [73, 9]. With nuclear structure calculations that include sufficient nuclear correlations, these high-resolution detectors can be used to verify specific models.

1.3 Ab-Initio Descriptions of β Decay

Since Enrico Fermi's originally rejected paper describing β decay in 1934 [44, 135], theorists have worked to refine this description within the ever-growing library of knowledge concerning the nature of the weak force, the characteristics of the neutrino, and the structure of nuclei. With the success of *ab initio* calculations for nuclear properties such as masses, radii, and electromagnetic phenomena, these techniques also seem promising ways to calculate relevant quantities involved in nuclear β decay. Because the kinematics of the decay and the underlying weak process are well understood, the remaining task for nuclear theory to tackle is calculating the transition amplitudes between the initial and final nuclei.

Modern calculations of these β -decay matrix elements were originally performed using phenomenological interactions in the shell model framework [134, 20, 131, 93]. Also, predecessors to current ab-initio techniques like the random-phase approximation (RPA) [124] included core-correlation effects in these early descriptions. These methods were able to successfully reproduce experimental lifetime data and address technical issues such as the

quenching of the axial-vector coupling constant. More recently, the success of the shell model has inspired an extension to the new method, known as the ab-initio shell model [6], where an effective interaction is constructed within a certain valence space using a many-body method such as CC [37] or IMSRG [16]. However, these techniques are computationally expensive and cannot currently reach heavy nuclei of interest. The most common method used in their place is known as the quasiparticle random-phase approximation (QRPA) [118, 39]. While these calculations can be performed for heavy nuclei in large spaces, they also rely on phenomenological effective interactions. Therefore, there is a demand for computationally-economical, *ab initio* techniques that can capture the relevant many-body correlations needed to accurately describe the nuclear structure aspects of electro-weak processes.

1.4 Thesis Structure

The main goal of this work is to explore the ab-initio description of nuclear β decay within the coupled-cluster theory framework of the *equation-of-motion coupled cluster with singles and doubles* (EOM-CCSD) method using renormalized chiral NN and 3N interactions. The organization of the thesis builds from a general description of the many-body problem of quantum mechanics in chapter 2. Then, in chapter 3, this many-body framework is applied within the coupled-cluster theory and applied to various systems including, atomic nuclei. In chapter 4, coupled-cluster theory is extended to the equations-of-motion method to describe open-shell systems. Chapter 5 outlines the procedure to express observables as effective coupled-cluster operators and how to calculate those observables in the equations-of-motion framework. Then, in chapter 5.1, the ability to calculate effective operators is applied to Fermi and Gamow-Teller β -decay operators and relevant quantities are determined for various nuclei. Lastly, conclusions and future perspectives are given in chapter 6 while technical

details concerning the formalism and implementation are given in the appendix 8.

Chapter 2

Many-Body Quantum Mechanics

Ab initio structure calculations of many-fermion systems such as those in nuclear and electronic structure aim to describe emergent phenomena from the constituent particles subject to the underlying microscopic Hamiltonian. This amounts to finding the solution to the many-body Schrödinger equation. However, a calculation of the exact solution needs to account for all possible correlations among the particles and thus scales factorially. This motivates the need for approximations to the exact solution that account for the most important correlations. This chapter first establishes the formalism necessary to define the many-body problem then illustrates several successive approximations to its solution. Because the type of fermions and the underlying Hamiltonian can be kept generic until specific systems are considered, the formalism and many-body methods can be kept generic as well.

2.1 Independent-Particle Model

The nonrelativistic A -body quantum problem begins with the Schrödinger equation,

$$\hat{H}\Psi_\nu(\mathbf{r}_1, \dots, \mathbf{r}_A) = E_\nu\Psi_\nu(\mathbf{r}_1, \dots, \mathbf{r}_A), \quad (2.1)$$

for the correlated wave function $\Psi_\nu(\mathbf{r}_1, \dots, \mathbf{r}_A)$ and the corresponding energy E_ν . The Hamiltonian can be written generically as a sum of k -body pieces which, in principle, can

contain up to A -body interactions,

$$\begin{aligned}\hat{H} &= {}^{(1)}\hat{H} + {}^{(2)}\hat{H} + {}^{(3)}\hat{H} + \dots \\ &= \sum_i^A {}^{(1)}\hat{H}(\mathbf{r}_i) + \sum_{i < j}^A {}^{(2)}\hat{H}(\mathbf{r}_i, \mathbf{r}_j) + \sum_{i < j < k}^A {}^{(3)}\hat{H}(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k) + \dots\end{aligned}\quad (2.2)$$

Regardless of the system, the one-body term contains the kinetic energy operator $\frac{-\hbar^2}{2m}\nabla_i^2$, while the higher-order terms result from inter-particle interactions.

An intuitive way to formulate the solution to the many-body Schrödinger equation is to express the collective wave function in terms of independent single-particle wave functions, or orbitals $\phi(\mathbf{r})$. In this *independent-particle model*, a selection of single-particle wave functions, known as the single-particle basis, are constructed by solving the Schrödinger equation for a single particle in either a mean-field potential for bound systems or in free space for infinite systems. Then a many-body wave function is constructed as a product of these single-particle orbits. This simple model is justified because it becomes exact when inter-particle interactions are completely suppressed and is useful because it provides an intuitive way to interpret complicated many-body dynamics as processes involving few single-particle wave functions.

A many-body wave function of fermions must be anti-symmetric with respect to particle exchange so that the Pauli exclusion principle is followed, such that no single-particle wave function is occupied by more than one fermion. This condition is satisfied by a wave function

in the form of a *Slater determinant* [111],

$$\Phi(\mathbf{r}_1, \dots, \mathbf{r}_A) = \frac{1}{\sqrt{A!}} \begin{vmatrix} \phi_1(\mathbf{r}_1) & \phi_1(\mathbf{r}_2) & \cdots & \phi_1(\mathbf{r}_A) \\ \phi_2(\mathbf{r}_1) & \phi_2(\mathbf{r}_2) & \cdots & \phi_2(\mathbf{r}_A) \\ \vdots & \vdots & \ddots & \vdots \\ \phi_A(\mathbf{r}_1) & \phi_A(\mathbf{r}_2) & \cdots & \phi_A(\mathbf{r}_A) \end{vmatrix}, \quad (2.3)$$

where A is the number of particles in the system and $\phi_p(\mathbf{r}_\mu)$ is the p -th orbital filled with the μ -th particle.

If the orbitals are constructed from an appropriate phenomenological potential, a Slater determinant composed of the A lowest orbitals can represent a fairly good approximation to the ground state for a closed-shell system, where the lowest-energy Slater determinant can be uniquely determined. The set of all Slater determinants in a certain model space of single-particle wave functions defines a complete A -body Hilbert space such that a generic wave function can be written as a linear combination of Slater determinants,

$$\Psi_\nu(\mathbf{r}_1, \dots, \mathbf{r}_A) = \sum_{\mu=1}^{\mathcal{N}} C_\nu^\mu \Phi_\mu(\mathbf{r}_1, \dots, \mathbf{r}_A), \quad (2.4)$$

where $C_\nu^\mu = \langle \Psi(\mathbf{r}_1, \dots, \mathbf{r}_A) | \Phi_\nu^\mu(\mathbf{r}_1, \dots, \mathbf{r}_A) \rangle$. The number of Slater determinants \mathcal{N} in an A -body Hilbert space with N orbits is given by,

$$\mathcal{N} = \binom{N}{A} = \frac{N!}{A!(N-A)!}, \quad (2.5)$$

which shows the factorial scaling of the exact problem. However, to reduce the size of the problem, progressively more significant Slater determinants can be chosen to systematically

refine approximations to the full solution.

2.2 Second Quantization

Even with the simplification of the independent-particle model, the many-body Schrödinger equation is an unwieldy and complex system of coupled differential equations. A useful reformulation of this equation is to promote the single-particle orbits to operators in a step known as *second quantization* (see e.g., [108, 45]). In this framework, a Slater determinant is represented by a string of occupied orbitals,

$$\Phi(\mathbf{r}_1, \dots, \mathbf{r}_A) \equiv \mathcal{A}(\phi_{p_1} \phi_{p_2} \phi_{p_3} \dots \phi_{p_N}) \equiv |p_1 p_2 p_3 \dots p_N\rangle, \quad (2.6)$$

where \mathcal{A} represents a permutation and normalization operator to correspond with Eq. (2.3). These second-quantized Slater determinants can be constructed with the use of operators that correspond to specific orbitals. A *creation* operator, \hat{a}_p^\dagger , places a particle in the p orbital, and an *annihilation* operator, \hat{a}_p , removes a particle from the p orbital,

$$\hat{a}_p^\dagger |0\rangle = |p\rangle \quad \hat{a}_p |p\rangle = |0\rangle, \quad (2.7)$$

where $|0\rangle$ represents the true vacuum, a state void of any particles. Because there must be a correspondence between the original first quantization and second quantization, these creation and annihilation operators obey the following anticommutation relations ($[\hat{A}, \hat{B}]_+ = \hat{A}\hat{B} + \hat{B}\hat{A}$),

$$[\hat{a}_p^\dagger, \hat{a}_q]_+ = \delta_{pq} \quad [\hat{a}_p^\dagger, \hat{a}_q^\dagger]_+ = [\hat{a}_p, \hat{a}_q]_+ = 0, \quad (2.8)$$

which guarantee that wave functions comprised of these operators obey antisymmetry and the Pauli exclusion principle required of fermionic systems.

The Hamiltonian in the form of Eq. (2.2) can be written with second-quantized operators as,

$$\hat{H} = \sum_{pq} {}^{(1)}H_q^p \hat{a}_p^\dagger \hat{a}_q + \frac{1}{4} \sum_{pqrs} {}^{(2)}H_{rs}^{pq} \hat{a}_p^\dagger \hat{a}_q^\dagger \hat{a}_s \hat{a}_r + \frac{1}{36} \sum_{pqrstu} {}^{(3)}H_{stu}^{pqr} \hat{a}_p^\dagger \hat{a}_q^\dagger \hat{a}_r^\dagger \hat{a}_u \hat{a}_t \hat{a}_s + \dots, \quad (2.9)$$

where the prefactors account for the double counting of particle-particle interactions, and the matrix elements represent integrals over the relevant single-particle wave functions,

$$\begin{aligned} {}^{(1)}H_q^p &\equiv \int d\mathbf{r}_1 \phi_p^*(\mathbf{r}_1) {}^{(1)}\hat{H}(\mathbf{r}_1) \phi_q(\mathbf{r}_1) \\ {}^{(2)}H_{rs}^{pq} &\equiv \int d\mathbf{r}_1 d\mathbf{r}_2 \phi_p^*(\mathbf{r}_1) \phi_q^*(\mathbf{r}_2) {}^{(2)}\hat{H}(\mathbf{r}_1, \mathbf{r}_2) [\phi_r(\mathbf{r}_1) \phi_s(\mathbf{r}_2) - \phi_s(\mathbf{r}_1) \phi_r(\mathbf{r}_2)] \\ &\vdots \end{aligned} \quad (2.10)$$

Matrix elements involving two or more particles include exchange terms which guarantee that they are also antisymmetric,

$$\begin{aligned} {}^{(2)}H_{rs}^{pq} &= -{}^{(2)}H_{rs}^{qp} = -{}^{(2)}H_{sr}^{pq} = {}^{(2)}H_{sr}^{qp} \\ {}^{(3)}H_{stu}^{pqr} &= -{}^{(3)}H_{stu}^{qpr} = -{}^{(3)}H_{tsu}^{pqr} = {}^{(3)}H_{tsu}^{qpr} = \dots \end{aligned} \quad (2.11)$$

These definitions apply regardless of the form of the Hamiltonian, and thus this formalism remains generic to the particular system. Second quantization is a crucial step in simplifying the many-body Schrödinger equation because it reduces the complexity of the spatial and spin degrees of freedom within the single-particle wave functions and interactions into precomputed matrix elements. The remaining effort is reduced to algebraic expressions in-

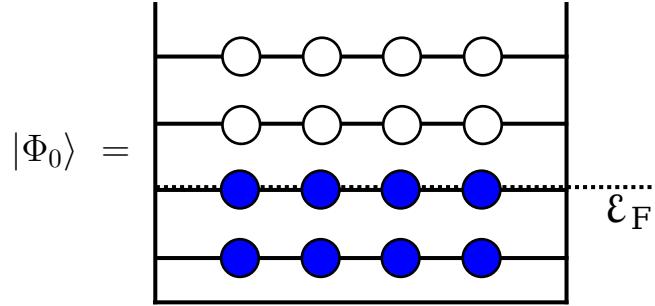


Figure 2.1: A depiction of the closed-shell reference state in the independent particle model. Each horizontal line represents a shell of single-particle orbits, represented by circles, and the dotted line represents the Fermi level which separates the unoccupied *particle* states from the occupied *hole* states.

volving creation and annihilation operators.

2.3 Normal Ordering

It's convenient to define an A -particle reference state, where states are filled from the true vacuum up to a closed shell, known as the Fermi level. This reference state must be uniquely determined from the number of particles in the system and therefore nondegenerate with other Slater determinants,

$$|\Phi_0\rangle = \left\{ \prod_i^A \hat{a}_i^\dagger \right\} |0\rangle. \quad (2.12)$$

This reference determinant defines a new *Fermi vacuum*. States above the Fermi vacuum are called *particle* states and will be denoted with the indices $a, b, c, d\dots$ while states below the Fermi vacuum are called *hole* states and will be denoted with the indices $i, j, k, l\dots$. Generic states above or below the Fermi vacuum will be denoted with the indices $p, q, r, s\dots$.

Any other Slater determinant can be constructed relative to this reference state by adding particles and/or removing holes. A Slater determinant with A particles added and B holes

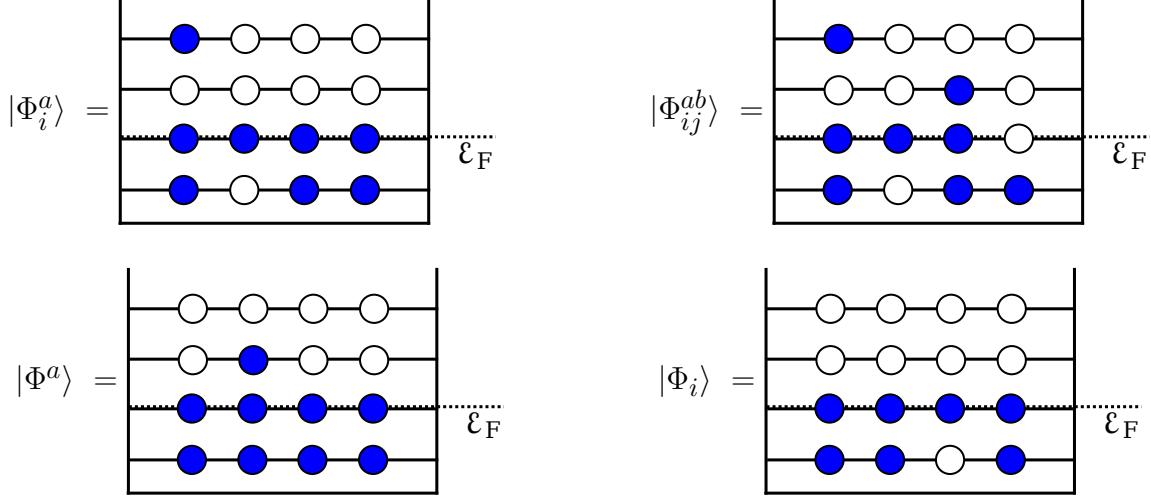


Figure 2.2: A depiction of $1p$ - $1h$, $2p$ - $2h$, $1p$ - $0h$, and $0p$ - $1h$ Slater determinants defined relative to the reference state in the independent particle model.

removed from reference state is known as a *Ap-Bh* excitation.

$$\begin{aligned}
 |\Phi_i^a\rangle &\equiv \hat{a}_a^\dagger \hat{a}_i |\Phi\rangle \\
 |\Phi_{ij}^{ab}\rangle &\equiv \hat{a}_a^\dagger \hat{a}_b^\dagger \hat{a}_j \hat{a}_i |\Phi\rangle \\
 |\Phi^a\rangle &\equiv \hat{a}_a^\dagger |\Phi\rangle \\
 |\Phi_i\rangle &\equiv \hat{a}_i |\Phi\rangle
 \end{aligned} \tag{2.13}$$

Using these definitions, hole-creation and particle-annihilation operators vanish when acting on the Fermi vacuum from the left, $\hat{a}_i^\dagger |\Phi_0\rangle = \hat{a}_i |\Phi_0\rangle = 0$. Conversely, hole-annihilation and particle-creation operators vanish when acting on the Fermi vacuum from the right, $\langle \Phi_0 | \hat{a}_i = \langle \Phi_0 | \hat{a}_a^\dagger = 0$.

These results can be exploited to simplify expressions involving strings of creation and annihilation operators by a procedure called *normal ordering* with respect to the Fermi vacuum. Denoted by $\{\cdots\}$, normal ordering permutes a string of creation and annihilation operators so that hole-annihilation and particle-creation operators are to the left of hole-

creation and particle-annihilation operators, which guarantees that normal ordered operators vanish on the Fermi vacuum, $\langle \Phi_0 | \{ \cdots \} = 0$ and $\{ \cdots \} | \Phi_0 \rangle = 0$.

$$\left\{ \hat{a}_j^\dagger \cdots \hat{a}_i \cdots \hat{a}_b \cdots \hat{a}_a^\dagger \right\} = (-1)^\sigma \hat{a}_i \cdots \hat{a}_a^\dagger \cdots \hat{a}_j^\dagger \cdots \hat{a}_b, \quad (2.14)$$

where σ is the number of two-state permutations required to do the normal ordering.

2.4 Wick's Theorem

At this point, the many-body problem has been reduced to computing long strings of creation and annihilation operators between the normal-ordered Hamiltonian and the correlated wave function using Eq. (2.8). Instead of using a brute-force approach by permuting over and over, a further simplification known as *Wick's theorem* [132] can be introduced. A Wick contraction of two operators with respect to the reference state is defined as

$$\overline{\hat{A}\hat{B}} = \hat{A}\hat{B} - \left\{ \hat{A}\hat{B} \right\}. \quad (2.15)$$

Which, given the definition in Eq. (2.14) and the anticommutation relations int Eq. (2.8), means that the only nonzero contractions are of the form,

$$\overline{\hat{a}_i^\dagger \hat{a}_j} = \delta_{ij} \quad \text{and} \quad \overline{\hat{a}_a \hat{a}_b^\dagger} = \delta_{ab}. \quad (2.16)$$

Because contracted operators simply represent a Kronecker delta, they can be removed from a normal ordered product by permuting the product σ times so that the contracted operators

are next to each other,

$$\{\hat{A} \cdots \hat{B} \cdots \hat{C} \cdots \hat{D}\} = (-1)^\sigma \{\hat{A} \cdots \hat{B} \hat{C} \cdots \hat{D}\} = (-1)^\sigma \hat{B} \hat{C} \{\hat{A} \cdots \hat{D}\}. \quad (2.17)$$

These different definitions for operator manipulation come together to define the time-independent Wick's theorem, which reformulates a product of operators as the sum of its normal-ordered form and all possible contractions of its normal-ordered form.

$$\hat{A} \hat{B} \hat{C} \cdots = \left\{ \hat{A} \hat{B} \hat{C} \cdots \right\} + \sum_{\text{one-contractions}} \left\{ \hat{A} \hat{B} \hat{C} \cdots \right\} + \sum_{\text{two-contractions}} \left\{ \hat{A} \hat{B} \hat{C} \cdots \right\} + \cdots + \sum_{\text{all-contractions}} \left\{ \hat{A} \hat{B} \hat{C} \cdots \right\} \quad (2.18)$$

Wick's theorem is incredibly useful in many-body techniques because complicated expressions of operators can be expressed as diagrams that are easy to compute with simple diagrammatic rules which correspond to Eqs. (2.8), (2.16), and (2.17). These diagrammatic techniques are an integral component to deriving expressions used in this work, and their underlying rules are summarized in ?? and are extensively discussed in [108].

A powerful application of Wick's theorem is to rewrite the Hamiltonian in Eq. (2.2) as a sum of normal-ordered operators in the form of Eq. (2.18),

$$\hat{H} = E_0 + \sum_{pq} f_q^p \left\{ \hat{a}_p^\dagger \hat{a}_q \right\} + \frac{1}{4} \sum_{pqrs} V_{rs}^{pq} \left\{ \hat{a}_p^\dagger \hat{a}_q^\dagger \hat{a}_s \hat{a}_r \right\} + \frac{1}{36} \sum_{pqrstu} W_{stu}^{pqr} \left\{ \hat{a}_p^\dagger \hat{a}_q^\dagger \hat{a}_r^\dagger \hat{a}_u \hat{a}_t \hat{a}_s \right\} + \cdots. \quad (2.19)$$

The first component, E_0 , contains fully-contracted terms, and because the Hamiltonian operators are ordered so that the creation operators appear before the annihilations operators, only terms that contract *hole* states in the form $\{\cdots \hat{a}_i^\dagger \cdots \hat{a}_j \cdots\}$ are nonzero. Therefore, the zero-body component of the normal ordered Hamiltonian can be written as a

sums over all hole states for each component of the original Hamiltonian,

$$E_0 = \sum_i {}^{(1)}H_i^i + \frac{1}{2} \sum_{ij} {}^{(2)}H_{ij}^{ij} + \frac{1}{6} \sum_{ijk} {}^{(3)}H_{ijk}^{ijk} \dots . \quad (2.20)$$

In a compact diagrammatic form, this sum can be drawn as the sum of connected vertices ($\bullet\cdots\cdots\otimes$, $\bullet\cdots\cdots\bullet$, etc...) corresponding to the components of the original Hamiltonian in Eq. (2.2),

$$E_0 = \text{Diagram } 1 + \text{Diagram } 2 + \text{Diagram } 3 + \dots . \quad (2.21)$$

The number of lines connected to each vertex defines its type such that the single vertex corresponds to the one-body Hamiltonian, the double vertex corresponds to the two-body Hamiltonian, etc... The looped lines represent a sum over all hole states,

The second component of the normal-ordered Hamiltonian, $f_q^p \left\{ \hat{a}_p^\dagger \hat{a}_q \right\}$, contains terms that have all operators contracted except for one pair. Like the fully-contracted term, these contractions must involve only hole states,

$$f_q^p = {}^{(1)}H_q^p + \sum_i {}^{(2)}H_{qi}^{pi} + \frac{1}{2} \sum_{ij} {}^{(3)}H_{qij}^{pij} + \dots . \quad (2.22)$$

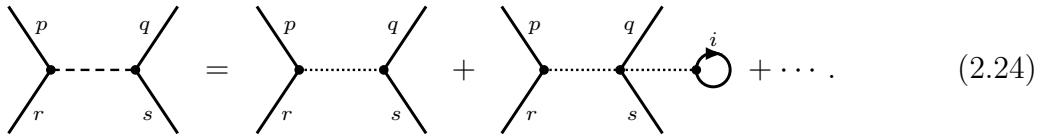
In diagrammatic notation, the uncontracted pair of operators is represented as two external lines connected to each vertex which, because they are generic states, are drawn as unoriented lines,

$$\text{Diagram } 1 = \text{Diagram } 1 + \text{Diagram } 2 + \text{Diagram } 3 + \dots . \quad (2.23)$$

When a line represents a particle state, it will contain an arrow directed upwards while a line representing a hole state will contain an arrow directed downwards.

The two-body term, $V_{rs}^{pq} \left\{ \hat{a}_p^\dagger \hat{a}_q^\dagger \hat{a}_s \hat{a}_r \right\}$, follows in the same manner such that it represents all terms of the original Hamiltonian with all but two pairs of operators contracted. Like the zero-body and one-body terms, the two-body term contains the original two-body term $(^2)\hat{H}$ as well as density-dependent terms that sum over hole states in higher-body Hamiltonian terms, leaving four external lines for each diagram vertex,

$$V_{rs}^{pq} = (^2)H_{rs}^{pq} + \sum_i (^3)H_{rsi}^{pqi} + \dots$$



Three- and four-body normal-ordered terms can also be calculated by following the same procedure, but in practice are truncated at the two- or three-body level. Because normal ordering the Hamiltonian has the effect of shuffling higher-order interactions into lower-order terms, and makes it feasible to include computationally expensive many-body interactions as normal-ordered few-body interactions. Also, it reorganizes many-body correlations into the reference state so that additional correlations around the Fermi surface can be treated as a perturbation. Therefore, from this point forward, the many-body problem will be formulated in terms of the normal-ordered Hamiltonian, and the bare interactions will be truncated beyond the three-body level for computational feasibility. Electronic systems are naturally truncated at the the level of the two-body Coulomb interaction, while nuclear systems can be successfully described with the two-body normal-ordered piece of the three-body force, in the form of Eq. (2.24).

The normal-ordered Hamiltonian (Eq. (2.19)) is written as a sum of the *reference energy*, E_0 , which is the fully-contracted expectation value of the Hamiltonian with respect to the reference state,

$$E_0 = \langle \Phi | \hat{H} | \Phi \rangle, \quad (2.25)$$

and the remaining normal-ordered pieces of the Hamiltonian, \hat{H}_N . Rewriting the many-body Schrödinger equation for the ground state, $|\Psi\rangle$, and using this partition gives,

$$\begin{aligned} \hat{H}|\Psi\rangle &= (E_0 + \hat{H}_N)|\Psi\rangle = E|\Psi\rangle \\ \longrightarrow \hat{H}_N|\Psi\rangle &= (E - E_0)|\Psi\rangle = \Delta E|\Psi\rangle, \end{aligned} \quad (2.26)$$

where ΔE is known as the *correlation energy*.

Now that the many-body quantum problem has been formulated, different approaches to solving that problem can be proposed and analyzed. Because taking account of correlations from all particles simultaneously is a demanding—and for some systems, computationally impossible—endeavor, methods for solving the many-body Schrödinger equation should be systematically improvable. Successful methods with this quality incorporate the most dominant correlations in lower-order solutions and approach the exact solution when more and more orders are included.

2.5 Hartree–Fock Method

A successful, first-order approximation to any many-body method comes from noticing that each individual particle feels a mean-field potential from the cumulative interactions with all the other particles. The *Hartree-Fock* (HF) method [59, 46] aims to transform the original

single-particle basis to a Hartree-Fock basis where each orbital is the eigenfunction of its corresponding mean-field. Because the transformation of a single orbital changes its effect on every other particle, this process must be performed iteratively until self-consistency between all the orbitals is reached, which is why this method is also known as the *Self-Consistent Field* (SCF) method.

This mean-field picture results from the following procedure. It begins by minimizing the reference energy with respect to the reference state. This functional is just the zero-body piece of the normal-ordered Hamiltonian,

$$E_{\text{HF}} [\Phi_0] = \langle \Phi_0 | \hat{H} | \Phi_0 \rangle = \sum_i {}^{(1)}H_i^i + \frac{1}{2} \sum_{ij} {}^{(2)}H_{ij}^{ij} + \frac{1}{6} \sum_{ijk} {}^{(3)}H_{ijk}^{ijk}. \quad (2.27)$$

Transforming the reference determinant can be accomplished by rotating the state within the single-particle basis by use of the *Thouless theorem* [120], which states that any Slater determinant can be written as the product of any other Slater determinant and an exponentiated single-excitation operator,

$$|\Phi'\rangle = e^{\hat{C}_1} |\Phi_0\rangle, \quad \text{where } \hat{C}_1 = \sum_{ai} C_i^a \left\{ \hat{a}_a^\dagger \hat{a}_i \right\}. \quad (2.28)$$

If the difference between the two Slater determinants is dominated by single excitations, this transformation can be approximated by expanding the exponential and ignoring higher-order terms,

$$|\Phi'\rangle \simeq \left(1 + \sum_{ai} C_i^a \left\{ \hat{a}_a^\dagger \hat{a}_i \right\} \right) |\Phi_0\rangle. \quad (2.29)$$

The reference energy functional can now be written as a sum of the original reference

state and new terms that incorporate the single-excitation variation,

$$E_{\text{HF}} [\Phi'] = \langle \Phi' | \hat{H} | \Phi' \rangle \simeq E_{\text{HF}} [\Phi_0] + \sum_{ai} C_i^a \langle \Phi_0 | \hat{H} | \Phi_i^a \rangle + \sum_{ai} C_i^{a*} \langle \Phi_i^a | \hat{H} | \Phi_0 \rangle. \quad (2.30)$$

The minimum of this functional is found by differentiating the expression with respect to the coefficients C_i^a and setting the result to zero,

$$\delta E_{\text{HF}} [\Phi'] \simeq \sum_{ai} \delta C_i^a \langle \Phi_0 | \hat{H} | \Phi_i^a \rangle + \sum_{ai} \delta C_i^{a*} \langle \Phi_i^a | \hat{H} | \Phi_0 \rangle = 0. \quad (2.31)$$

Because this expression is Hermitian, both terms must vanish independently so that,

$$\langle \Phi_0 | \hat{H} | \Phi_i^a \rangle = \langle \Phi_i^a | \hat{H} | \Phi_0 \rangle = 0. \quad (2.32)$$

This condition is the result of the *Brillouin theorem* [19], which states that the Hamiltonian matrix element must vanish between an optimized Hartree-Fock ground state and any single excitation from it. The Brillouin condition is satisfied by diagonalizing the one-body piece of the normal-ordered Hamiltonian f_q^p (Eq. (2.22)), known as the *Fock* operator, such that off-diagonal pieces like $\langle \Phi_0 | \hat{H} | \Phi_i^a \rangle = f_a^i$ and $\langle \Phi_i^a | \hat{H} | \Phi_0 \rangle = f_i^a$ vanish. Diagonalizing the Fock operator can be written schematically as,

$$\varepsilon_q^p \delta_{pq} \leftarrow (1)H_q^p + \sum_i (2)H_{qi}^{pi} + \frac{1}{2} \sum_{ij} (3)H_{qij}^{pi,j}$$

$$(2.33)$$

where ε_q^p is the eigenvalue of the Fock operator, and its diagrammatic form vanishes when

the external indices differ.

A practical way of solving this system of equations is to express each new orbital in the unknown Hartree-Fock basis, $|p'\rangle \equiv \phi_{p'}(\mathbf{r})$, denoted with primed labels, as a linear combination of the known single-particle basis states, $|p\rangle \equiv \phi_p(\mathbf{r})$, denoted without primed labels. These two bases are related by a unitary transformation $C_{p'}^p \equiv \langle p|p'\rangle$,

$$|p'\rangle = \sum_p \langle p|p'\rangle |p\rangle = \sum_p C_{p'}^p |p\rangle. \quad (2.34)$$

Then the Fock matrix can be written in terms of the Hartree-Fock basis,

$$\begin{aligned} f_{q'}^{p'} &= {}^{(1)}H_{q'}^{p'} + \sum_{i'} {}^{(2)}H_{q'i'}^{p'i'} + \frac{1}{2} \sum_{i'j'} {}^{(3)}H_{q'i'j'}^{p'i'j'} \\ &= \sum_{pq} C_p^{p'*} {}^{(1)}H_q^p C_{q'}^q + \sum_{\substack{i' \\ prqs}} C_p^{p'*} C_r^{i'*} {}^{(2)}H_{qs}^{pr} C_{q'}^q C_{i'}^s + \frac{1}{2} \sum_{\substack{i'j' \\ prsqtu}} C_p^{p'*} C_r^{i'*} C_s^{j'*} {}^{(3)}H_{qtu}^{prs} C_{q'}^q C_{i'}^t C_{j'}^u. \end{aligned} \quad (2.35)$$

Defining the first-order density matrix γ_q^p as the product of expansion coefficients, summed over all shared hole states,

$$\gamma_q^p = \sum_{i'} C_{i'}^p C_{q'}^{i'*}, \quad (2.36)$$

Eq. (2.34) is simplified to,

$$f_{q'}^{p'} = \sum_{pq} C_p^{p'*} \left[{}^{(1)}H_q^p + \sum_{rs} \gamma_s^r {}^{(2)}H_{qs}^{pr} + \frac{1}{2} \sum_{rstu} \gamma_t^r \gamma_u^s {}^{(3)}H_{qtu}^{prs} \right] C_{q'}^q \rightarrow \varepsilon_{q'}^{p'} \delta_{p'q'}. \quad (2.37)$$

Therefore, the Hartree-Fock equations are ultimately expressed as an eigenvalue problem

according to Eq. (2.5) where the matrix to diagonalize is the Fock matrix in the form,

$$\hat{F}_q^p(\hat{C}) = {}^{(1)}H_q^p + \sum_{rs} \gamma_r^s {}^{(2)}H_{qs}^{pr} + \frac{1}{2} \sum_{rstu} \gamma_r^t \gamma_s^u {}^{(3)}H_{qtu}^{prs}, \quad (2.38)$$

and the matrix of coefficients, $\hat{C} = C_{p'}^p$, is the unitary operator that transforms the matrix to a diagonal form,

$$\sum_{pq} C_p^{p'*} F_q^p(\hat{C}) C_{q'}^q = \varepsilon_{q'}^{p'} \delta_{p'q'} \quad (2.39)$$

The iterative nature of the solution comes from the dependence of the Fock matrix on the transformation coefficients. These Hartree-Fock equations are solved numerically by using an iterative algorithm where the Fock matrix is built using a known set of coefficients and diagonalized to obtain an updated set of coefficients. This process is repeated until the unitary set of coefficients is unchanged within a certain tolerance. For most calculations, using the identity matrix as an initial guess for the coefficients is sufficient. To improve the rate of convergence, techniques such as the direct inversion of the iterative subspace (DIIS) [100, 99] or Broyden's method [21] can be implemented. And, to avoid any oscillatory behavior around the solution, techniques such as the level-shifting method or *ad hoc* linear mixing can be implemented to dampen the large changes between iterations.

To make use of the HF solution as the reference state for post-HF calculations, the Hamiltonian matrix elements must be transformed to the new basis and the normal-ordered version redefined to account for the additional reordering of one-particle correlations into the HF energy. The one-body piece from Eq. (2.22) is simply the resulting eigenvalues of the diagonalized Fock matrix,

$$f_{q'}^{p'} = \varepsilon_{q'}^{p'} \delta_{p'q'} \quad (2.40)$$

For the two-body term, Eq. (2.24), first the density-dependent component of the three-body

interaction is written with the first-order density matrix. Then the remaining states are transformed according to Eq. (2.34),

$$V_{r's'}^{p'q'} = \sum_{pqrs} C_p^{p'*} C_q^{q'*} \left({}^{(2)}H_{rs}^{pq} + {}^{(3)}H_{rsu}^{pqt} \gamma_t^u \right) C_{r'}^r C_{s'}^s. \quad (2.41)$$

The reference energy from Eqns. (2.20) and (2.25) can be written in terms of the transformed one- and two-body Hamiltonian, Eqns. (2.22) and (2.24), and the original three-body term using first-order density matrices,

$$E_0 = \sum_{i'} \varepsilon_{i'}^{i'} - \frac{1}{2} \sum_{i'j'} V_{i'j'}^{i'j'} + \frac{1}{6} \sum_{pqrstu} {}^{(3)}H_{stu}^{pqr} \gamma_p^s \gamma_q^t \gamma_u^r$$

Additionally, any operators that are constructed in the original basis must be transformed in a similar manner. For example, a one-body operator \hat{O} in the Hartree-Fock basis is,

$$\begin{aligned} \hat{O} &= \sum_{p'q'} O_{q'}^{p'} \left\{ \hat{a}_{p'}^\dagger \hat{a}_{q'} \right\} = \sum_{p'q'pq} C_p^{p'*} O_q^p C_{q'}^q \left\{ \hat{a}_{p'}^\dagger \hat{a}_{q'} \right\} \\ &\longrightarrow O_{q'}^{p'} = \sum_{pq} C_p^{p'*} O_q^p C_{q'}^q. \end{aligned} \quad (2.43)$$

Because the Hartree-Fock basis is diagonal in the one-body piece of the Hamiltonian, any terms that include off-diagonal elements automatically vanish, greatly simplifying any post-Hartree-Fock methods. From this point on, all calculations will use the Hartree-Fock basis unless stated otherwise, and prime symbols will be omitted.

2.6 Configuration-Interaction

The most generic way to write a correlated wave function in a given basis is as a linear combination of all possible Slater determinants. In normal-ordered form, this expansion can, in principle, consist of the $0p$ - $0h$ reference state and all possible Np - Nh excitations up to Ap - Ah excitations,

$$|\Psi_\nu\rangle = \sum_{\nu_i}^{\mathcal{N}} C_{\nu_i} |\Phi_{\nu_i}\rangle = C_0 |\Phi_0\rangle + \sum_{N=1}^A \left(\frac{1}{N!}\right)^2 \sum_{\substack{a_1 \dots a_N \\ i_1 \dots i_N}} C_{i_1 \dots i_N}^{a_1 \dots a_N} |\Phi_{i_1 \dots i_N}^{a_1 \dots a_N}\rangle. \quad (2.44)$$

Using this form of the wave function in Eq. (2.44), the Schrödinger equation can be reformulated as a standard matrix eigenvalue problem,

$$\begin{aligned} \hat{H}_N |\Psi_\nu\rangle &= \Delta E_\nu |\Psi_\nu\rangle \quad \longrightarrow \quad \langle \Psi_\mu | \hat{H}_N | \Psi_\nu \rangle = \Delta E_\nu \langle \Psi_\mu | \Psi_\nu \rangle \\ &= \sum_{\mu_i \nu_i} C_{\mu_i}^* \langle \Phi_{\mu_i} | \hat{H}_N | \Phi_{\nu_i} \rangle C_{\nu_i} = \Delta E_\nu \sum_{\mu_i \nu_i} C_{\mu_i}^* C_{\nu_i} \delta_{\mu_i \nu_i} \\ &\longrightarrow \mathbf{C}_\mu^T \left(\langle \Phi_{\mu_i} | \hat{H}_N | \Phi_{\nu_i} \rangle - \Delta E_\nu \mathbf{I} \right) \mathbf{C}_\nu = 0. \end{aligned} \quad (2.45)$$

In this case, the matrix elements are Hamiltonian terms that connect two Slater determinants, and the eigenvectors are the ground and excited states in the form of Eq. (2.44). The matrix elements can be found with the help of the Slater-Condon rules [111, 33] which, because the Hamiltonian is restricted to one- and two-body terms, require that any terms connecting Slater determinants which differ by more than two single-particle states vanish. Also, because the one-body Hamiltonian is diagonal in the Hartree-Fock basis, it only contributes to diagonal elements the CI matrix. Some examples of these matrix elements

are,

$$\langle \Phi_i^a | \hat{H} | \Phi_i^a \rangle = \varepsilon_a - \varepsilon_i - V_{ia}^{ia} \quad (2.46)$$

$$\langle \Phi_{ij}^{ab} | \hat{H} | \Phi_{ij}^{cd} \rangle = V_{cd}^{ab} \quad (2.47)$$

$$\langle \Phi_{ijk}^{abc} | \hat{H} | \Phi_{ijl}^{abd} \rangle = -V_{kd}^{lc} \quad (2.48)$$

Because the configuration-interaction method exhaustively captures all the correlations of a many-body system, it is considered an “exact” method within a certain model space and becomes truly exact as the number of single-particle states is increased to infinity. However, there is a price to pay for this exactness. The number of Slater determinants, \mathcal{N} , in model space scales factorially according to Eq. (2.5) and the configuration-interaction matrix scales as \mathcal{N}^2 . For sufficiently-sized model spaces, the memory required for this matrix quickly becomes unimaginable even for the largest supercomputers, see Fig. 2.3.

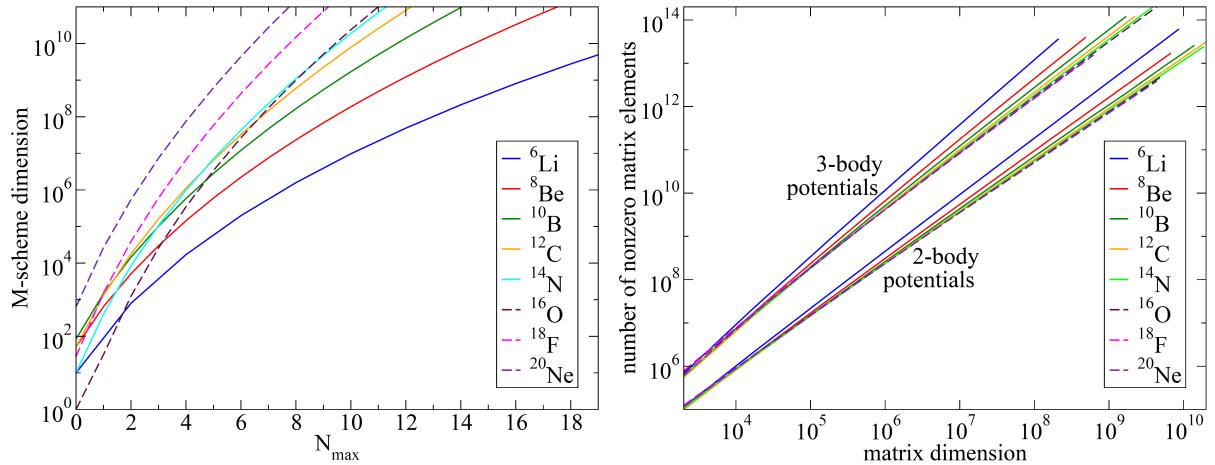


Figure 2.3: Scaling of the matrix size and number of non-zero matrix elements for nuclear CI calculations of light nuclei. Even for modest N_{\max} , the memory requirements approach the limit of petascale supercomputers (10^{10}). Figure taken from [107].

However, for a reference state that is a good approximation to the true ground state, few-body excitations generally dominate the wave functions for low-lying states [109]. This

can be exploited by truncating the expansion in Eq. (2.44). Owing to the two-body nature of the interaction, the lowest appropriate truncation is also at the two-body level, known as configuration interaction with singles and doubles (CISD),

$$|\Psi_\nu\rangle = C_0 |\Phi_0\rangle + \sum_{ai} C_i^a |\Phi_i^a\rangle + \frac{1}{4} \sum_{abij} C_{ij}^{ab} |\Phi_{ij}^{ab}\rangle. \quad (2.49)$$

This is a very straightforward and tractable way to approximate the many-body Schrödinger equation, and it can be systematically improved by adding more excitations such as triples (CISDT) or triples and quadruples (CISDTQ). But the drawback to this simplicity is that any truncated CI method is not size-extensive such that any extensive property of a system, like the energy, would scale with the size of the system. A desirable many-body method will be both systematically improvable and size-extensive while maintaining computational feasibility.

2.7 Many-Body Perturbation Theory

One many-body method that is both size-extensive and systematically improvable treats particle-particle interactions as a perturbation to the mean-field potential and is known as many-body perturbation theory (MBPT) [88, 66, 67, 108]. The Hamiltonian is partitioned into a diagonal piece and the interaction piece,

$$\begin{aligned} \hat{H} &= \hat{H}_0 + \hat{V}, \quad \text{with} \\ \hat{H}_0 &= E_0 + \sum_p f_p^p \left\{ \hat{a}_p^\dagger \hat{a}_p \right\} \quad \text{and} \\ \hat{V} &= \frac{1}{4} \sum_{pqrs} V_{rs}^{pq} \left\{ \hat{a}_p^\dagger \hat{a}_q^\dagger \hat{a}_s \hat{a}_r \right\} \end{aligned} \quad (2.50)$$

When not in the Hartree-Fock basis, the interaction piece has the additional off-diagonal Fock term, $\sum_{p \neq q} f_q^p \left\{ \hat{a}_p^\dagger \hat{a}_q \right\}$. This means that the reference state is an eigenstate of the zero-order piece of the Hamiltonian,

$$\hat{H}_0 |\Phi_0\rangle = \left(E_0 + \sum_i f_i^i \left\{ \hat{a}_i^\dagger \hat{a}_i \right\} \right) |\Phi_0\rangle = \left(E_0 + \sum_i \varepsilon_i \right) |\Phi_0\rangle = E_0^{(0)} |\Phi_0\rangle. \quad (2.51)$$

Using *intermediate normalization*, which sets $\langle \Phi_0 | \Psi \rangle = 1$, the Schrödinger equation, Eq. (2.1) for the ground state becomes,

$$\begin{aligned} \langle \Phi_0 | (\hat{H}_0 + \hat{V}) | \Psi \rangle &= \langle \Phi_0 | \hat{H}_0 | \Psi \rangle + \langle \Phi_0 | \hat{V} | \Psi \rangle = E \langle \Phi | \Psi \rangle \\ &= E^{(0)} \langle \Phi | \Psi \rangle + \langle \Phi_0 | \hat{V} | \Psi \rangle = E^{(0)} + \Delta E_0 = E, \end{aligned} \quad (2.52)$$

where the energy difference is $\Delta E_0 \equiv \langle \Phi_0 | \hat{V} | \Psi \rangle$.

Next, the projection operators \hat{P} and \hat{Q} can be introduced,

$$\hat{P} = |\Phi_0\rangle\langle\Phi_0|, \quad (2.53)$$

$$\hat{Q} = \sum_{n \neq 0} |\Phi_n\rangle\langle\Phi_n| = 1 - |\Phi_0\rangle\langle\Phi_0|. \quad (2.54)$$

The \hat{P} operator isolates the reference-state component of any Slater determinant while the \hat{Q} operator isolates all components *except* the reference-state component out of any Slater determinant. Both these operators are idempotent, which means that $\hat{P}^2 = \hat{P}$ and $\hat{Q}^2 = \hat{Q}$, and because of intermediate normalization, the correlated wave function can be written as $|\Psi\rangle = (\hat{P} + \hat{Q})|\Psi\rangle = |\Phi\rangle + \hat{Q}|\Psi\rangle$. Also, both operators commute with the unperturbed part of the Hamiltonian, $\hat{H}_0 \hat{P} = \hat{P} \hat{H}_0$ and $\hat{H}_0 \hat{Q} = \hat{Q} \hat{H}_0$. These identities can be applied to an alternate version of the Schrödinger equation which defines a particular version of

perturbation theory known as Raleigh-Schrödinger perturbation theory (RSPT) [101, 106].

In this version, the zeroth-order energy $E^{(0)}$ is added to both sides of the Schrödinger equation. Acting with \hat{Q} and rearranging terms gives,

$$\begin{aligned}\hat{Q}(E^{(0)} - \hat{H}_0)|\Psi\rangle &= \hat{Q}(E^{(0)} + \hat{V} - E)|\Psi\rangle \\ \hat{Q}(E^{(0)} - \hat{H}_0)\hat{Q}|\Psi\rangle &= \hat{Q}(\hat{V} - \Delta E_0)|\Psi\rangle,\end{aligned}\quad (2.55)$$

where $\Delta E_0 \equiv E - E^{(0)} = \langle \Phi_0 | \hat{V} | \Psi \rangle$. The operator $\hat{Q}(E^{(0)} - \hat{H}_0)\hat{Q}$ is invertible because $(E^{(0)} - \hat{H}_0)^{-1}$ is never singular in Q -space. Therefore, the operator $\hat{R}_0 = \hat{Q}(E^{(0)} - \hat{H}_0)^{-1}\hat{Q}$, known as the *resolvent*, can be applied to both sides to result in the generating equation for RSPT,

$$\begin{aligned}\hat{Q}(E^{(0)} - \hat{H}_0)^{-1}(E^{(0)} - \hat{H}_0)\hat{Q}|\Psi\rangle &= \hat{Q}(E^{(0)} - \hat{H}_0)^{-1}\hat{Q}(\hat{V} - \Delta E_0)|\Psi\rangle \\ \hat{Q}|\Psi\rangle &= \hat{R}_0(\hat{V} - \Delta E_0)|\Psi\rangle \\ |\Psi\rangle &= |\Phi\rangle + \hat{R}_0(\hat{V} - \Delta E_0)|\Psi\rangle\end{aligned}\quad (2.56)$$

Because the single-particle states are eigenfunctions of the zeroth-order Hamiltonian, they are also eigenfunctions of the resolvent, with the resulting eigenvalues, ε , are known as *energy denominators*. Applying the resolvent operator to any orthogonal state to the reference determinant in the form of Eq. (2.13) gives the following relation,

$$\begin{aligned}\hat{R}_0|\Phi_{i_1 \dots i_N}^{a_1 \dots a_N}\rangle &= \frac{1}{\varepsilon_{i_1 \dots i_N}^{a_1 \dots a_N}}|\Phi_{i_1 \dots i_N}^{a_1 \dots a_N}\rangle, \quad \text{where} \\ \varepsilon_{i_1 \dots i_N}^{a_1 \dots a_N} &= \varepsilon_{i_1} + \dots + \varepsilon_{i_N} - \varepsilon_{a_1} - \dots - \varepsilon_{a_N}.\end{aligned}\quad (2.57)$$

Equation (2.56) can be iterated infinitely to give the solution for the fully correlated wave function,

$$|\Psi\rangle = \sum_{n=0}^{\infty} \left[\hat{R}_0 (\hat{V} - \Delta E_0) \right]^n |\Phi_0\rangle. \quad (2.58)$$

Applying this form of the correlated wave function into Eq. (2.52) results in the energy difference,

$$\Delta E_0 = \langle \Phi_0 | \hat{V} | \Psi \rangle = \sum_{n=0}^{\infty} \langle \Phi_0 | \hat{V} \left[\hat{R}_0 (\hat{V} - \Delta E_0) \right]^n |\Phi_0\rangle \quad (2.59)$$

The immediate problem with this equation is that the right-hand side of the equations contain the target energy difference ΔE_0 for which these equations are meant to solve. This can be remedied by expanding the right-hand sides and rearranging terms. Using the fact that $\hat{R}_0 \Delta E_0 |\Phi_0\rangle = \Delta E_0 \hat{R}_0 |\Phi_0\rangle = 0$, the first-order energy $E^{(1)} = \langle \Phi_0 | \hat{V} | \Phi_0 \rangle$, and the shifted term $\tilde{V} \equiv \hat{V} - E^{(1)}$, these simplify to,

$$|\Psi\rangle - |\Phi_0\rangle = \hat{R}_0 \hat{V} |\Phi_0\rangle + \hat{R}_0 \tilde{V} \hat{R}_0 \hat{V} |\Phi_0\rangle \\ + \hat{R}_0 \tilde{V} \hat{R}_0 \tilde{V} \hat{R}_0 \hat{V} |\Phi_0\rangle - \langle \Phi_0 | \hat{V} \hat{R}_0 \hat{V} |\Phi_0\rangle \hat{R}_0^2 \hat{V} |\Phi_0\rangle + \dots \quad (2.60)$$

$$\Delta E_0 = \langle \Phi_0 | \hat{V} | \Phi_0 \rangle + \langle \Phi_0 | \hat{V} \hat{R}_0 \hat{V} | \Phi_0 \rangle + \langle \Phi_0 | \hat{V} \hat{R}_0 \tilde{V} \hat{R}_0 \hat{V} | \Phi_0 \rangle \\ + \langle \Phi_0 | \hat{V} \hat{R}_0 \tilde{V} \hat{R}_0 \tilde{V} \hat{R}_0 \hat{V} | \Phi_0 \rangle - \langle \Phi_0 | \hat{V} \hat{R}_0 \hat{V} | \Phi_0 \rangle \langle \Phi_0 | \hat{V} \hat{R}_0^2 \hat{V} | \Phi_0 \rangle + \dots \quad (2.61)$$

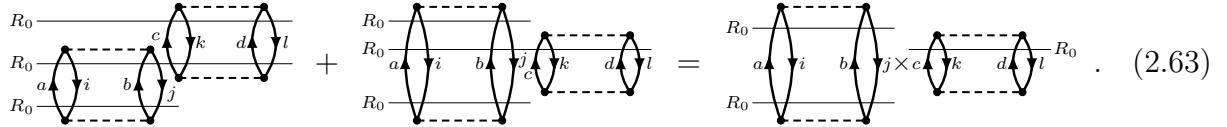
The order of each term can be easily identified by counting the numbers of times that \hat{V} or \tilde{V} appears. At the third order in the wave function and the fourth order in the energy, *renormalization* terms make their first appearance. These terms contain separated and closed factors in the form of lower-order energy terms, such as $\langle \Phi_0 | \hat{V} \hat{R}_0 \hat{V} | \Phi_0 \rangle \equiv E^{(2)}$. Terms that do not contain normalization factors are known as *principal* terms.

2.7.1 Factorization Theorem

A powerful application of diagrammatic techniques known as the *factorization theorem* [67, 47, 18] can immediately be used to simplify these expansions. By factoring sums of *unlinked* diagrams, where two or more parts of a diagram are closed and separated, from the principal terms, it can be shown that they exactly cancel with the renormalization terms at each order. In the following factorization, two fourth-order energy diagrams which differ by only the time-ordering of the interaction vertices are added together. By multiplying each term by an appropriate factor so that they share a common denominator, the additive property of the energy denominators ($\varepsilon_{ij}^{ab} + \varepsilon_{kl}^{cd} = \varepsilon_{ijkl}^{abcd}$) can be exploited to remove the addition of both terms, now written as the product of two terms,

$$\begin{aligned} \frac{1}{16} \sum_{\substack{abcd \\ i j k l}} \frac{V_{ab}^{ij} V_{ij}^{ab} V_{cd}^{kl} V_{kl}^{cd}}{\varepsilon_{ij}^{ab} \varepsilon_{ijkl}^{abcd} \varepsilon_{kl}^{cd}} + \frac{1}{16} \sum_{\substack{abcd \\ i j k l}} \frac{V_{ab}^{ij} V_{ij}^{ab} V_{cd}^{kl} V_{kl}^{cd}}{\varepsilon_{ij}^{ab} \varepsilon_{ijkl}^{abcd} \varepsilon_{ij}^{ab}} &= \frac{1}{16} \sum_{\substack{abcd \\ i j k l}} V_{ab}^{ij} V_{ij}^{ab} V_{cd}^{kl} V_{kl}^{cd} \frac{\varepsilon_{ij}^{ab} + \varepsilon_{kl}^{cd}}{\left(\varepsilon_{ij}^{ab}\right)^2 \varepsilon_{ijkl}^{abcd} \varepsilon_{kl}^{cd}} \\ &= \frac{1}{4} \sum_{abi j} \frac{V_{ab}^{ij} V_{ij}^{ab}}{\left(\varepsilon_{ij}^{ab}\right)^2} \cdot \frac{1}{4} \sum_{cdkl} \frac{V_{cd}^{kl} V_{kl}^{cd}}{\varepsilon_{kl}^{cd}} = \langle \Psi_n^{(1)} | \Psi_n^{(1)} \rangle E_n^{(2)}. \end{aligned} \quad (2.62)$$

In diagrammatic form, these sums are represented by internal lines between vertices. The common resolvent in each term, drawn as a line through the relevant state, is removed, and the common diagrams which result are shown as a product (see [108] for more details),



A similar factorization can be performed on the wave function terms. The following example uses two similar third-order terms with different time-ordered interaction vertices. Once again, the additive property of the energy denominators is used to factor the common

denominator between the terms, resulting in the product of two lower-order terms,

$$\begin{aligned}
& \frac{1}{16} \sum_{\substack{abcd \\ i j k l}} \frac{V_{ij}^{ab} V_{cd}^{kl} V_{kl}^{cd}}{\varepsilon_{kl}^{cd} \varepsilon_{abcd} \varepsilon_{ij}^{ab}} |\Phi_{ij}^{ab}\rangle + \frac{1}{16} \sum_{\substack{abcd \\ i j k l}} \frac{V_{ij}^{ab} V_{cd}^{kl} V_{kl}^{cd}}{\varepsilon_{ij}^{ab} \varepsilon_{ijkl} \varepsilon_{ij}^{ab}} |\Phi_{ij}^{ab}\rangle = \frac{1}{16} \sum_{\substack{abcd \\ i j k l}} V_{ij}^{ab} V_{cd}^{kl} V_{kl}^{cd} \frac{\varepsilon_{ij}^{ab} + \varepsilon_{kl}^{cd}}{\left(\varepsilon_{ij}^{ab}\right)^2 \varepsilon_{ijkl}^{abcd} \varepsilon_{kl}^{cd}} |\Phi_{ij}^{ab}\rangle \\
& = \frac{1}{4} \sum_{abij} \frac{V_{ij}^{ab}}{\left(\varepsilon_{ij}^{ab}\right)^2} |\Phi_{ij}^{ab}\rangle \cdot \frac{1}{4} \sum_{cdkl} \frac{V_{cd}^{kl} V_{kl}^{cd}}{\varepsilon_{kl}^{cd}} = \frac{|\Psi_n^{(1)}\rangle}{\varepsilon_n} E_n^{(2)}
\end{aligned}$$
(2.64)

The factorization theorem is also valid with off-diagonal Fock terms and applies to the MBPT expansions of both the wave function and energy. Therefore, the MBPT expansions in Eqs. (2.58) and (2.59) can be written in terms of *linked diagrams* only [1],

$$|\Psi\rangle = \sum_{n=0}^{\infty} \left[\hat{R}_0 (\hat{V} - \Delta E_0) \right]^n |\Phi_0\rangle_L, \quad (2.65)$$

$$\Delta E_0 = \sum_{n=0}^{\infty} \langle \Phi_0 | \hat{V} \left[\hat{R}_0 (\hat{V} - \Delta E_0) \right]^n | \Phi_0 \rangle_L, \quad (2.66)$$

where “L” denotes that no diagrams with closed, disconnected pieces should be included. This result not only simplifies the MBPT expressions, but it guarantees the size-extensivity of the MBPT wave function at each order [108]. Also, it is a useful step towards coupled-cluster theory which reorganizes the connected diagrams from MBPT such that certain classes can be summed to infinite order, see section 3.2.

Chapter 3

Coupled-Cluster Theory

Coupled-cluster theory is a powerful method for approximating solutions to the many-body Schrödinger equation. Because of its effectiveness and economical scaling it has been a staple of many-body quantum mechanics for decades. This chapter details various aspects of the coupled-cluster approach and presents ground-state results for multiple systems. First, the CC wave operator and the corresponding effective Hamiltonian will be introduced and used to derive the CC equations. Then, results from MBPT are used to illuminate the underlying many-body physics of the CC wave function. After the mathematical foundations of coupled cluster theory are outlined, specific implementation details are discussed and demonstrated by focusing on two simple examples. Finally, the nuclear many-body problem is formally introduced, along with a brief description of the nuclear interactions used in this work, and selected results are shown.

3.1 Exponential Ansatz

Coupled cluster theory is based on expressing the A -particle correlated wave function $|\Psi\rangle$ using the exponential ansatz [31, 28],

$$|\Psi\rangle = e^{\hat{T}} |\Phi_0\rangle. \quad (3.1)$$

The cluster operator $\hat{T} \equiv \hat{T}_1 + \hat{T}_2 + \cdots + \hat{T}_A$, is composed of k -particle k -hole excitation operators, \hat{T}_k , which have the form,

$$\begin{aligned}\hat{T}_1 &\equiv \sum_{ai} t_i^a \left\{ \hat{a}_a^\dagger \hat{a}_i \right\}, \\ \hat{T}_2 &\equiv \frac{1}{4} \sum_{abij} t_{ij}^{ab} \left\{ \hat{a}_a^\dagger \hat{a}_b^\dagger \hat{a}_j \hat{a}_i \right\}, \\ &\vdots \\ \hat{T}_k &\equiv \left(\frac{1}{k!} \right)^2 \sum_{\substack{a_1 \dots a_k \\ i_1 \dots i_k}} t_{i_1 \dots i_k}^{a_1 \dots a_k} \left\{ \hat{a}_1^\dagger \dots \hat{a}_k^\dagger \hat{a}_{i_k} \dots \hat{a}_{i_1} \right\}.\end{aligned}\tag{3.2}$$

where the unknown matrix elements, $t_{i_1 \dots i_k}^{a_1 \dots a_k}$, are known as *cluster amplitudes* [108].

Using the CC ansatz, the Schrödinger equation can be rewritten by left-multiplying with $\langle \Phi_0 | e^{-\hat{T}}$ as,

$$\begin{aligned}\hat{H} e^{\hat{T}} |\Phi_0\rangle &= E e^{\hat{T}} |\Phi_0\rangle \\ \longrightarrow \quad \langle \Phi_0 | \bar{H} |\Phi_0\rangle &= E,\end{aligned}\tag{3.3}$$

where the *coupled cluster effective Hamiltonian* is defined as,

$$\bar{H} \equiv e^{-\hat{T}} \hat{H} e^{\hat{T}},\tag{3.4}$$

in which the wave operator, $e^{\hat{T}}$, acts as a similarity transform on the Hamiltonian.

An important characteristic of the effective Hamiltonian, \bar{H} , is that because the cluster operator, which contains no de-excitations, is not Hermitian, the exponential wave operator cannot be unitary, and thus \bar{H} is not Hermitian. This initially seems like an explicit con-

tradiction to any standard quantum mechanics formulation where observables are associated with the real eigenvalues of Hermitian matrices. Technically, the existence and reality of the CC solution is only guaranteed when the full cluster operator is used $\hat{T} = \hat{T}_1 + \cdots + \hat{T}_A$, see [138, 87]. However, while the non-Hermiticity does require some special considerations which will be discussed, this fundamental problem does not materialize in this work.

The effective Hamiltonian in Eq. (3.4) can be rewritten with commutators ($[\hat{A}, \hat{B}] = \hat{A}\hat{B} - \hat{B}\hat{A}$) according to the Baker–Campbell–Hausdorff expansion as,

$$\bar{H} = \hat{H} + [\hat{H}, \hat{T}] + \frac{1}{2!}[[\hat{H}, \hat{T}], \hat{T}] + \frac{1}{3!}[[[\hat{H}, \hat{T}], \hat{T}], \hat{T}] + \frac{1}{4!}[[[[\hat{H}, \hat{T}], \hat{T}], \hat{T}], \hat{T}] + \cdots ,$$

which terminates at four-nested commutators when using a two-body interaction. This commutator expression ensures that CC theory is size-extensive and contains only connected terms. In addition, because \hat{T} is an excitation operator, terms of the form $\hat{T}\hat{H}$ are necessarily disconnected and thus vanish [108]. Therefore the CC effective Hamiltonian can be further reduced to

$$\bar{H} = \left(\hat{H} e^{\hat{T}} \right)_c , \quad (3.5)$$

where the subscript “c” indicates that only connected terms are used.

3.1.1 The Coupled Cluster Equations

In practice, the cluster operator \hat{T} must be truncated for calculations to be computationally feasible. In this work, we use only single and double excitations where applicable,

$$\hat{T} = \hat{T}_1 + \hat{T}_2 .$$

This is known as coupled cluster with singles and doubles (CCSD), with an asymptotic computational cost that scales like $\mathcal{O}(n_p^4 n_h^2)$, where n_h is the number of hole states and n_p is the number of particle states. This truncation has been successfully applied to many problems in quantum chemistry [7] and nuclear physics [74, 55].

The unknown cluster amplitudes in CCSD, t_i^a and t_{ij}^{ab} , can be calculated by left-multiplying Eq. (3.1) with $\langle \Phi_i^a | e^{-\hat{T}}$ and with $\langle \Phi_{ij}^{ab} | e^{-\hat{T}}$, respectively,

$$\begin{aligned}\langle \Phi_i^a | \bar{H} | \Phi_0 \rangle &= 0, \\ \langle \Phi_{ij}^{ab} | \bar{H} | \Phi_0 \rangle &= 0.\end{aligned}\tag{3.6}$$

After the Fock matrix has been diagonalized, the diagonal components of Eq. (3.6) can be separated and, after expanding the exponent in Eq. (3.5), the non-vanishing terms of the CCSD amplitude equations become,

$$\begin{aligned}\langle \Phi_i^a | \left[\hat{H}_2 \left(\hat{T}_1 + \hat{T}_2 + \hat{T}_1 \hat{T}_2 + \frac{1}{2!} \hat{T}_1^2 + \frac{1}{3!} \hat{T}_1^3 \right) \right]_c | \Phi_0 \rangle &= \varepsilon_i^a t_i^a \\ \langle \Phi_{ij}^{ab} | \left[\hat{H}_2 \left(1 + \hat{T}_1 + \hat{T}_2 + \frac{1}{2} \hat{T}_1^2 + \hat{T}_1 \hat{T}_2 + \frac{1}{2!} \hat{T}_2^2 + \frac{1}{3!} \hat{T}_1^3 + \frac{1}{2!} \hat{T}_1^2 \hat{T}_2 + \frac{1}{4!} \hat{T}_1^4 \right) \right]_c | \Phi_0 \rangle &= \varepsilon_{ij}^{ab} t_{ij}^{ab},\end{aligned}\tag{3.7}$$

where ε are equivalent to the MBPT energy denominators from Eq. (2.7).

These non-linear equations are solved using an iterative procedure where the cluster amplitudes on the right-hand side of Eq. (3.6) and Eq. (3.7) are updated by calculating the terms on the left-hand side until a fixed point is reached. Like the HF iterative procedure, employing convergence acceleration techniques can reduce the number of CC iterations required. Detailed techniques for solving these equations are discussed in section 3.4.

3.2 Linked-Cluster Theorem and MBPT

The exponential ansatz in Eq. (3.1) and the cluster amplitudes in Eq. (3.1) are not just useful mathematical constructs for solving the many-body problem. They represent physical many-body dynamics and can be derived from the results of many-body perturbation theory, see section 2.7. The *linked-cluster theorem* [67, 47, 18] states that the sum of all time orderings of a disconnected diagram is equal to the product of the two subdiagrams. Using the results and techniques from the factorization theorem, 2.7.1, the linked-cluster theorem can be used to factorize specific MBPT terms such that they can be analytically summed to infinite order. This infinite summation is the main principle behind coupled cluster theory and can be shown to lead naturally to the exponential ansatz.

The first step in deriving the exponential ansatz is to group all the linked MBPT diagrams of Eq. (2.65) into classes according to their number of disconnected pieces. The first class, where all the terms are connected, can be defined as the cluster excitation operator \hat{T} , depicted by the vertex type . The connected terms can then be characterized by their excitation type such that \hat{T}_k corresponds to $kp-kh$ excitations.

The \hat{T}_1 operator represents the class of all connected 1p-1h MBPT diagrams, which are determined by the number of particle-hole pairs (or pair of up- and down-lines) at the top of the diagram,

$$\begin{aligned} \hat{T}_1 |\Phi_0\rangle \equiv & \text{Diagram with one vertex} = (\hat{T}_1^{(1)} + \hat{T}_1^{(2)} + \dots) |\Phi_0\rangle = \text{Diagram with one vertex} + \text{Diagram with one vertex and one loop} \\ & + \text{Diagram with one vertex and two loops} + \text{Diagram with one vertex and three loops} + \dots, \end{aligned} \quad (3.8)$$

where only first-order $\hat{T}^{(1)}$ and second-order $\hat{T}^{(2)}$ terms are shown while resolvant lines and

labels are removed for clarity. The \hat{T}_2 operator similarly represents the class of all connected $2p\text{-}2h$ MBPT diagrams. The first- and second-order terms that belong to this class are,

$$\begin{aligned}\hat{T}_2|\Phi_0\rangle \equiv & \quad \text{Diagram: two vertical lines with arrows pointing down, connected by a horizontal bar at the bottom.} = (\hat{T}_2^{(1)} + \hat{T}_2^{(2)} + \dots) |\Phi_0\rangle = \\ & \quad \text{Diagram: two vertical lines with arrows pointing down, connected by a dashed horizontal bar at the bottom.} + \quad \text{Diagram: two vertical lines with arrows pointing down, one has a dashed horizontal bar at the bottom and the other has a cross symbol.} + \quad \text{Diagram: two vertical lines with arrows pointing down, one has a dashed horizontal bar at the bottom and the other has a cross symbol.} \\ & + \quad \text{Diagram: two vertical lines with arrows pointing down, one has a dashed horizontal bar at the bottom and the other has a cross symbol.} + \quad \text{Diagram: two vertical lines with arrows pointing down, one has a dashed horizontal bar at the bottom and the other has a cross symbol.} + \quad \text{Diagram: two vertical lines with arrows pointing down, one has a dashed horizontal bar at the bottom and the other has a cross symbol.} + \dots\end{aligned}\tag{3.9}$$

Additionally, the \hat{T}_3 operator represents the class of all connected $3p\text{-}3h$ MBPT diagrams, of which there is only two second-order terms,

$$\begin{aligned}\hat{T}_3|\Phi_0\rangle \equiv & \quad \text{Diagram: three vertical lines with arrows pointing down, connected by a horizontal bar at the bottom.} = (\hat{T}_3^{(2)} + \dots) |\Phi_0\rangle = \\ & \quad \text{Diagram: three vertical lines with arrows pointing down, one has a dashed horizontal bar at the bottom and the other has a cross symbol.} + \quad \text{Diagram: three vertical lines with arrows pointing down, one has a dashed horizontal bar at the bottom and the other has a cross symbol.} + \dots\end{aligned}\tag{3.10}$$

So far, this is merely a redefinition of the connected class of MBPT diagrams up to all orders and is not particularly useful. But the disconnected diagrams, neglected up to this point, can be recombined using the factorization theorem 2.7.1 to provide a powerful simplification (see [108] for a more thorough derivation).

As an example, the remaining disconnected first- and second-order diagrams can be written as the product of connected diagrams. First, the second-order disconnected diagram from the term $\hat{f}^2|\Phi_0\rangle$ can be rewritten by adding a duplicate with the left and right subdiagrams exchanged, which doesn't change the value because the state labels are generic. Then, the two disconnected diagrams can be rewritten using the factorization theorem following the

example of Eq. (2.7.1),

$$\begin{array}{c} \text{Diagram: Two vertices } i \text{ and } j \text{ connected by a dashed line, each with a } R_0 \text{ line above it.} \\ \times \end{array} = \frac{1}{2} \left(\begin{array}{c} \text{Diagram: Two vertices } i \text{ and } j \text{ connected by a dashed line, each with a } R_0 \text{ line above it.} \\ \times \end{array} + \begin{array}{c} \text{Diagram: Two vertices } i \text{ and } j \text{ connected by a dashed line, each with a } R_0 \text{ line above it.} \\ \times \end{array} \right) = \frac{1}{2} \left(\begin{array}{c} \text{Diagram: Two vertices } i \text{ connected by a dashed line, each with a } R_0 \text{ line above it.} \\ \times \end{array} \right)^2. \quad (3.11)$$

The resulting product involves the first-order component of the \hat{T}_1 operator in Eq. (3.8), $\hat{T}_1^{(1)}$. Algebraically, this process is written below,

$$\begin{aligned}
 \sum_{abij} \frac{f_i^a f_j^b}{\varepsilon_{ij}^{ab} \varepsilon_i^a} |\Phi_{ij}^{ab}\rangle &= \frac{1}{2} \sum_{abij} \frac{f_i^a f_j^b}{\varepsilon_{ij}^{ab} \varepsilon_i^a} |\Phi_{ij}^{ab}\rangle + \frac{1}{2} \sum_{abij} \frac{f_i^a f_j^b}{\varepsilon_{ij}^{ab} \varepsilon_j^b} |\Phi_{ij}^{ab}\rangle = \frac{1}{2} \sum_{abij} f_i^a f_j^b \frac{\varepsilon_j^b + \varepsilon_i^a}{\varepsilon_{ij}^{ab} \varepsilon_i^a \varepsilon_j^b} |\Phi_{ij}^{ab}\rangle \\
 &= \frac{1}{2} \left(\sum_{ai} \frac{f_i^a}{\varepsilon_i^a} |\Phi_i^a\rangle \right) \left(\sum_{bj} \frac{f_j^b}{\varepsilon_j^b} |\Phi_j^b\rangle \right) = \frac{1}{2} (\hat{T}_1^{(1)})^2. \quad (3.12)
 \end{aligned}$$

This procedure can be repeated for the single second-order disconnected term from $\hat{V}^2 |\Phi_0\rangle$,

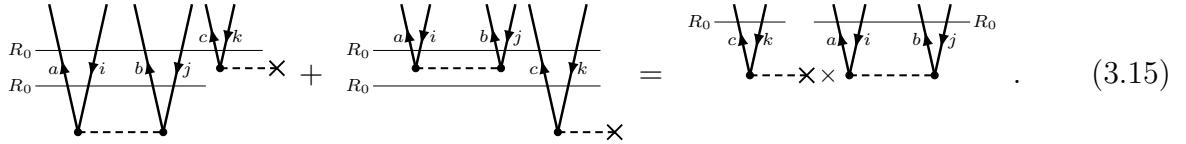
$$\begin{array}{c} \text{Diagram: Four vertices } a, b, c, d \text{ connected by a dashed line, each with a } R_0 \text{ line above it.} \\ \times \end{array} = \frac{1}{2} \left(\begin{array}{c} \text{Diagram: Four vertices } a, b, c, d \text{ connected by a dashed line, each with a } R_0 \text{ line above it.} \\ \times \end{array} + \begin{array}{c} \text{Diagram: Four vertices } a, b, c, d \text{ connected by a dashed line, each with a } R_0 \text{ line above it.} \\ \times \end{array} \right) = \frac{1}{2} \left(\begin{array}{c} \text{Diagram: Two vertices } i \text{ and } j \text{ connected by a dashed line, each with a } R_0 \text{ line above it.} \\ \times \end{array} \right)^2. \quad (3.13)$$

A similar results gives the product involving the first-order component of the \hat{T}_2 operator in

Eq. (3.9), $\hat{T}_2^{(1)}$. Again, the factorization process is written algebraically,

$$\begin{aligned}
\frac{1}{16} \sum_{\substack{abcd \\ i j k l}} \frac{V_{ij}^{ab} V_{kl}^{cd}}{\varepsilon_{ijkl}^a \varepsilon_{ij}^b} |\Phi_{ijkl}^{abcd}\rangle &= \frac{1}{32} \sum_{\substack{abcd \\ i j k l}} \frac{V_{ij}^{ab} V_{kl}^{cd}}{\varepsilon_{ijkl}^a \varepsilon_{ij}^b \varepsilon_{ij}^c} |\Phi_{ijkl}^{abcd}\rangle + \frac{1}{32} \sum_{\substack{abcd \\ i j k l}} \frac{V_{ij}^{ab} V_{kl}^{cd}}{\varepsilon_{ijkl}^a \varepsilon_{ij}^b \varepsilon_{kl}^d} |\Phi_{ijkl}^{abcd}\rangle \\
&= \frac{1}{32} \sum_{\substack{abcd \\ i j k l}} V_{ij}^{ab} V_{kl}^{cd} \frac{\varepsilon_{kl}^{cd} + \varepsilon_{ij}^{ab}}{\varepsilon_{ijkl}^a \varepsilon_{ij}^b \varepsilon_{kl}^d} |\Phi_{ijkl}^{abcd}\rangle \\
&= \frac{1}{2} \left(\frac{1}{4} \sum_{abij} \frac{V_{ij}^{ab}}{\varepsilon_{ij}^{ab}} |\Phi_{ij}^{ab}\rangle \right) \left(\frac{1}{4} \sum_{cdkl} \frac{V_{kl}^{cd}}{\varepsilon_{kl}^{cd}} |\Phi_{kl}^{cd}\rangle \right) = \frac{1}{2} (\hat{T}_2^{(1)})^2. \quad (3.14)
\end{aligned}$$

Lastly, for the disconnected terms from $\hat{V}\hat{f}|\Phi_0\rangle$ and $\hat{f}\hat{V}|\Phi_0\rangle$, the first duplication step can be skipped and the diagrams can be factorized following the procedure in Eq. (2.7.1),



This factorization results in a mixed term between the the first-order components from the \hat{T}_1 and \hat{T}_2 operators,

$$\begin{aligned}
\frac{1}{4} \sum_{\substack{abc \\ ijk}} \frac{V_{ij}^{ab} f_k^c}{\varepsilon_{ijk}^a \varepsilon_{ij}^b} |\Phi_{ijk}^{abc}\rangle + \frac{1}{4} \sum_{\substack{abc \\ ijk}} \frac{V_{ij}^{ab} f_k^c}{\varepsilon_{ijk}^a \varepsilon_{ij}^b \varepsilon_k^c} |\Phi_{ijk}^{abc}\rangle &= \frac{1}{4} \sum_{\substack{abcd \\ i j k l}} V_{ij}^{ab} f_k^c \frac{\varepsilon_k^c + \varepsilon_{ij}^{ab}}{\varepsilon_{ijk}^a \varepsilon_{ij}^b \varepsilon_k^c} |\Phi_{ijk}^{abc}\rangle \\
&= \left(\sum_{ck} \frac{f_k^c}{\varepsilon_k^c} |\Phi_k^c\rangle \right) \left(\frac{1}{4} \sum_{abij} \frac{V_{ij}^{ab}}{\varepsilon_{ij}^{ab}} |\Phi_{ij}^{ab}\rangle \right) = \hat{T}_1^{(1)} \hat{T}_2^{(1)} \quad (3.16)
\end{aligned}$$

Adding these factorized contributions from Eqs. (3.12), (3.14), and (3.16) gives $\frac{1}{2} (\hat{T}_1^{(1)})^2 + \hat{T}_1^{(1)} \hat{T}_2^{(1)} + \frac{1}{2} (\hat{T}_2^{(1)})^2 = \frac{1}{2} (\hat{T}_1^{(1)} + \hat{T}_2^{(1)})^2$. Repeating this procedure for all diagrams with two disconnected parts ($L = 2$) adds similar terms of all orders. The final contribution from

the $L = 2$ class of diagrams with two disconnected parts is,

$$\sum_{n=0}^{\infty} \left[\hat{R}_0 (\hat{V} - \Delta E_0) \right]^n |\Phi_0\rangle_{L=2} = \frac{1}{2} \hat{T}^2. \quad (3.17)$$

A similar form results from any class of diagrams with k disconnected pieces,

$$\sum_{n=0}^{\infty} \left[\hat{R}_0 (\hat{V} - \Delta E_0) \right]^n |\Phi_0\rangle_{L=k} = \frac{1}{k!} \hat{T}^k. \quad (3.18)$$

Therefore, summing over all classes of diagrams gives the final result that justifies the exponential ansatz,

$$|\Psi\rangle = \sum_{k=0}^{\infty} \sum_{n=0}^{\infty} \left[\hat{R}_0 (\hat{V} - \Delta E_0) \right]^n |\Phi_0\rangle_{L=k} = \sum_{k=0}^{\infty} \frac{1}{k!} \hat{T}^k |\Phi_0\rangle = e^{\hat{T}} |\Phi_0\rangle. \quad (3.19)$$

This equation shows the true strength and elegance of coupled cluster theory. By an ingenious reorganization and factorization of certain MBPT diagrams, the exponential ansatz captures the contribution of these excitations to infinite order. A more comprehensive derivation of the linked-cluster theorem can be found in [108].

3.3 Example: Pairing Model

It's beneficial to illustrate a simplified example of coupled cluster theory. For this purpose, we turn our attention to the simple pairing model. This system uses a model space of N shells, or degenerate groups of single-particle states, each with two opposite spin orbitals.

With a closed-shell reference state, the Hamiltonian is restricted to interact only between

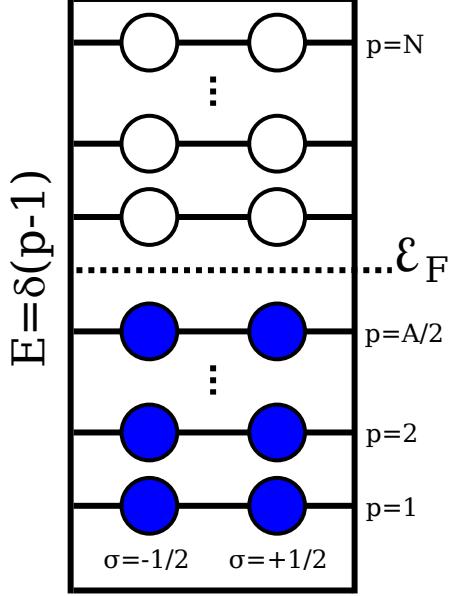


Figure 3.1: Schematic representation of the pairing model space. The shells are equally spaced and doubly degenerate with one spin-up and one spin-down state.

unbroken pairs, which can be written as,

$$\begin{aligned}
 {}^{(1)}\hat{H} &= \delta \sum_p^N (p-1) \left[\hat{a}_{p+}^\dagger \hat{a}_{p+} + \hat{a}_{p-}^\dagger \hat{a}_{p-} \right], \quad \text{and} \\
 {}^{(2)}\hat{H} &= -\frac{g}{2} \sum_{pq}^N \hat{a}_{p+}^\dagger \hat{a}_{p-}^\dagger \hat{a}_{q-} \hat{a}_{q+},
 \end{aligned} \tag{3.20}$$

where δ and g are free parameters and the \pm labels represent the spin-up and spin-down states, respectively.

As with all other coupled cluster calculations in this work, the first step is transforming the problem to the Hartree-Fock basis. In this case, the restriction to unbroken pairs means that the original single-particle states do not mix with states in other shells. In fact, the original basis is already an eigenbasis of the Fock operator, Eq. (2.5), so that all is left of the HF transformation is to redefine the single-particle energies to their corresponding

Hartree-Fock energies while the two-body interaction is left unchanged,

$$\begin{aligned}\varepsilon_{pmp} &= {}^{(1)}H_{pmp}^{pmp} + \sum_{im_i} {}^{(2)}H_{pmp}^{pmpim_i} = \delta(p-1) - \frac{g}{2} \\ V_{rs}^{pq} &= {}^{(2)}H_{rs}^{pq}\end{aligned}\quad (3.21)$$

Again, because of the pairing restriction, only hole-state energies have to be redefined.

The next step in calculating the ground-state correlation energy is to solve the CCD equations in the Hartree-Fock basis. The system of equations comes from the terms of Eq. (3.7) that contain only the \hat{T}_2 operator, and are most easily derived with diagrammatic techniques, see [108].

$$\begin{aligned}\langle \Phi_{ij}^{ab} | (\hat{H} e^{\hat{T}_2})_C | \Phi_0 \rangle - & \left[\text{Diagram with dashed cross at } i \right] - \left[\text{Diagram with dashed cross at } j \right] = \left[\text{Diagram with dashed cross at } i \right] + \left[\text{Diagram with dashed cross at } j \right] \\ + & \left[\text{Diagram with dashed cross at } k \right] + \left[\text{Diagram with dashed cross at } l \right] + \left[\text{Diagram with dashed cross at } c \right] + \left[\text{Diagram with dashed cross at } d \right] + \left[\text{Diagram with dashed cross at } k \right] + \left[\text{Diagram with dashed cross at } l \right] \\ + & \left[\text{Diagram with dashed cross at } d \right] + \left[\text{Diagram with dashed cross at } c \right] + \left[\text{Diagram with dashed cross at } k \right] + \left[\text{Diagram with dashed cross at } l \right] \\ (\varepsilon_i + \varepsilon_j - \varepsilon_a - \varepsilon_b) t_{ij}^{ab} &= V_{ij}^{ab} + \frac{1}{2} \sum_{kl} V_{ij}^{kl} t_{kl}^{ab} + \frac{1}{2} \sum_{cd} V_{cd}^{ab} t_{ij}^{cd} - \hat{P}(ij|ab) \sum_{kc} V_{ic}^{kb} t_{kj}^{ac} \\ + \frac{1}{4} \sum_{klcd} & V_{cd}^{kl} t_{kl}^{ab} t_{ij}^{cd} + \hat{P}(ab) \sum_{klcd} V_{cd}^{kl} t_{lj}^{ac} t_{ki}^{bd} - \hat{P}(ij) \frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_{lj}^{ab} t_{ki}^{cd} - \hat{P}(ab) \frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_{ij}^{db} t_{kl}^{ca}\end{aligned}\quad (3.22)$$

The CCD equations are written in this particular form so that an initial guess for all the

amplitudes t_{ij}^{ab} can be used to calculate the right-hand side of Eq. (3.3) and update the amplitudes on the left-hand side iteratively until the amplitudes do not change within a certain tolerance.

Lastly, the CCD correlation energy can be found with the \hat{T}_2 term of Eq. (3.1),

$$\Delta E_{\text{CCD}} = \langle \Phi_0 | (\hat{H} e^{\hat{T}_2})_{\text{C}} | \Phi_0 \rangle = \text{Diagram} = \frac{1}{4} \sum_{klcd} V_{cd}^{kl} t_{kl}^{cd} \quad (3.23)$$

The correlation energies for a specific case, with $\delta = 1.0$, $N = 4$, and $A = 4$, were calculated for a number of different interaction strengths, g . The results are shown in Fig. 3.2 along with the MBPT correlation energies to third (MBPT3) and fourth (MBPT4) orders for comparison. The MBPT expressions for second and third order are,

$$\Delta E_{\text{MBPT2}} = \frac{1}{4} \sum_{ijab} \frac{V_{ab}^{ij} V_{ij}^{ab}}{\varepsilon_{ij}^{ab}}, \quad (3.24)$$

$$\Delta E_{\text{MBPT3}} = \Delta E_{\text{MBPT2}} + \frac{1}{8} \sum_{ijabcd} \frac{V_{ab}^{ij} V_{cd}^{ab} V_{ij}^{cd}}{\varepsilon_{ij}^{ab} \varepsilon_{ij}^{cd}} + \frac{1}{8} \sum_{ijklab} \frac{V_{ab}^{ij} V_{ij}^{kl} V_{kl}^{ab}}{\varepsilon_{ij}^{ab} \varepsilon_{kl}^{ab}}. \quad (3.25)$$

Generally, the fourth-order expression contains 39 additional terms. However, most of these vanish in this case because of the form of the pairing interaction.

Also shown are the exact results from the CI method. With an example this small, it's possible to diagonalize, and even show explicitly, the full CI Hamiltonian matrix for an exact result. There are six possible Slater determinants with no broken pairs, one representing the reference state, four representing various $2p$ - $2h$ excitations, one representing a $4p$ - $4h$ excitation. The diagonal elements of the matrix include the single-particle energies of the constituent states, and the matrix elements between Slater determinants with no overlap

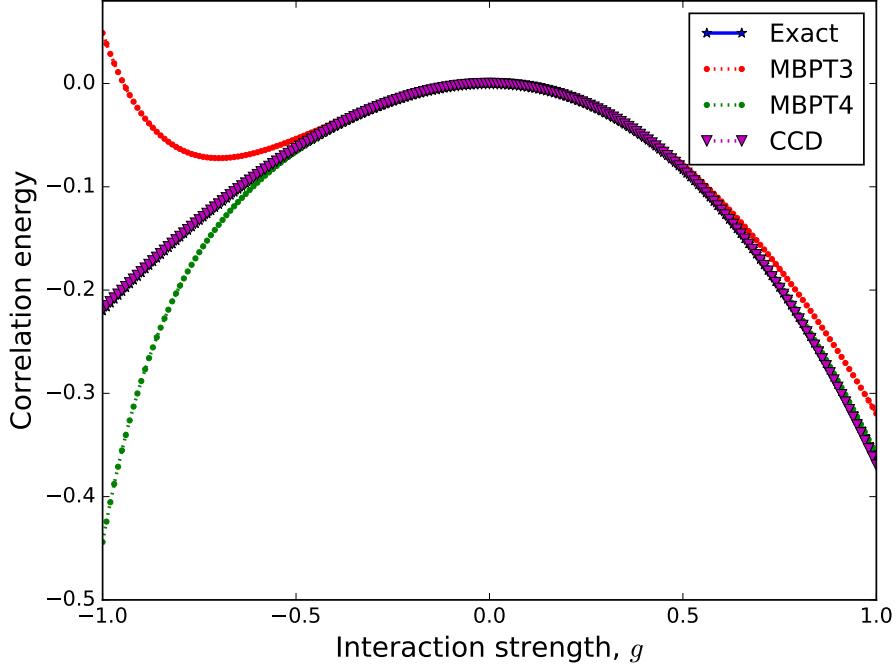


Figure 3.2: Correlation energy for the pairing model with exact diagonalization, CCD, and perturbation theory to third (MBPT3) and fourth order (MBPT4) for a range of interaction values, g .

vanish in accordance with the Slater-Condon rules, see Eq. (2.46) and [111, 33]. The full Hamiltonian matrix to be diagonalized is,

$$H = \begin{bmatrix} 2\delta - g & -g/2 & -g/2 & -g/2 & -g/2 & 0 \\ -g/2 & 4\delta - g & -g/2 & -g/2 & -0 & -g/2 \\ -g/2 & -g/2 & 6\delta - g & 0 & -g/2 & -g/2 \\ -g/2 & -g/2 & 0 & 6\delta - g & -g/2 & -g/2 \\ -g/2 & 0 & -g/2 & -g/2 & 8\delta - g & -g/2 \\ 0 & -g/2 & -g/2 & -g/2 & -g/2 & 10\delta - g \end{bmatrix}. \quad (3.26)$$

As methods to obtain the ground-state correlation energy, both CI and CC decouple, to some degree, the reference state and excitations from it. This decoupling has the effect of

shuffling correlations into the reference state and suppressing matrix elements connected to it. Full decoupling between the reference state and all other Slater determinants can only be achieved with untruncated versions of these techniques, while decoupling of the strongest correlations can be approximately obtained with appropriate truncations. However, unlike other many-body methods, the most unique aspect of the CC similarity transformation is that because of its non-unitarity, the resulting Hamiltonian will not be Hermitian. This decoupling and non-Hermiticity can be seen in Fig. 3.3, which shows the effect of the CC similarity transformation on the Hamiltonian for a pairing case with $N = 6$ and $A = 4$. The effective Hamiltonian \bar{H} shown in Fig. 3.3 can be explicitly built, and it happens to be

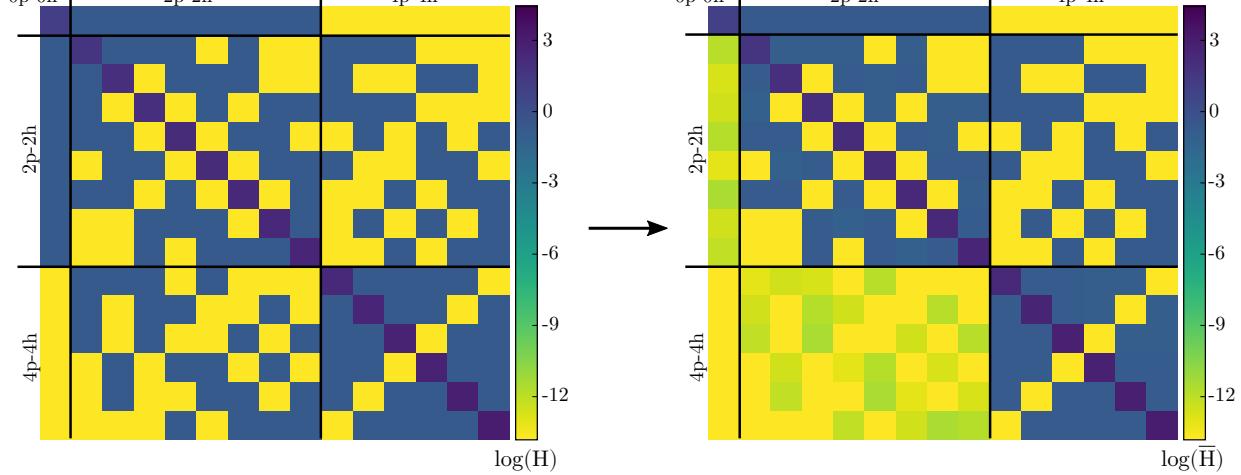


Figure 3.3: Visualization of the CCD similarity transform on the pairing Hamiltonian for four particles and six shells. This shows the main function of CCD, which is to decouple $2p$ - $2h$ excitations from the ground state, shown by the suppression of matrix elements on the first column. In the pairing model, this also has the effect of decoupling $2p$ - $2h$ excitations from $4p$ - $4h$ excitations. Also, the non-unitary nature of the transformation is obvious given the asymmetry of the resulting Hamiltonian.

beneficial to do so as part of most CC calculations, both for solving the CC equations and for use in post-CC methods. This topic is discussed in detail in the next section.

3.4 Solving the Coupled Cluster Equations

As described above, the coupled cluster equations are solved by first initializing all of the cluster amplitudes, then updating them by computing various sums over particle and hole combinations, $\text{CC}(\hat{T})$. This updating procedure is performed over multiple iterations until the amplitudes stay unchanged within a certain tolerance.

$$\begin{aligned} \text{Initialize : } & \quad \hat{T}^{(0)} = \hat{T}_{\text{init}} \\ \text{Iterate : } & \quad \hat{T}^{(n+1)} \leftarrow \text{CC}(\hat{T}^{(n)}) \end{aligned} \quad (3.27)$$

Generally, the convergence of this algorithm depends on the relative magnitudes between the inter-particle force and the single-particle energy spacing. For a relatively small Fermi gap between the closed shell of occupied states and the unoccupied particle states, the energy denominators will approach zero and cause a divergent or chaotic solution [119]. Physically, this situation occurs when a system exhibits strong many-particle clustering, which is difficult to capture with only the single and double excitations of CCSD. A simple way to avoid this ill-defined behavior is to scale the energy denominators for early iterations or to employ linear mixing to dampen the solution.

$$\hat{T}^{(n+1)} \leftarrow \alpha \text{CC}(\hat{T}^{(n)}) + (1 - \alpha) \hat{T}^{(n)} \quad (3.28)$$

If a solution to a highly-collective system does not diverge, it typically converges very slowly. To improve the convergence rate, techniques already utilized for the Hartree-Fock iterations can also be employed here, such as DIIS [100, 99] or Broyden's method [21]. The additional computational complexity for the CC iterations is simply a multiplicative factor equal to the

number of iterations performed. Typical calculations in this work with DIIS acceleration are converged within ~ 30 iterations. Therefore, any significant improvements to the CC algorithm will involve the expensive sums embedded within the function $\text{CC}(\hat{T}^{(n)})$.

3.4.1 Symmetry Channels

For the coupled cluster equations, as well as many other many-body methods, the first way to simplify the various sums is to exploit any symmetries of the underlying Hamiltonian of a particular problem. These symmetries manifest as conserved quantities, and because of the underlying nature of the cluster operators, see Section 3.2, these must conserve these quantum numbers as well. For example, the pairing Hamiltonian of Section 3.3 has a symmetry that conserves both the total spin projection and the number of pairs. The Coulomb Hamiltonian of Section 3.5, which has translational symmetry, conserves the linear momentum of any state. Finally, the spherical symmetry of the nuclear Hamiltonian ensures that angular momentum and parity are conserved. To utilize these symmetries, any sums that contain many-body states with different conserved quantum numbers can be ignored. For efficiency, these symmetry groups can be pre-sorted into *channels*, $\Sigma_{\vec{\xi}}$, where $\vec{\xi}$ represents the relevant quantum numbers of a certain channel.

For CCSD calculations, useful types of channels include the direct two-body channels, $\Sigma_{\vec{\xi}_1}$ —which categorizes the vector sum of two single-particle-state quantum numbers—and the cross two-body channels, $\Sigma_{\vec{\xi}_2}$ —which categorizes the vector difference of two single-particle-state quantum numbers or, equivalently, the vector sum of the quantum numbers of a

single-particle state and a time-reversed single-particle state.

$$\vec{\xi}_{pq} = \vec{\xi}_p + \vec{\xi}_q \quad \longrightarrow \quad |pq\rangle \in \Sigma_{\vec{\xi}_1=\vec{\xi}_{pq}} \quad (3.29)$$

$$\vec{\xi}_{p\bar{q}} = \vec{\xi}_p - \vec{\xi}_q = \vec{\xi}_p + \vec{\xi}_{\bar{q}} \quad \longrightarrow \quad |p\bar{q}\rangle \in \Sigma_{\vec{\xi}_2=\vec{\xi}_{p\bar{q}}} \quad (3.30)$$

Also useful is the one-body channels, $\Sigma_{\vec{\xi}_3}$, which categorizes both single-particle states by their conserved quantum numbers. These one-body channels can also characterize a special type of three-body state: the vector difference between the quantum numbers of a direct two-body state and a single-particle state or, equivalently, the vector sum of the quantum numbers of a two-body direct state and a time-reversed single-particle state,

$$\vec{\xi}_p \quad \longrightarrow \quad |p\rangle \in \Sigma_{\vec{\xi}_3=\vec{\xi}_p} \quad (3.31)$$

$$\vec{\xi}_{pq\bar{r}} = \vec{\xi}_p + \vec{\xi}_q - \vec{\xi}_r = \vec{\xi}_p + \vec{\xi}_q + \vec{\xi}_{\bar{r}} \quad \longrightarrow \quad |pq\bar{r}\rangle \in \Sigma_{\vec{\xi}_3=\vec{\xi}_{pq\bar{r}}}. \quad (3.32)$$

Using these channel structures, the interaction matrix elements and cluster amplitudes can be built in different ways. The full applicability of these structures are shown in detail in the appendix B, but a few examples using sums in the CCD equations (3.3) are shown here. The direct two-body channels can be used when two summed indices appear in the bra- or ket-state of multiple matrix-elements,

$$\frac{1}{2} \sum_{cd} V_{cd}^{ab} t_{ij}^{cd} = \frac{1}{2} \sum_{|cd\rangle} V_{cd}^{ab} t_{ij}^{cd} \quad \text{for } |cd\rangle \in \Sigma_{\vec{\xi}_{ab}} = \Sigma_{\vec{\xi}_{ij}}. \quad (3.33)$$

The cross two-body channels are used when two summed indices appear in the opposite

corresponding bra- and ket-states of multiple matrix-matrix elements,

$$\sum_{kc} V_{ic}^{kb} t_{kj}^{ac} = \sum_{|k\bar{c}\rangle} V_{i\bar{b}}^{k\bar{c}} t_{k\bar{c}}^{a\bar{j}} \text{ for } |k\bar{c}\rangle \in \Sigma_{\vec{\xi}_{i\bar{b}}} = \Sigma_{\vec{\xi}_{a\bar{j}}}. \quad (3.34)$$

Lastly, the one- and three-body channels are used when a single summed index appears opposite a direct two-body state in multiple matrix elements,

$$\frac{1}{2} \sum_{kled} V_{cd}^{kl} t_{ij}^{db} t_{kl}^{ca} = \frac{1}{2} \sum_{\substack{|k\bar{l}\bar{c}\rangle \\ |d\rangle}} V_d^{k\bar{l}\bar{c}} t_{ij\bar{b}}^d t_{k\bar{l}\bar{c}}^a \text{ for } |k\bar{l}\bar{c}\rangle, |d\rangle \in \Sigma_{\vec{\xi}_{ij\bar{b}}} = \Sigma_{\vec{\xi}_a}. \quad (3.35)$$

3.4.2 Matrix Structures and Intermediates

Symmetry channels not only provide an organized structure for the interaction matrix elements and cluster amplitudes, and remove any terms that violate the underlying symmetry of a problem, but they also naturally provide an efficient way of performing sums using matrix-matrix multiplications. For example, the sums in Eq. (??) can be reformulated as matrix-matrix multiplications by structuring the channel-separated interaction matrix elements and cluster amplitudes into individual matrices. These operations can be performed very quickly using highly optimized linear algebra algorithms like those found in BLAS (Basic Linear Algebra Subprograms) [2]. The matrices can be reordered so that the summed indices correspond to the internal columns and rows of those matrices.

For the direct two-body case of Eq. (3.33), the structures are already in the correct order such that the state $|cd\rangle$, indexed by the columns of \mathbf{V} and the rows of \mathbf{t} , is summed by multiplying the two matrices,

$$\frac{1}{2} \sum_{cd} V_{cd}^{ab} t_{ij}^{cd} = \frac{1}{2} \mathbf{V}_{cd}^{ab} \cdot \mathbf{t}_{ij}^{cd} \text{ for } |ab\rangle, |ij\rangle, |cd\rangle \in \Sigma_{\vec{\xi}_1}. \quad (3.36)$$

For the cross two-body case of Eq. (3.34), the states $|j\rangle$, $|b\rangle$, and $|c\rangle$ are time reversed so that the summed variables are collected in a state $|k\bar{c}\rangle$. Then the matrix structures are reordered so this state is indexed by columns and rows of \mathbf{t} and \mathbf{V} , respectively,

$$\sum_{kc} V_{ic}^{kb} t_{kj}^{ac} = \mathbf{t}_{k\bar{c}}^{a\bar{j}} \cdot \mathbf{V}_{ib}^{k\bar{c}} \text{ for } |a\bar{j}\rangle, |i\bar{b}\rangle, |k\bar{c}\rangle \in \Sigma_{\vec{\xi}_2}. \quad (3.37)$$

Lastly, for the case of Eq. (3.35), the states $|b\rangle$ and $|c\rangle$ are time-reversed so that the states $|d\rangle$ and $|kl\bar{c}\rangle$ appear in two different matrix elements. Then, the matrix structures are reorganized so the summed states occur in the appropriate rows and columns for matrix-matrix multiplication,

$$\frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_{ij}^{db} t_{kl}^{ca} = \frac{1}{2} \mathbf{t}_{kl\bar{c}}^a \cdot \mathbf{V}_d^{kl\bar{c}} \cdot \mathbf{t}_{ij\bar{b}}^d \text{ for } |a\rangle, |ij\bar{b}\rangle, |kl\bar{c}\rangle, |d\rangle \in \Sigma_{\vec{\xi}_3}. \quad (3.38)$$

These sums correspond to different components of the updated cluster amplitudes according to Eq. (3.27). So the different channel structures of the matrix-matrix multiplications correspond to different amplitude structures according to the sum's external indices. The two external direct two-body states of Eq. (3.36), $|ab\rangle$ and $|ij\rangle$, naturally map to the direct amplitude structure,

$$\mathbf{t}_{ij}^{ab} \leftarrow \frac{1}{2} \mathbf{V}_{cd}^{ab} \cdot \mathbf{t}_{ij}^{cd} \text{ for } |ab\rangle, |ij\rangle, |cd\rangle \in \Sigma_{\vec{\xi}_1}. \quad (3.39)$$

Similarly, the two external cross two-body states of Eq. (3.37), $|a\bar{j}\rangle$ and $|i\bar{b}\rangle$, naturally map to the cross amplitude structure,

$$\mathbf{t}_{ib}^{a\bar{j}} \leftarrow \mathbf{t}_{k\bar{c}}^{a\bar{j}} \cdot \mathbf{V}_{ib}^{k\bar{c}} \text{ for } |a\bar{j}\rangle, |i\bar{b}\rangle, |k\bar{c}\rangle \in \Sigma_{\vec{\xi}_2}. \quad (3.40)$$

Lastly, the one- and three-body external states of Eq. (3.38), $|a\rangle$ and $|i\bar{b}\rangle$, naturally map to the one-body amplitude structure characterized by the index a ,

$$\mathbf{t}_{ij\bar{b}}^a \leftarrow \frac{1}{2} \mathbf{t}_{kl\bar{c}}^a \cdot \mathbf{V}_d^{kl\bar{c}} \cdot \mathbf{t}_{ij\bar{b}}^d \quad \text{for } |a\rangle, |ij\bar{b}\rangle, |kl\bar{c}\rangle, |d\rangle \in \Sigma_{\vec{\xi}_3}. \quad (3.41)$$

The last summation in the matrix-matrix form of Eqs. (??) and (??) involves two multiplications, which suggests the need for an *intermediate* matrix to hold the result of the first operation. This is the last main ingredient to an efficient CC algorithm. To see the benefit of intermediate structures, it's helpful to examine an expensive sum from the CCD equations. For typical calculations, particle states outnumber hole states by an order of magnitude, $n_p \sim 10n_h$, which means that one of the most expensive sums is,

$$\frac{1}{4} \sum_{klcd} V_{cd}^{kl} t_{kl}^{ab} t_{ij}^{cd}. \quad (3.42)$$

Because this term must be computed for each t_{ij}^{ab} , its computational cost naively scales as $\mathcal{O}(N_h^4 N_p^4)$. However, using the matrix form of this sum and an intermediate matrix,

$$\frac{1}{4} \sum_{klcd} V_{cd}^{kl} t_{kl}^{ab} t_{ij}^{cd} = \frac{1}{4} \mathbf{t}_{kl}^{ab} \cdot (\mathbf{V}_{cd}^{kl} \cdot \mathbf{t}_{ij}^{cd}) = \frac{1}{4} \mathbf{t}_{kl}^{ab} \cdot \mathbf{X}_{ij}^{kl} \rightarrow \mathbf{t}_{ij}^{ab}. \quad (3.43)$$

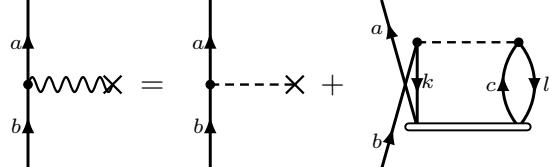
this term is now computed as the combination of two sums, each scaling as $\mathcal{O}(N_h^4 N_p^2)$. These intermediates can also be used as a way to combine similar sums. For example, the last step of Eq. 3.43 has a very similar structure to the first sum in Eq. 3.3. Therefore, the

two sums can be written with a common intermediate as,

$$\frac{1}{2} \sum_{kl} V_{ij}^{kl} t_{kl}^{ab} + \frac{1}{4} \sum_{klcd} V_{cd}^{kl} t_{kl}^{ab} t_{ij}^{cd} = \frac{1}{2} \mathbf{t}_{kl}^{ab} \cdot \left[\mathbf{V}_{ij}^{kl} + \frac{1}{2} \mathbf{V}_{cd}^{kl} \cdot \mathbf{t}_{ij}^{cd} \right] = \frac{1}{4} \mathbf{t}_{kl}^{ab} \cdot \mathbf{X}_{ij}^{kl} \rightarrow \mathbf{t}_{ij}^{ab},$$

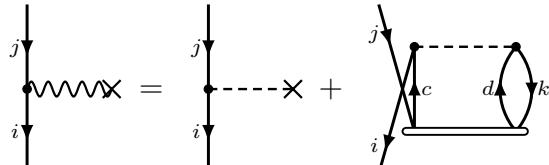
where, $\mathbf{X}_{ij}^{kl} = \mathbf{V}_{ij}^{kl} + \frac{1}{2} \mathbf{V}_{cd}^{kl} \cdot \mathbf{t}_{ij}^{cd}$ (3.44)

It just so happens that this form of the intermediate X_{ij}^{kl} is equivalent to the hhhh component of the CCD similarity transformed Hamiltonian, \bar{H} . Constructing other intermediates in this way gives similar results, so it's a natural extension to actually construct the effective Hamiltonian at each iteration for the express purpose of using it as different intermediate components for the CC equations. This has the added benefit of having already computed the effective Hamiltonian for post-CC methods. The different components of the CCD effective Hamiltonian, $\bar{H}_{\text{CCD}} = (\hat{H} e^{\hat{T}_2})_c$, are written below in both algebraic and diagrammatic form. The pp, one-body component of \bar{H}_{CCD} is,



$$X_b^a = f_b^a - \frac{1}{2} \sum_{klc} V_{bc}^{kl} t_{kl}^{ac}. \quad (3.45)$$

The hh, one-body component is,



$$X_j^i = f_j^i + \frac{1}{2} \sum_{kcd} V_{cd}^{ik} t_{jk}^{cd}. \quad (3.46)$$

The hhhh, two-body component, which appears as the intermediate in Eqs. (3.43) and (3.4.2), is,

$$X_{kl}^{ij} = V_{kl}^{ij} + \frac{1}{2} \sum_{cd} V_{cd}^{ij} t_{kl}^{cd}. \quad (3.47)$$

Lastly, the hphp, two-body component is,

$$X_{jb}^{ia} = V_{jb}^{ia} - \frac{1}{2} \sum_{kc} V_{cb}^{ik} t_{jk}^{ca}. \quad (3.48)$$

Using these terms, the CCD equations can be written in pseudo-linear form using the $2p$ - $2h$ component of effective Hamiltonian form of the equations, Eq. (3.6). This also explicitly shows the decoupling of the effective Hamiltonian with $2p$ - $2h$ excitations. The pphh,

two-body component, which should vanish when the CCS amplitudes have converged, is,

$$X_{ij}^{ab} = 0 = V_{ij}^{ab} + \hat{P}(ab) \sum_c X_c^a t_{ij}^{cb} - \hat{P}(ij) \sum_k X_i^k t_{kj}^{ab} \\ + \frac{1}{2} \sum_{cd} X_{cd}^{ab} t_{ij}^{cd} + \frac{1}{2} \sum_{kl} X_{ij}^{kl} t_{kl}^{ab} - \hat{P}(ab|ij) \sum_{kc} X_{ic}^{kb} t_{kj}^{ac}. \quad (3.49)$$

The components and intermediates of the CCSD effective Hamiltonian are much more complicated and are shown with their corresponding sums in the appendix ??.

3.5 Example: Homogeneous Electron Gas

Another relatively simple calculation using the CCD approximation is the homogeneous electron gas. This example aims to calculate the ground state energy of a three-dimensional gas of electrons subject to Coulomb repulsion. This is an approximate model of the valence electrons in a metal, subject to a uniform background of positive charge from the nuclei and core electrons [50]. As will be explained below, this calculation employs pure-momentum eigenstates such that $1p$ - $1h$ excitations from the reference state are forbidden by momentum conservation. This means that the problem reduces to the doubles approximation. To obtain realistic results, a sufficiently-sized basis with a sufficient number of electrons must be used. Therefore, the improvements discussed in Section 3.4 are necessary to keep the computation

time manageable as the system size increases.

With a uniform background potential, the electron gas can be constructed using eigenfunctions of the kinetic energy operator, $(^1)\hat{H} = \hat{T} = \frac{-\hbar^2}{2m}\nabla^2$. In an infinite volume, however, there are an unlistable number of plane wave modes which satisfy this condition due to the continuous nature of the linear momentum eigenstates. Therefore, the single-particle orbits will be constructed in a finite box of volume Ω and length L , and then the limit $L \rightarrow \infty$ can be taken after various expectation values have been computed.

$$\begin{aligned} \frac{-\hbar^2}{2m}\nabla^2\phi_{\mathbf{k}\sigma}(\mathbf{r}) &= \epsilon_{\mathbf{k}}\phi_{\mathbf{k}\sigma}(\mathbf{r}) \\ \phi_{\mathbf{k}\sigma}(\mathbf{r}) &= \frac{1}{\sqrt{\Omega}}\exp(i\mathbf{kr})\xi_{\sigma} \end{aligned} \quad (3.50)$$

where m is the electron mass, \mathbf{k} is the wave number, and ξ_{σ} is the spin function for either spin up or down electrons

$$\xi_{\sigma=+1/2} = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad \xi_{\sigma=-1/2} = \begin{pmatrix} 0 \\ 1 \end{pmatrix}. \quad (3.51)$$

Assuming the single-particle orbits follow periodic boundary conditions within the containing box ($\phi(\mathbf{r}_i) = \phi(\mathbf{r}_i + L)$ for $i = x, y, z$) the wave numbers are quantized,

$$k_i = \frac{2\pi n_i}{L} \quad i = x, y, z \quad n_i = 0, \pm 1, \pm 2, \dots \quad (3.52)$$

A state can therefore be characterized by the quantum numbers n_x , n_y , and n_z as well as the spin quantum number σ . The energy of such a state, independent of the spin, can be

written as

$$\epsilon_{n_x, n_y, n_z} = \frac{\hbar^2}{2m} (k_x^2 + k_y^2 + k_z^2) = \frac{\hbar^2}{2m} \left(\frac{2\pi}{L}\right)^2 (n_x^2 + n_y^2 + n_z^2). \quad (3.53)$$

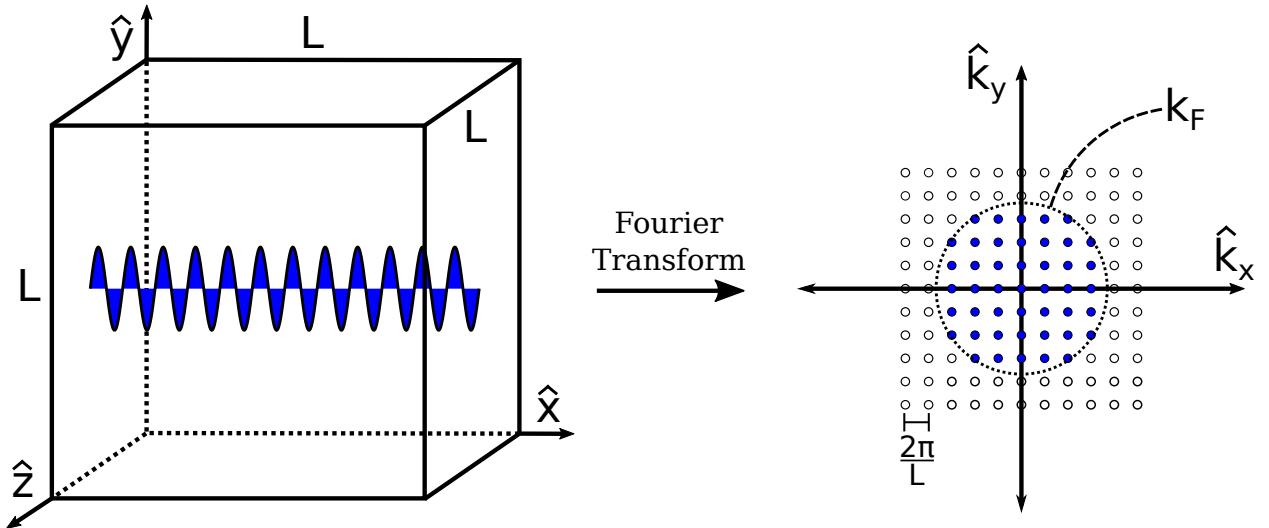


Figure 3.4: Visualization of the Fourier transform of a finite box. This transformation characterizes the construction of the single-particle basis for infinite matter, mapping plane waves in coordinate space onto finitely-spaced points in momentum space.

Now that the single-particle orbits are established, a particular basis consisting of these orbits can be chosen such that all states are included up to a closed shell. This basis is then filled with electrons until a closed Fermi level is obtained. Additionally, only the unpolarized case, in which all orbitals are occupied with one spin-up and one spin-down electron, will be considered here. For this spherical-type level structure, the number of electrons required for closed shells increases quickly. For example, the first six shells contain 2, 14, 38, 54, 66 and 114 states, respectively.

A finite number of electrons A in a finite box of volume Ω naturally leads to the characterization of an infinite system by its number density density $\rho = A/\Omega$. The average

inter-electron distance, or *Wigner-Seitz radius*, is defined as

$$\frac{4}{3}\pi r_s^3 = \frac{1}{\rho}, \quad r_s = \left(\frac{3}{4\pi\rho}\right)^{1/3}. \quad (3.54)$$

In practice, these calculations are defined by the total number of shells included in the basis, the number of electrons, and the Wigner-Seitz radius, usually given in units of the Bohr radius, $r_b = \frac{\hbar}{mc\alpha}$, where c is the speed of light and α is the fine-structure constant.

The last ingredient to this many-body calculation is the interaction between the electrons, the well-known Coulomb force. Using atomic units, where the elementary charge $e = 1$ and the Coulomb constant $\frac{1}{4\pi\varepsilon_0} = 1$, this potential is simply

$$V(\mathbf{r}_1, \mathbf{r}_2) = \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|}. \quad (3.55)$$

As mentioned in chapter 2, this potential can be utilized in second-quantization by computing antisymmetrized integrals over the basis states. In this case, the integrals have the form,

$$(2)H_{rs}^{pq} \equiv \int d\mathbf{r}_1 d\mathbf{r}_2 \phi_{\mathbf{k}p\sigma_p}^*(\mathbf{r}_1) \phi_{\mathbf{k}q\sigma_q}^*(\mathbf{r}_2) \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} [\phi_{\mathbf{k}r\sigma_r}(\mathbf{r}_1) \phi_{\mathbf{k}s\sigma_s}(\mathbf{r}_2) - \phi_{\mathbf{k}s\sigma_s}(\mathbf{r}_1) \phi_{\mathbf{k}r\sigma_r}(\mathbf{r}_2)]. \quad (3.56)$$

The symmetries of the Coulomb potential guarantee that the total linear momentum and total spin projection are conserved such that,

$$\mathbf{k}_p + \mathbf{k}_q = \mathbf{k}_r + \mathbf{k}_s \quad \text{and} \quad \sigma_p + \sigma_q = \sigma_r + \sigma_s. \quad (3.57)$$

The integral is relatively simple given the form of the basis functions. The result is given in

terms of the momentum transfer, $\mathbf{q}_1 = \mathbf{k}_p - \mathbf{k}_r$ and $\mathbf{q}_2 = \mathbf{k}_p - \mathbf{k}_r$,

$${}^{(2)}H_{rs}^{pq} = \frac{4\pi\hbar c\alpha}{\Omega} \left[\frac{\delta_{\sigma_p\sigma_r}\delta_{\sigma_q\sigma_s}}{|\mathbf{q}_1|^2} - \frac{\delta_{\sigma_p\sigma_s}\delta_{\sigma_q\sigma_r}}{|\mathbf{q}_2|^2} \right] \quad (3.58)$$

The last preparation step before performing the coupled cluster algorithm is the Hartree-Fock transformation. As with the pairing model, the single-particle orbitals are already eigenfunctions of the Fock operator, in this case because the translational invariance of the plane wave basis functions ensure that the HF terms of the form ${}^{(2)}H_{qi}^{pi}$ vanish due to momentum conservation, see Eq. (2.5). Therefore, the HF transformation consists simply of redefining the single-particle energies while the two-body interaction is left unchanged.

$$\begin{aligned} \varepsilon_p &= \epsilon_{\mathbf{k}p} + \sum_i {}^{(2)}H_{pi}^{pi} \\ V_{rs}^{pq} &= {}^{(2)}H_{rs}^{pq} \end{aligned} \quad (3.59)$$

As mentioned above, in the plane-wave basis, any single excitation from the reference state vanishes automatically due to momentum conservation so that $t_i^a = 0$. Therefore, it's necessary to include only double excitations (before adding triples, etc.). Therefore, calculations for the electron gas use the pseudo-linear form of the CCD equations (A.40) and an effective Hamiltonian that excludes single excitations in Eqns. (3.45)-(A.23). To explore the HEG equation-of-state, the total energy per electron can be calculated as a function of the Wigner-Seitz radius. In the limit $N, L \rightarrow \infty$, the plot in Fig. 3.5 represents the equation-of-state for a 3D electron gas at absolute zero. This curve can reveal many thermodynamic properties of the electron gas including the saturation density and saturation energy, which occur at the lowest point on the curve. The CCD results are compared

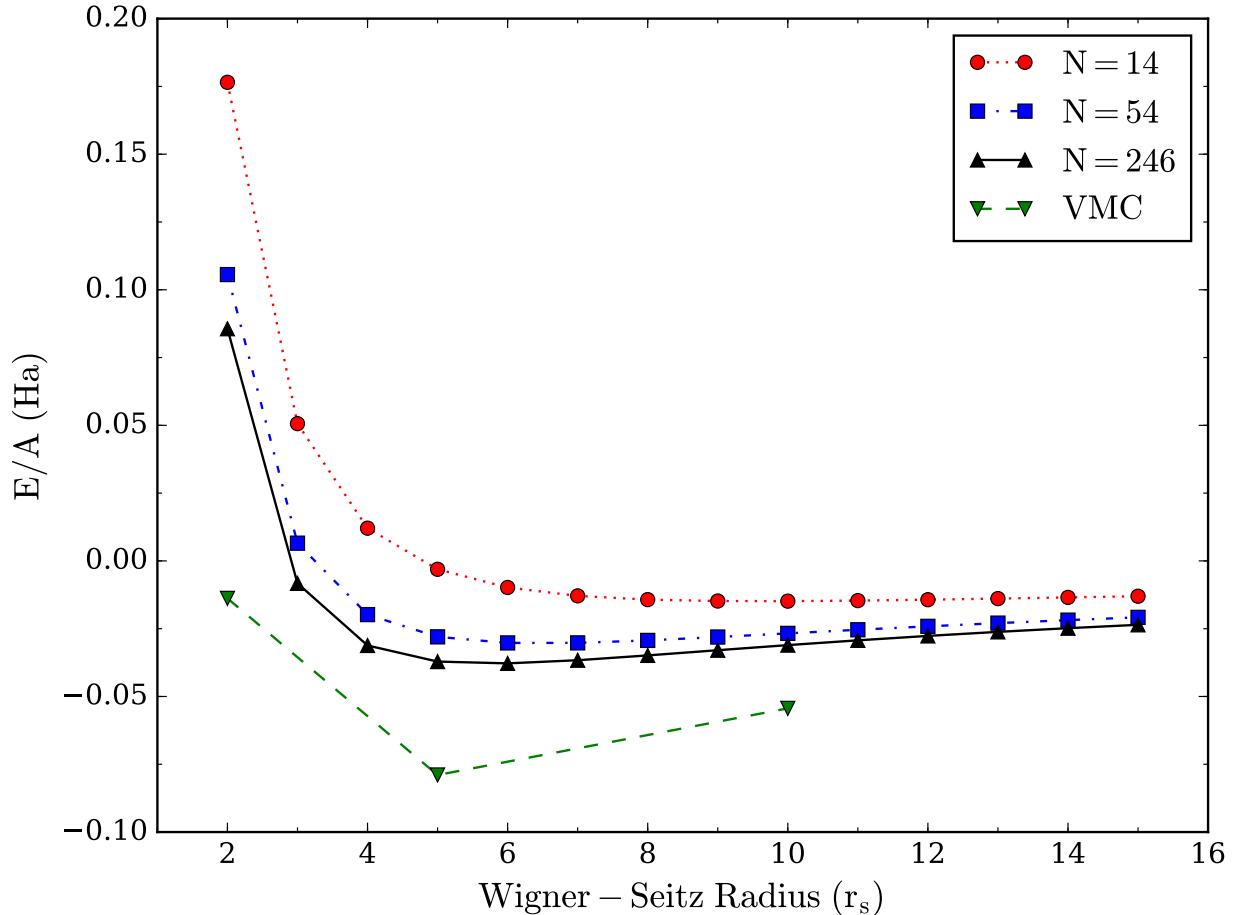


Figure 3.5: CCD energy per electron in Hartrees for the 3D homogeneous electron gas as function of the Wigner-Seitz radius in units of Bohr radii. The calculation used periodic boundary conditions and a basis with 25 shells, resulting in a total of 1238 single-particle states. Also plotted are the variational quantum Monte Carlo (VMC) results from [82].

with the quasi-exact results from variational quantum Monte Carlo calculations from [82]. The discrepancies between the saturation energies from the two methods can be partially attributed to an insufficient basis size. However, even an appropriate extrapolation to an infinite basis won't be able to recover all of the required correlations, which suggests that CCSDT might be necessary. Regardless of the value to the saturation energy, these CCD results do qualitatively reproduce the saturation radius at $r_s \approx 5.0$.

3.6 Coupled Cluster for Finite Nuclei

The main purpose of this work is to calculate properties of atomic nuclei with coupled cluster theory. From a many-body perspective, the main process is computing the converged cluster amplitudes and thus the important correlations of the system. These amplitudes comprise the CC similarity transformation, which is versatile for constructing any effective operator, such as the Hamiltonian, that can act on the correlated system. Therefore, the first step in calculating beta-decay properties of nuclei is solving for the ground-state wave function of specific closed-shell nuclei.

3.6.1 Harmonic Oscillator Basis

Calculations of finite nuclei follow the basic structure of the algorithms used for the pairing model and the homogeneous electron gas, but they also differ in some significant ways. Like the other examples, the first step is to construct a proper single-particle basis and reference state. Because the nuclear Hamiltonian conserves angular momentum and parity, it's useful to construct orbits that are eigenfunctions of these operators. For a system with no external potential, like the electron gas, this suggests a basis made of plane waves. However, plane waves do not represent the bound states of a nucleus very well. This property can be satisfied by introducing a fictitious external potential that mimics the mean field from the collection of nucleons. Some phenomenological potentials, like the Woods-Saxon potential, properly consider the resonance and continuum states of realistic nuclei in addition to the bound states. Many-body techniques discussed in this work have been applied to model spaces that include all three types of single-particle states [85, 52] with some success. However, for the many-body states considered in this work, it's sufficient to consider only bound single-particle states. Therefore, the nuclear basis will be constructed from the isotropic harmonic

oscillator,

$$V(r) = \frac{1}{2}m\omega^2 r^2. \quad (3.60)$$

An eigenstate of the harmonic oscillator potential is defined by its principal quantum number n and its orbital angular momentum quantum number l , which is denoted by the letters s, p, d, f... for the values $l = 0, 1, 2, 3\dots$ respectively. Because of spin-orbit terms in the nuclear Hamiltonian, the orbital angular momentum is coupled to a particle spin to a total angular momentum of $j = |\mathbf{l} + \mathbf{s}|$, which results in a degeneracy of $2j + 1$ for each orbit. This basis does not provide any simplification to eliminate single excitations, so CCSD will be used for all the following calculations. A schematic version of this single-particle basis is shown in Fig. 3.6. The shell structure of this basis is characterized by the energy quantum numbers, $e = 2n + l$, of the HO single-particle spectrum. This can be used to define the maximum-energy shell and the size of a HO basis with the parameter e_{\max} .

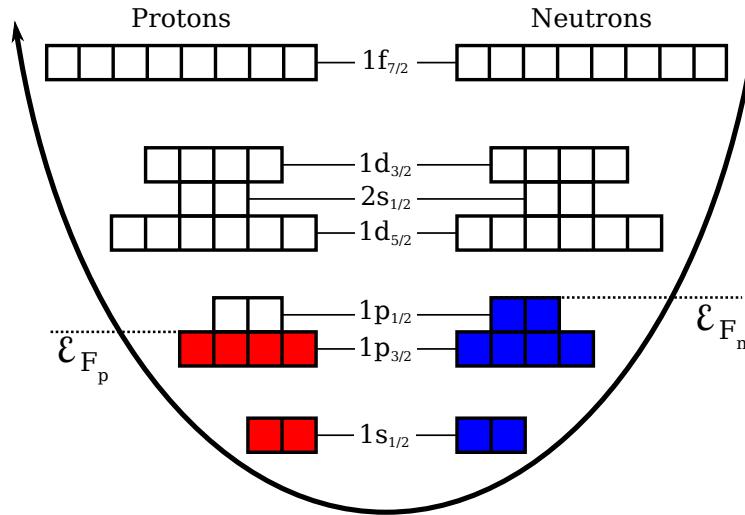


Figure 3.6: A schematic illustration of the harmonic oscillator basis used for calculations of nuclei. Shown is an example of a initial reference state for carbon-14, with 6 protons filled to the $p3/2$ -subshell closure and 8 neutrons filled to the $p1$ -shell closure. See text for details on the single-particle states.

One issue with this construction, is that while the single-particle orbits are eigenstates of

the angular momentum operator and localized to the external potential, they are not translationally invariant, which is required by the nuclear Hamiltonian. This means that there is a fictitious center-of-mass kinetic energy which must be removed from the Hamiltonian. The COM kinetic energy can be written as the sum of one- and two-body pieces,

$$\hat{T}_{\text{cm}} = \frac{\mathbf{P}_{\text{cm}}}{2mA} = \sum_{pq}^A \frac{\mathbf{p}_p \cdot \mathbf{p}_q}{2mA} = \sum_p^A \frac{\mathbf{p}_p^2}{2mA} + \sum_{p < q}^A \frac{\mathbf{p}_p \cdot \mathbf{p}_q}{mA}. \quad (3.61)$$

The one-body piece is just a scaled form of the original kinetic energy operator, and the two-body piece is given in a similar form to the original two-body Hamiltonian. Both can be integrated into matrix elements like Eq. (2.2),

$$\begin{aligned} {}^{(1)}T_q^p &\equiv \int d\mathbf{r}_1 \phi_p^*(\mathbf{r}_1) \frac{\mathbf{p}_1^2}{2mA} \phi_q(\mathbf{r}_1), \\ {}^{(2)}T_{rs}^{pq} &\equiv \int d\mathbf{r}_1 d\mathbf{r}_2 \phi_p^*(\mathbf{r}_1) \phi_q^*(\mathbf{r}_2) \frac{\mathbf{p}_1 \cdot \mathbf{p}_2}{mA} [\phi_r(\mathbf{r}_1) \phi_s(\mathbf{r}_2) - \phi_s(\mathbf{r}_1) \phi_r(\mathbf{r}_2)]. \end{aligned} \quad (3.62)$$

Subtracting the COM kinetic energy results in the *intrinsic* Hamiltonian for finite nuclear systems,

$$\begin{aligned} \hat{H}_{\text{in}} &= \left(1 - \frac{1}{A}\right) \sum_{pq} {}^{(1)}T_q^p \hat{a}_p^\dagger \hat{a}_q + \frac{1}{4} \sum_{pqrs} \left({}^{(2)}H_{rs}^{pq} - {}^{(2)}T_{rs}^{pq} \right) \hat{a}_p^\dagger \hat{a}_q^\dagger \hat{a}_s \hat{a}_r \\ &\quad + \frac{1}{36} \sum_{pqrsstu} {}^{(3)}H_{stu}^{pqr} \hat{a}_p^\dagger \hat{a}_q^\dagger \hat{a}_r^\dagger \hat{a}_u \hat{a}_t \hat{a}_s + \dots, \end{aligned} \quad (3.63)$$

This form of the bare Hamiltonian (up to the three-body force) is used in the Hartree-Fock transformation. Then, after normal-ordering, the three-body piece is discarded, which is referred to as a NN+3N-induced interaction. The use of a localized external potential has further complications involving the COM wave function that are discussed in section 3.7.

A special property of this single-particle basis that can be exploited to reduce the computational complexity of the problem is the degeneracy of each orbital, due to the angular momentum projection of each single-particle state, $m_j = \{-j, -j+1, \dots, j-1, j\}$. According to the Wigner-Eckart theorem [133, 36], the geometrical component of a wave function, dependent on its projection m_j , can be isolated as a Clebsch-Gordon coefficient. Because these coefficients have compact summation rules, any diagram and corresponding sum can be written in terms of the j -orbitals instead of the single-particle m_j states, commonly known as J -scheme and M -scheme, respectively. Calculations in J -scheme require complicated angular momentum coupling, detailed in the appendix ??, but involve roughly an order of magnitude fewer states compared with an M -scheme calculation in the same model space.

3.6.2 The Nuclear Interaction

Perhaps the most important component in nuclear structure calculations, and also perhaps the most easily overlooked component from a many-body perspective, is the nuclear Hamiltonian. Further complicated by the composite nature of protons and neutrons, bound by gluon exchange within the nucleon, the inter-nucleon interaction is a residual force of virtual pion exchanges and other, more exotic processes. Early *ab initio* calculations avoided this complexity by using phenomenological interactions, tuned to reproduce certain properties of a nucleus. These phenomenological forces were effectively used for calculations using shell-model CI and density-functional theory, but were restricted by the conditions of the fitted parameters.

These problems, along with the success of quantum field theories in high-energy physics, motivated the effort to describe the inter-nucleon interaction in terms of the underlying theory of the strong force, quantum chromodynamics (QCD) [60, 78]. However, while cal-

culations of nuclei in terms of their constituent quarks using lattice QCD have made some progress with increases in computing power, they have been confined to few nucleon systems [8]. The problem is finding a way to express the high-energy QCD interactions as low-energy forces between nucleons. Such a problem, containing two vastly different scales, can be rewritten as an effective theory.

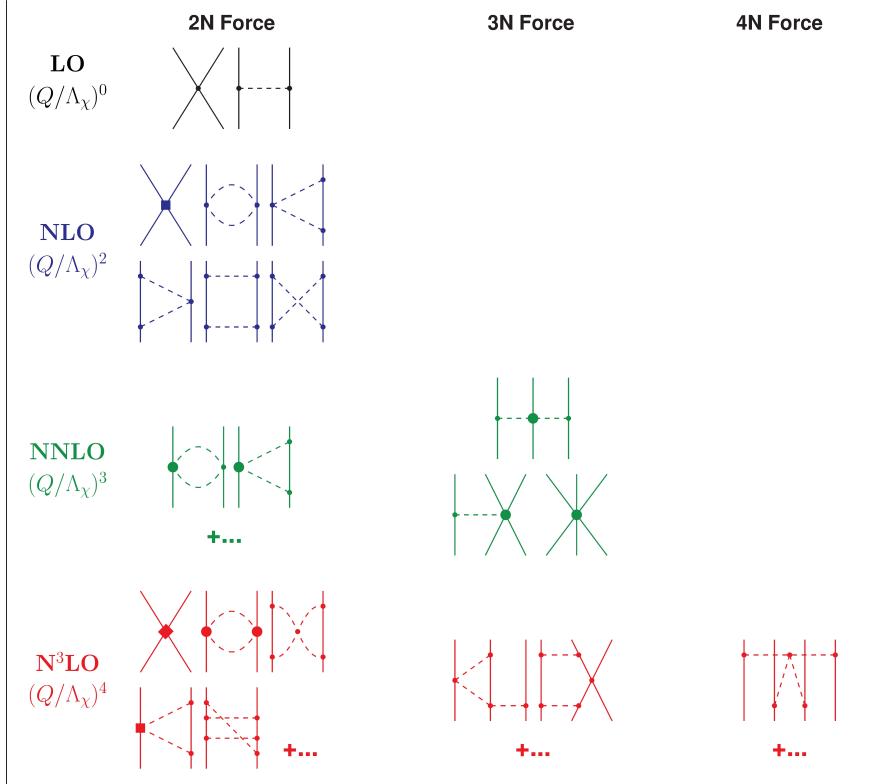


Figure 3.7: Diagrammatic form of the chiral EFT expansion up to N^3LO . The solid lines represent nucleons and the dashed lines represent pions. The different vertices represent higher-order interactions. Figure taken from [84].

Chiral effective field theory (EFT), which exploits the large difference in scales between the low-energy regime of nuclear physics and the high-energy regime of QCD, is built from a general Lagrangian consistent with the broken chiral symmetry of QCD [41, 84]. This broken symmetry, a consequence of non-zero quark masses, results in several hadronic structures including protons, neutrons, and mesons, the lightest of which is the pion, $m_\pi \approx 140\text{MeV}/c^2$

[10]. This can be exploited by systematically writing a Lagrangian as the sum of pion exchanges of increasing order. Additional contact interactions, which represent exchanges of heavier mesons, are also included and must be fit to low-energy nuclear data. The hierarchy of EFT terms, which contain 3N and higher many-body forces, are ordered by power counting the expansion term (m_π/Λ), where Λ is an energy cutoff between the low- and high-energy scales, and is shown up to N³LO in Fig. 3.7.

This work exclusively employs the NN force of the N³LO interaction from Entem and Machleidt with a cutoff of $\Lambda = 500$ MeV [40]. For most calculations, this interaction is coupled with the N²LO 3N interaction from Navrátil with a cutoff of $\Lambda = 400$ MeV [89]. This NN+3N(400) interaction is successful at reproducing low- and medium-mass nuclei, but begins to overbind beyond the *sd*-shell. As mentioned in the introduction, these bare Hamiltonians exhibit strong repulsion at short ranges among high-momentum states. Therefore to soften the interaction, the similarity renormalization group method is used to integrate high-momentum modes out of the interaction while preserving observables [17, 104].

3.6.3 Ground-State Results for Nuclei

The main object of this section is to demonstrate the validity of all the ingredients which have been discussed so far: the harmonic oscillator basis, the NN+3N(400) chiral interaction, and the *J*-scheme CCSD algorithm. To accomplish this, calculations for different nuclei will be compared to the corresponding experimental values. Additionally, results for different input parameters will be presented to verify that the observables are independent of non-physical variables. Once again, all results are computed with a HF-optimized basis, see section 2.5.

First, the ground-state energies should be independent of the SRG cutoff parameter

λ_{SRG} . While the SRG evolution should preserve any observables, the renormalization process induces 3N and higher-body forces which can be missed by truncations of the many-body method in both the cluster amplitudes and the Hamiltonian. The trade-off here is that larger cutoff parameters produce interactions that contain higher-momentum components, which reduce a system's convergence properties, but induce fewer many-body forces, so that systems can be accurately described with fewer correlations. Conversely, a smaller cutoff parameter means that solutions can be more easily converged, but it also requires a many-body method that includes more correlations or higher-order forces [103]. To show this effect, the ground state for oxygen-16 is shown for different cutoff parameters and for both the NN and NN+3N-induced interactions. Accounting for both the small dependence on the SRG cutoff parameter and the minor inaccuracies from the truncations made to the cluster operator and the Hamiltonian, the rest of this work will use the NN+3N(400)-induced interaction with an SRG cutoff parameter of $\lambda_{\text{SRG}} = 2.0 \text{ fm}^{-1}$.

Next, any nuclear observables calculated with this framework should be independent of the fictitious confining potential. This can be verified by showing the ground-state energies of various nuclei as a function of the underlying harmonic oscillator energy, $\hbar\omega$. Convergence is reached by increasing the size of the model space until the resulting curve is flat. Figure 3.9 shows the convergence for the *doubly-magic*, $N = Z$ nuclei, ${}^4\text{He}$, ${}^{16}\text{O}$, ${}^{20}\text{Ca}$, and ${}^{56}\text{Ni}$, where both protons and neutrons fill the same major shell closure. All the results converge to a variance of $< 1\%$ at $e_{\text{max}} = 12$ for intermediate values of $\hbar\omega$. While a larger model space is always desirable, this level of variance justifies the use of $e_{\text{max}} = 12$ for post-CC calculations. Additionally, these results show the limitations of the NN+3N(400) interaction, as overbinding increases with the system size, where the ground-state energies of ${}^{20}\text{Ca}$ and ${}^{56}\text{Ni}$ differ from their experimental binding energies by $\sim 8\%$ and $\sim 13\%$, respectively.

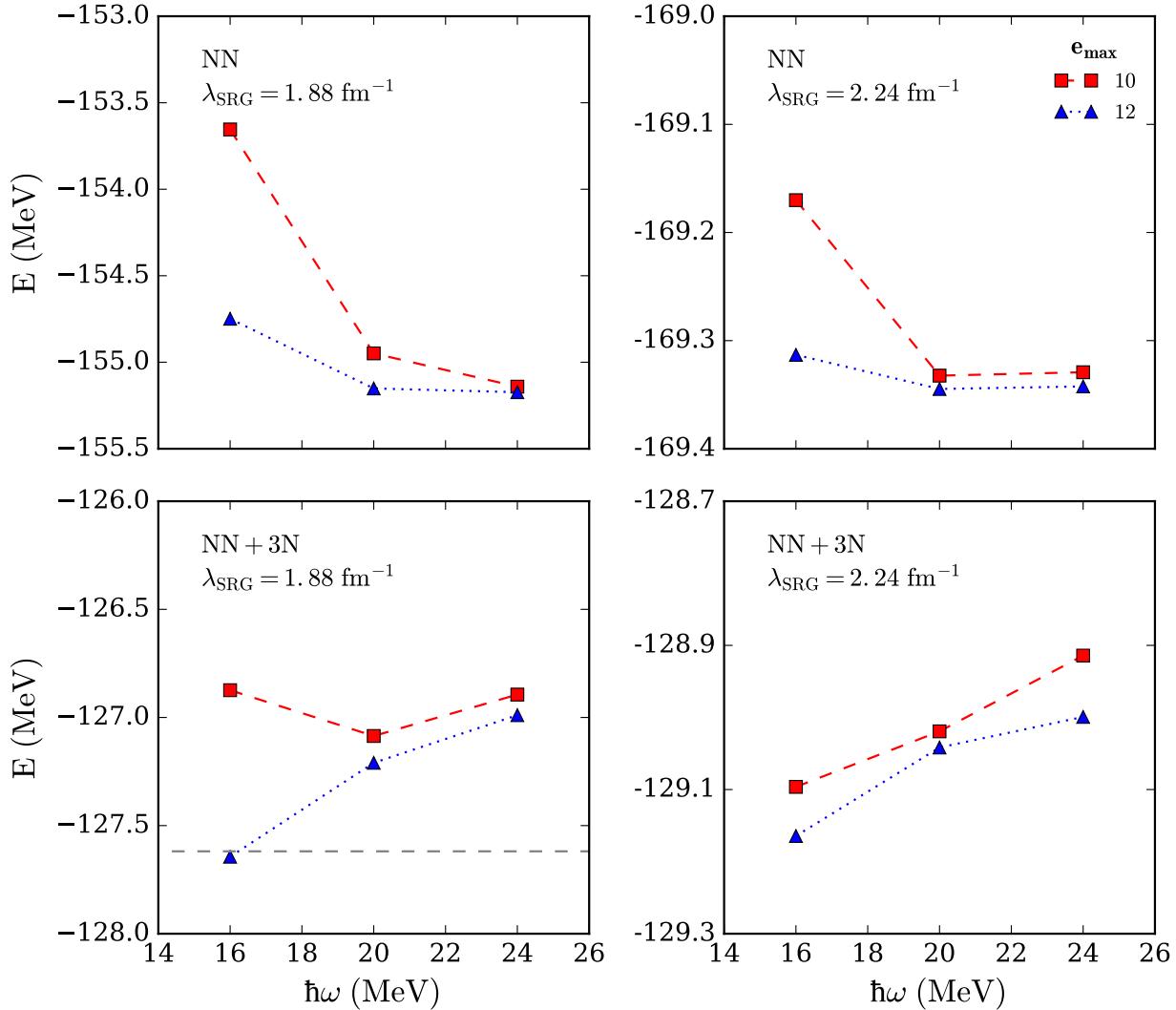


Figure 3.8: Ground-state energies for ^{16}O for the EM N³LO NN only interaction and with the added 3N interaction from Navrátil, both SRG softened with $\lambda_{\text{SRG}} = 1.88, 2.24 \text{ fm}^{-1}$. The energies are plotted for $e_{\max} = 10, 12$. The most obvious difference is between the NN and NN+3N calculations, showing the importance of including 3N forces. The differences between the cutoff parameters are resolved within $\sim 1\%$ with the inclusion of 3N forces and can be rectified further by including additional correlations or full 3N forces. The experimental binding energy is shown with the grey dashed line.

The ground-state results are also shown for singly-magic nuclei, where either the protons or neutrons fill a sub-shell closure. This has the potential complication of a vanishing energy gap between the hole and particle states, like the picture in Fig. ??, which causes undefined behavior in the CC algorithm (see section 3.4). However, the subshell orbitals repel each

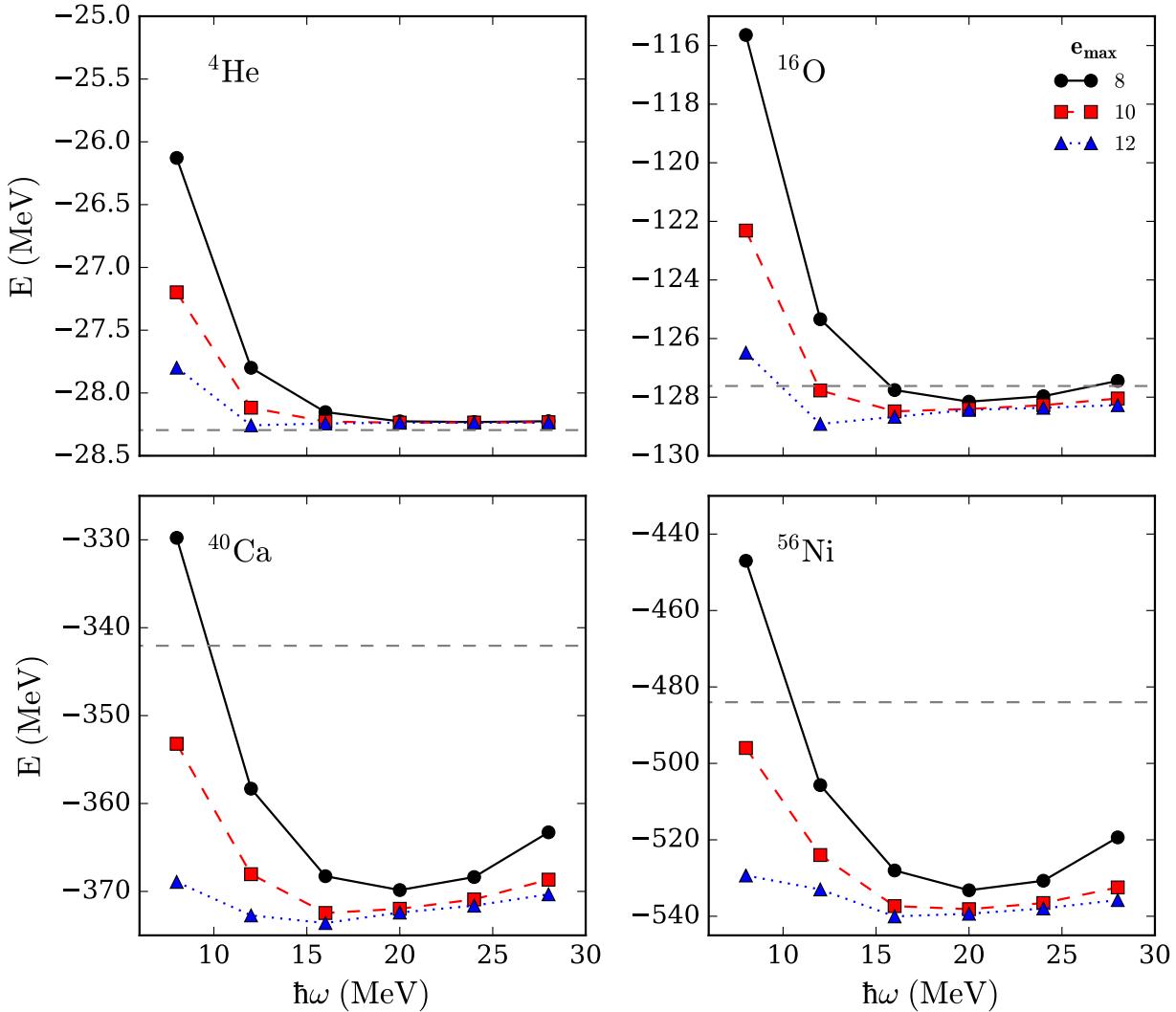


Figure 3.9: Ground-state energies for doubly magic nuclei as a function of the harmonic oscillator energy $\hbar\omega$ with the NN+3N(400) interaction, SRG softened with $\lambda_{\text{SRG}} = 2\text{fm}^{-1}$. The energies are plotted for $e_{\max} = 8, 10, 12$, showing the convergence as the model space increases. The results are independent of the underlying oscillator frequency to $\sim 1\%$ for $e_{\max} = 12$. The grey dashed line is the experimental binding energy. The overbinding of this interaction becomes apparent as the system size increases.

other when transformed during the Hartree-Fock algorithm [79], so these systems are valid in some cases. The ground-state energies for ^{14}C , ^{22}O , and ^{34}Si are plotted as a function of the underlying oscillator potential in Fig. 3.10. The smaller energy gap involved in these systems results in stronger excitations missed by the CCSD approximation, causing further deviations from the experimental values.

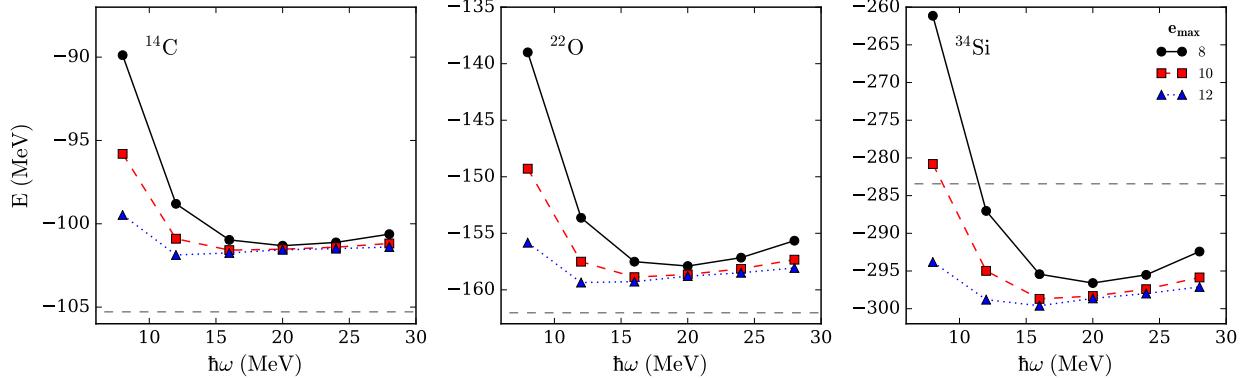


Figure 3.10: Ground-state energies for singly magic nuclei as a function of the harmonic oscillator energy $\hbar\omega$ with the NN+3N(400) interaction, SRG softened with $\lambda_{\text{SRG}} = 2\text{fm}^{-1}$. The energies are plotted for different e_{\max} . The results are independent of the underlying oscillator frequency to $\sim 1\%$ for $e_{\max} = 12$. The grey dashed line is the experimental binding energy. These results underbind with respect to their doubly-magic counterparts in Fig. 3.9.

3.7 Ground-State Center-of-Mass Factorization

While an intrinsic Hamiltonian can be built by removing the center-of-mass (COM) kinetic energy, Eq. (3.63), there is still an inconsistency between the translational invariance of the underlying harmonic oscillator basis and translationally-invariant nuclear many-body states [81, 48]. This inconsistency can materialize in certain calculations in the form of spurious, non-physical states.

Of course, this problem can be avoided by using more complicated basis states that obey translational invariance, such as the use of Jacobi coordinates, but such methods are limited to few-body problems [14, 92]. Another possible solution is to use the harmonic oscillator basis in an untruncated space of Slater determinants up to a certain harmonic oscillator shell. Known as the N_{\max} space, this treatment can be successfully applied within no-core shell model calculations [91]. However, the factorial scaling of this method restricts its use to light nuclei. It can be shown that in the N_{\max} space, the eigenstates of the intrinsic Hamiltonian are also eigenstates of the COM Hamiltonian, and any state perfectly factorizes into a COM

component and a translationally-invariant, intrinsic component,

$$|\Psi\rangle = |\Psi_{\text{in}}\rangle |\Psi_{\text{cm}}\rangle. \quad (3.64)$$

This factorization results in a compound energy spectrum, where the intrinsic component of the spectrum is degenerate for each COM excitation. Therefore, the intrinsic spectrum can be recovered by offsetting the COM Hamiltonian by the corresponding excitation energies, such that the COM energies vanish, $E_{\text{cm}} = 0$. However, with truncated methods like CCSD, this factorization is not guaranteed, and COM energies, no longer eigenenergies of the COM Hamiltonian, can take on values $E_{\text{cm}} \neq 0$. In this case, the intrinsic spectrum is contaminated with nonphysical, *spurious* states.

Because the specific form is irrelevant [130], the shifted COM Hamiltonian can be assumed to take the form of a harmonic trap with a oscillator strength of $\hbar\tilde{\omega}$, not necessarily equal to the oscillator strength of the underlying basis $\hbar\tilde{\omega}$,

$$\hat{H}_{\text{cm}}(\tilde{\omega}) = \frac{\mathbf{P}_{\text{cm}}}{2mA} + \frac{1}{2}mA\tilde{\omega}^2\mathbf{R}_{\text{cm}} - \frac{3}{2}\hbar\tilde{\omega}, \quad (3.65)$$

offset by the ground-state energy, $\frac{3}{2}\hbar\tilde{\omega}$. In the N_{max} space, the factorization in Eq. (3.64) occurs regardless of $\hbar\tilde{\omega}$, while in truncated methods like CCSD, the COM oscillator strength is a free parameter which can be used to probe the level of COM contamination. If a frequency exists such that $E_{\text{cm}}(\tilde{\omega}) \approx 0$, then the wave function is approximately factorized, and the COM wave function is in its ground state [54, 68].

The COM energy, E_{cm} , can be calculated by using a version of the Hellmann-Feynman theorem by adding the COM Hamiltonian as a perturbation and computing the difference

quotient [34, 42],

$$E_{\text{cm}}(\tilde{\omega}) \equiv \langle \Psi | \hat{H}_{\text{cm}} | \Psi \rangle \approx \frac{1}{2\delta} \left(\langle \Psi | \hat{H} + \delta \hat{H}_{\text{cm}}(\tilde{\omega}) | \Psi \rangle - \langle \Psi | \hat{H} - \delta \hat{H}_{\text{cm}}(\tilde{\omega}) | \Psi \rangle \right). \quad (3.66)$$

Because the operator \mathbf{R}_{cm} depends only on the underlying single-particle basis regardless of the COM oscillator frequency, it can be rewritten in terms of \hat{H}_{cm} to find the relationship between ω and $\tilde{\omega}$,

$$\frac{1}{\tilde{\omega}^2} \left(\hat{H}_{\text{cm}}(\tilde{\omega}) - \hat{T}_{\text{cm}} + \frac{3}{2}\hbar\tilde{\omega} \right) = \frac{1}{\omega^2} \left(\hat{H}_{\text{cm}}(\omega) - \hat{T}_{\text{cm}} + \frac{3}{2}\hbar\omega \right). \quad (3.67)$$

Using the known value $\langle \Psi | \hat{T}_{\text{cm}} | \Psi \rangle = \frac{3}{4}\hbar\tilde{\omega}$ and the requirement that $E_{\text{cm}}(\tilde{\omega}) = 0$ gives the following relation that relates the COM oscillator frequency to the underlying basis frequency,

$$\hbar\tilde{\omega} = \hbar\omega + \frac{2}{3}E_{\text{cm}}(\omega) \pm \sqrt{\left(\frac{2}{3}E_{\text{cm}}(\omega)\right)^2 + \frac{4}{3}\hbar\omega E_{\text{cm}}(\omega)}. \quad (3.68)$$

The ground-state COM energies are plotted for ^{16}O and ^{40}Ca using the COM Hamiltonian with two different oscillator strengths in Fig. 3.11: that of the underlying basis, $\hbar\omega$, and one of the two solutions to Eq. (3.68), $\hbar\tilde{\omega}_\pm$. Of the two $\hbar\tilde{\omega}_\pm$, which are shown as the solutions to Eq. (3.68) in the inset, one typically results in a large COM energy while the other vanishes, which is plotted. Because the COM energies approximately vanish regardless of the underlying basis frequency, the COM wave function is in its ground state and approximately factorized from the intrinsic nuclear wave function.

Unfortunately, when intrinsic states are coupled to COM excited-states, they can contaminate the spectrum of intrinsic states coupled to the COM ground-state. These spurious states can be essentially removed from the ground-state spectrum with the Lawson-Gloeckner

method [48]. When the proper COM oscillator strength is chosen such that the COM ground-state energy vanishes, the COM Hamiltonian can be added to the intrinsic Hamiltonian at an arbitrarily large scale, β , without changing the ground-state spectrum,

$$\hat{H}_{\text{in}} \rightarrow \hat{H}_{\text{in}} + \beta \hat{H}_{\text{cm}}. \quad (3.69)$$

When β is arbitrarily large, eigenenergies of intrinsic states coupled to COM excited states will increase by the COM energy quanta, $\beta\hbar\tilde{\omega}$, such that they are removed from the range of low-lying states of interest. The method will be used to remove spurious states from the spectra of open-shell states in section 4.

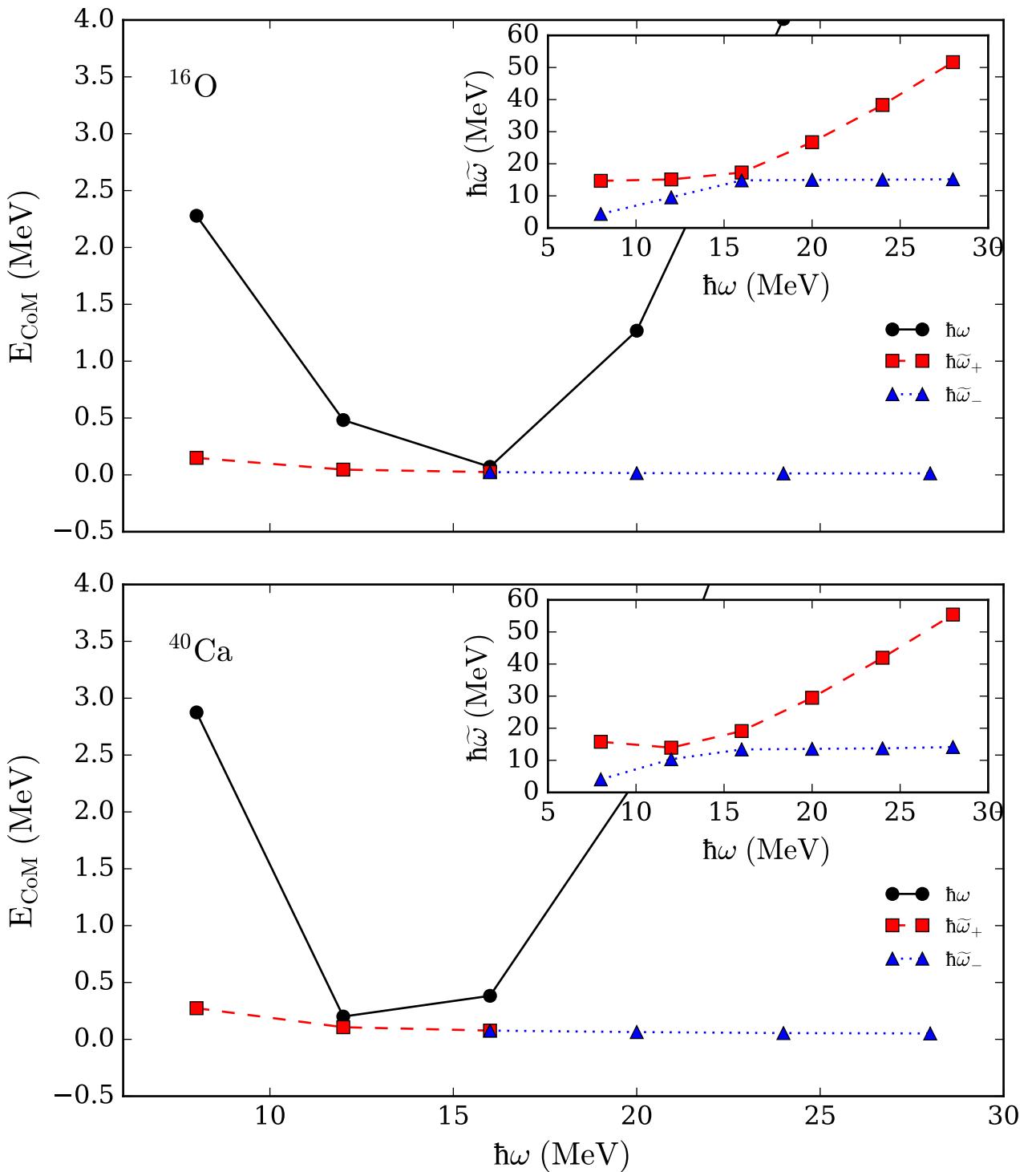


Figure 3.11: Ground-state COM energies, Eq. (3.65), for ^{16}O and ^{40}Ca at varies harmonic oscillator frequencies with the NN+3N(400)-induced with $\lambda_{\text{SRG}} = 2.0 \text{ fm}^{-1}$ at $e_{\text{max}} = 12$. Using the proper COM oscillator frequencies shows the approximate factorization of Eq. (3.64).

Chapter 4

Equation-of-Motion Method

Once the ground state energy and the effective Hamiltonian has been calculated, any further properties of the ground state can be calculated using the correlated wave function written as a Slater determinant expansion in the form of Eqn. [?]. However, many of the most interesting processes in nuclear physics involve excited-state properties. Additionally, because the coupled cluster method requires a doubly closed-shell reference state, most topics in nuclear physics that can benefit from an ab initio description are unreachable with the standard coupled cluster approach alone.

These restrictions motivates a class of techniques known as equations-of-motion methods (EOM). Applied with the CC effective Hamiltonian, the *coupled cluster equation-of-motion* (CC-EOM) method begins with a standard CC calculation of a closed-shell ground state. Then, EOM target states are built onto the correlated ground-state wave function in the same way that CI states were built from the reference state, Eqn. (2.44). Unfortunately, because the CCSD similarity transformation only decouples the ground state from $1p\text{-}1h$ and $2p\text{-}2h$ excitations, these target states are still coupled to each other. Therefore, capturing the relevant correlations to describe EOM states involves a CI-like diagonalization of the effective Hamiltonian.

4.1 Equations-of-Motion Method

4.2 Equation-of-Motion States

The CC equations-of-motion states are built by applying a particular type of excitation operator to the correlated ground-state wave function.

$$|\Psi_\mu\rangle = \hat{R}_\mu |\Psi\rangle = \hat{R}_\mu e^{\hat{T}} |\Phi_0\rangle, \quad (4.1)$$

where \hat{R}_μ can be written as a linear combination of strings of creation and annihilation operators. The form of these strings is determined by the structure of the desired target state. For example, excited states maintain the particle number of the reference state and ground state so that the constituent operator strings are $kp-kh$ operators. The A -particle excited-state EOM operator \hat{R}_μ^A is,

$$\hat{R}_\mu^A = {}^\mu r_0 + \sum_{ai} {}^\mu r_i^a \left\{ \hat{a}_a^\dagger \hat{a}_i \right\} + \frac{1}{4} \sum_{abij} {}^\mu r_{ij}^{ab} \left\{ \hat{a}_a^\dagger \hat{a}_b^\dagger \hat{a}_j \hat{a}_i \right\} + \dots \quad (4.2)$$

Additionally, particle-attached states can be reached with strings of the form p, pph, ppphh, etc... These operator strings increase the number of particles by one and gives the *particle-attached equation-of-motion* PA-EOM operator \hat{R}_μ^{A+1} ,

$$\hat{R}_\mu^{A+1} = \sum_a {}^\mu r^a \left\{ \hat{a}_a^\dagger \right\} + \frac{1}{2} \sum_{abi} {}^\mu r_i^{ab} \left\{ \hat{a}_a^\dagger \hat{a}_b^\dagger \hat{a}_i \right\} + \dots \quad (4.3)$$

Lastly, particle-removed states can be reached with strings of the form h, hhp, hhpp, etc... These operator strings decrease the number of particles by one and gives the *particle-removed*

equation-of-motion PR-EOM operator \hat{R}_μ^{A-1} ,

$$\hat{R}_\mu^{A-1} = \sum_i {}^\mu r_i \{ \hat{a}_i \} + \frac{1}{2} \sum_{aij} {}^\mu r_{ij}^a \left\{ \hat{a}_a^\dagger \hat{a}_j \hat{a}_i \right\} + \dots \quad (4.4)$$

$$\hat{L}_\mu^{A+1} = \sum_a {}^\mu l_a \{ \hat{a}_a \} + \frac{1}{2} \sum_{abi} {}^\mu l_{ab}^i \left\{ \hat{a}_i^\dagger \hat{a}_b \hat{a}_a \right\} + \dots \quad (4.5)$$

$$\hat{L}_\mu^{A-1} = \sum_i {}^\mu l^i \left\{ \hat{a}_i^\dagger \right\} + \frac{1}{2} \sum_{aij} {}^\mu l_a^{ij} \left\{ \hat{a}_i^\dagger \hat{a}_j^\dagger \hat{a}_a \right\} + \dots \quad (4.6)$$

$$\langle \hat{L}_\mu | \hat{R}_\nu \rangle = \delta_{\mu\nu} \quad (4.7)$$

Here, ${}^\mu r$ are the normal-ordered matrix elements of \hat{R}_μ .

Substitution of Eq. (4.12) into the energy eigenvalue problem

$$\hat{H} |\Psi_\mu\rangle = E_\mu |\Psi_\mu\rangle, \quad (4.8)$$

$$\begin{aligned} \hat{H}_N |\Psi_\mu\rangle &= \Delta E_\mu |\Psi_\mu\rangle \\ \hat{H}_N \hat{R}_\mu e^{\hat{T}} |\Phi_0\rangle &= \Delta E_\mu \hat{R}_\mu e^{\hat{T}} |\Phi_0\rangle \\ e^{-\hat{T}} \hat{H}_N \hat{R}_\mu e^{\hat{T}} |\Phi_0\rangle &= e^{-\hat{T}} \Delta E_\mu \hat{R}_\mu e^{\hat{T}} |\Phi_0\rangle \end{aligned} \quad (4.9)$$

$$\begin{aligned} e^{-\hat{T}} \hat{H}_N e^{\hat{T}} \hat{R}_\mu |\Phi_0\rangle &= \Delta E_\mu e^{-\hat{T}} e^{\hat{T}} \hat{R}_\mu |\Phi_0\rangle \\ \bar{H}_N \hat{R}_\mu |\Phi_0\rangle &= \Delta E_\mu \hat{R}_\mu |\Phi_0\rangle \end{aligned} \quad (4.10)$$

$$\begin{aligned}
\bar{H}_N \hat{R}_\mu |\Phi_0\rangle &= \Delta E_\mu \hat{R}_\mu |\Phi_0\rangle \\
-\hat{R}_\mu \bar{H}_N |\Phi_0\rangle &= \hat{R}_\mu \Delta E |\Phi_0\rangle \\
\longrightarrow [\bar{H}_N, \hat{R}_\mu] |\Phi_0\rangle &= (\Delta E_\mu - \Delta E) \hat{R}_\mu |\Phi_0\rangle = \omega_\mu \hat{R}_\mu |\Phi_0\rangle
\end{aligned} \tag{4.11}$$

$$\langle \Psi_\mu | = \langle \Phi_0 | \hat{L}_\mu \tag{4.12}$$

$$\begin{aligned}
\langle \Psi_\mu | \bar{H}_N &= \langle \Psi_\mu | \Delta E_\mu \\
\langle \Phi_0 | \hat{L}_\mu \bar{H}_N &= \langle \Phi_0 | \hat{L}_\mu \Delta E_\mu,
\end{aligned} \tag{4.13}$$

$$\begin{aligned}
[\bar{H}_N, \hat{R}_\mu] &= \omega_\mu \hat{R}_\mu \\
\hat{L}_\mu \bar{H}_N &= \hat{L}_\mu \Delta E_\mu
\end{aligned} \tag{4.14}$$

gives

$$[\hat{H}, \hat{R}_\mu] |\Psi\rangle = \pm \varepsilon_u^{(\pm)} \hat{X}_u^{(N\pm 1)} |\Psi_0^{(N)}\rangle, \tag{4.15}$$

which constitutes a generalized eigenvalue problem for the amplitudes x , where $\varepsilon_u^{(\pm)}$ are the single-particle addition (+) and removal (-) energies. The quality of this calculation depends on the ansatz for the N -particle ground state, as well as the systematically improvable truncation on the ladder operators. In this work we include 1p and 2p1h excitations in the $N + 1$ ladder operator and likewise 1h and 2h1p operators for the $N - 1$ ladder operators.

After a single-reference ground state IM-SRG calculation, the Hamiltonian has been

rotated such that the reference state is an eigenfunction with corresponding eigenvalue $E_0^{(N)}$, which is the correlated N -particle ground state energy. The EOM equation is therefore

$$[\bar{H}, \bar{X}_u^{(N\pm 1)}]|\Phi_0^{(N)}\rangle = \pm \varepsilon_u^{(\pm)} \bar{X}_u^{(N\pm 1)} |\Phi_0^{(N)}\rangle, \quad (4.16)$$

where bars denote rotated operators. Now the reference state is used in place of the bare correlated ground state. The ground state IM-SRG procedure has implicitly re-summed contributions from higher order excitations (3-particle-2-hole, 2-particle-3-hole, 2-particle-3-hole, 4-particle-3-hole, ...) into the lower order amplitudes of the ladder operators (1-particle-0-hole, 0-particle-1-hole, 2-particle-1-hole, 1-particle-2-hole).

Despite these gains, the EOM calculation is still a partial diagonalization method, limited by the truncation to 2-particle-1-hole and 1-particle-2-hole operators. We expect $N + 1$ (or $N - 1$) states to be described appropriately by EOM-IM-SRG if their wavefunctions are dominated by 1-particle-0-hole (or 0-particle-1-hole) contributions in the rotated frame. We use partial norms of the EOM ladder operators to estimate these contributions:

$$n_{1\text{-particle}} = \sqrt{\sum_a |\mu_r a|^2}, \quad (4.17)$$

$$n_{1\text{-hole}} = \sqrt{\sum_i |\mu_r i|^2}. \quad (4.18)$$

Large single particle partial norms indicate that the EOM truncation is reasonable for the relevant state. States with lower single particle norms should be treated with a higher EOM approximation, which can be accomplished directly or perturbatively [95].

Like EOM-IM-SRG, the equations-of-motion technique can be applied after a CC ground-state calculation, by using the CC effective Hamiltonian. Here, the non-Hermitian nature

of \bar{H}_{CC} becomes apparent. In this case, in addition to constructing excitation ladder operators Eq. (??) and Eq. (??) that correspond to the right-eigenvectors of the generalized eigenvalue problem Eq. (4.16), there exist analogous de-excitation ladder operators, $\hat{L}^{(N+1)}$ and $\hat{L}^{(N-1)}$, that correspond to the left-eigenvectors,

$$\begin{aligned}\hat{L}_u^{(N+1)} &= \sum_a l_a^{(u,+)} \{\hat{a}_a\} + \frac{1}{2} \sum_{iab} l_{iab}^{(u,+)} \left\{ \hat{a}_i^\dagger \hat{a}_b \hat{a}_a \right\} + \dots, \\ \hat{L}_u^{(N-1)} &= \sum_i l_i^{(u,-)} \left\{ \hat{a}_i^\dagger \right\} + \frac{1}{2} \sum_{ija} l_{ija}^{(u,-)} \left\{ \hat{a}_i^\dagger \hat{a}_j^\dagger \hat{a}_a \right\} + \dots,\end{aligned}$$

where $l_p^{(u,\pm)}$ and $l_{pqr}^{(u,\pm)}$ are likewise the normal-ordered matrix elements of $\hat{L}_u^{(N\pm 1)}$. These left-eigenvectors satisfy the left-eigenvalue problem, with left-eigenvalues $\varepsilon_u^{(\pm)}$, analogous to Eq. (4.16),

$$\langle \Phi_0^{(N)} | [\bar{H}, \bar{L}_u^{(N\pm 1)}] = \pm \varepsilon_u^{(\pm)} \langle \Phi_0^{(N)} | \bar{L}_u^{(N\pm 1)}$$

and form a bi-orthogonal set with the right-eigenvectors, $\langle \bar{L}_u^{(N\pm 1)} | \bar{X}_v^{(N\pm 1)} \rangle = \delta_{uv}$.

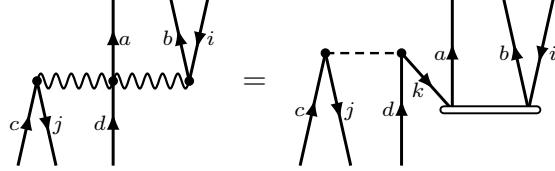
In this paper, because the effective Hamiltonian is real, the corresponding left- and right-eigenvalues are equal. In addition, while the left- and right-eigenvectors are generally not equivalent, the differences in their single-particle Eq. (4.17) or single-hole Eq. (4.18) natures are, in practice, not significant. Therefore, only the right-eigenvectors are used in this paper.

4.2.1 Induced Three-Body Interaction

While the original three-body Hamiltonian was only used for the normal-ordered zero- and one-body pieces, the CC similarity transformation induces higher-body interactions. The two such terms which are relevant to CC-EOM(2) calculations are shown below. The effec-

tive hpphpp three-body interaction can couple two particle-attached operators of the form

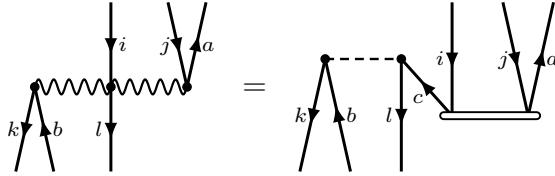
$$\mu r_i^{ab} \left\{ \hat{a}_a^\dagger \hat{a}_b^\dagger \hat{a}_i \right\},$$



$$X_{jcd}^{iab} = - \sum_k V_{cd}^{jk} t_{ki}^{ab}. \quad (4.19)$$

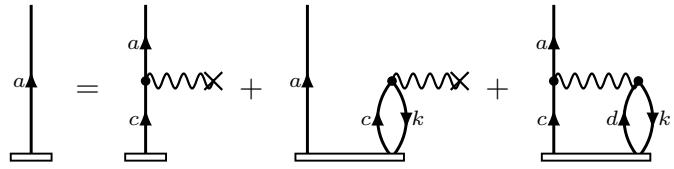
Likewise, the effective hhphhp three-body interaction couples two particle-removed operators

$$\mu r_{ij}^a \left\{ \hat{a}_a^\dagger \hat{a}_j \hat{a}_i \right\},$$

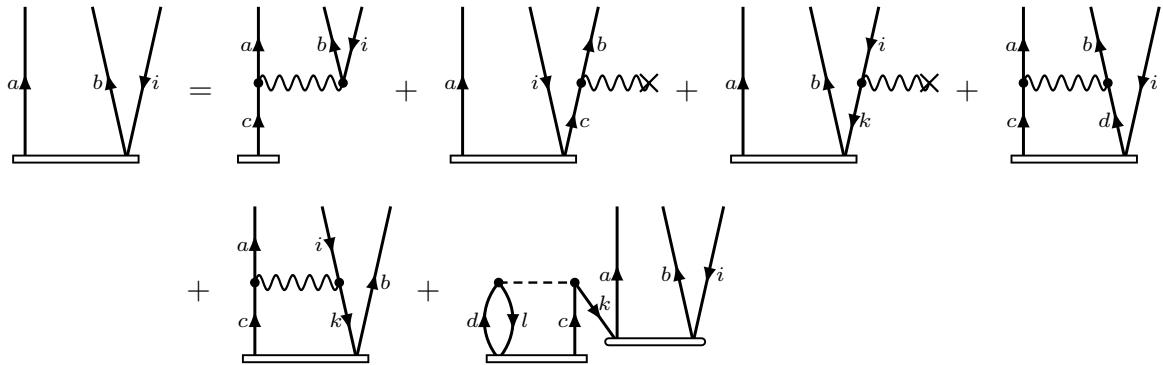


$$X_{ijb}^{kla} = \sum_k V_{cd}^{jk} t_{ki}^{ab}. \quad (4.20)$$

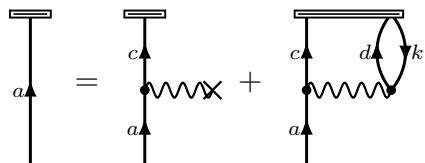
Because the pphpph structure in Eq. (4.19) scales as $\mathcal{O}(n_h^2 n_p^4)$, it can quickly overtake a calculation's memory allocation. Therefore, these structures are never actually built. Instead, the different components are summed as special intermediates when they are needed during PA-EOM-CC or PR-EOM-CC calculations.



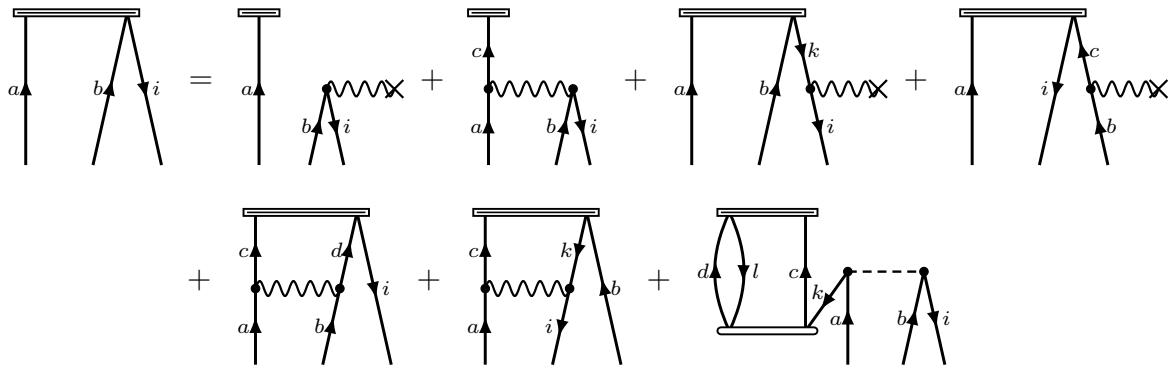
$$\omega_k r^a = \sum_c X_c^a r^c + \sum_{kc} X_c^k r_k^{ac} + \frac{1}{2} \sum_{kcd} X_{cd}^{ak} r_k^{cd} \quad (4.21)$$



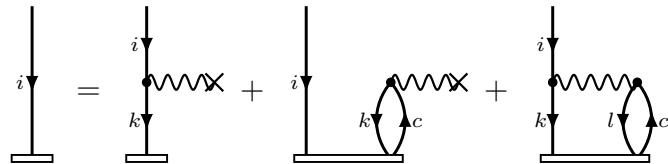
$$\begin{aligned} \omega_k r_i^{ab} = & \sum_c X_{ci}^{ab} r^c + \hat{P}(ab) \sum_c X_c^b r_i^{ac} - \sum_k X_i^k r_k^{ab} + \frac{1}{2} \sum_{cd} X_{cd}^{ab} r_i^{cd} \\ & - \hat{P}(ab) \sum_{kc} X_{ci}^{ak} r_k^{cb} - \frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_{ki}^{ab} r_l^{cd} \end{aligned} \quad (4.22)$$



$$E_k l_a = \sum_c l_c X_a^c + \frac{1}{2} \sum_{kcd} l_{cd}^k X_{ak}^{cd} \quad (4.23)$$



$$\begin{aligned}
E_k l_{ab}^i &= \hat{P}(ab) l_a X_b^i + \sum_c l_c X_{ab}^{ci} - \sum_k l_{ab}^k X_k^i + \hat{P}(ab) \sum_c l_{ac}^i X_b^c \\
&\quad + \frac{1}{2} \sum_{cd} l_{cd}^i X_{ab}^{cd} - \hat{P}(ab) \sum_{kc} l_{cb}^k X_{ak}^{ci} - \frac{1}{2} \sum_{klcd} l_{cd}^l V_{ab}^{ki} t_{kl}^{cd}
\end{aligned} \tag{4.24}$$



$$\omega_k r_i = - \sum_k X_i^k r_k + \sum_{kc} X_c^k r_{ik}^c - \frac{1}{2} \sum_{klc} X_{ic}^{kl} r_{kl}^c \tag{4.25}$$

$$\begin{aligned}
& \text{Diagram 1: } i \downarrow \text{---} \text{V}^{\downarrow a} \text{---} a \uparrow = \\
& \quad \text{Diagram 2: } i \downarrow \text{---} \text{V}^{\downarrow a} \text{---} a \uparrow + \text{Diagram 3: } i \downarrow \text{---} a \uparrow \text{---} \text{V}^{\downarrow j} \text{---} j \uparrow + \text{Diagram 4: } i \downarrow \text{---} j \uparrow \text{---} \text{V}^{\downarrow a} \text{---} a \uparrow + \text{Diagram 5: } i \downarrow \text{---} \text{V}^{\downarrow a} \text{---} c \uparrow \text{---} \text{V}^{\downarrow j} \text{---} j \uparrow + \\
& \quad \text{Diagram 6: } i \downarrow \text{---} \text{V}^{\downarrow a} \text{---} c \uparrow \text{---} \text{V}^{\downarrow j} \text{---} j \uparrow + \text{Diagram 7: } i \downarrow \text{---} \text{V}^{\downarrow a} \text{---} c \uparrow \text{---} \text{V}^{\downarrow l} \text{---} l \uparrow \text{---} a \uparrow \\
& + \text{Diagram 8: } i \downarrow \text{---} \text{V}^{\downarrow a} \text{---} c \uparrow \text{---} \text{V}^{\downarrow l} \text{---} l \uparrow \text{---} a \uparrow + \text{Diagram 9: } i \downarrow \text{---} \text{V}^{\downarrow a} \text{---} c \uparrow \text{---} \text{V}^{\downarrow l} \text{---} l \uparrow \text{---} d \uparrow \text{---} \text{V}^{\downarrow k} \text{---} k \uparrow \text{---} \text{V}^{\downarrow a} \text{---} a \uparrow
\end{aligned}$$

$$\begin{aligned}
\omega_k r_{ij}^a &= - \sum_k X_{ij}^{ka} r_k - \hat{P}(ij) \sum_k X_j^k r_{ik}^a + \sum_c X_c^a r_{ij}^c + \frac{1}{2} \sum_{kl} X_{ij}^{kl} r_{kl}^a \\
&\quad - \hat{P}(ij) \sum_{kc} X_{ci}^{ak} r_{kj}^c - \frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_{ij}^{ca} r_{kl}^d
\end{aligned} \tag{4.26}$$

$$\begin{aligned}
& \text{Diagram 1: } i \downarrow = \text{Diagram 2: } i \downarrow \text{---} \text{V}^{\downarrow i} \text{---} i \downarrow + \text{Diagram 3: } i \downarrow \text{---} \text{V}^{\downarrow i} \text{---} i \downarrow + \text{Diagram 4: } i \downarrow \text{---} \text{V}^{\downarrow i} \text{---} i \downarrow + \text{Diagram 5: } i \downarrow \text{---} \text{V}^{\downarrow i} \text{---} i \downarrow
\end{aligned}$$

$$E_k l^i = - \sum_k l^k X_k^i - \frac{1}{2} \sum_{klc} l_c^{kl} X_{kl}^{ic}
\tag{4.27}$$

The diagram shows a series of Feynman-like diagrams representing a perturbative expansion. The first diagram on the left shows a vertical line with an arrow labeled i entering from below, followed by a diagonal line with an arrow labeled j , which then splits into two lines meeting at a vertex labeled a . This is followed by an equals sign. To the right of the equals sign are several diagrams separated by plus signs. These include:

- A diagram where the j line has a wavy line (representing a field) attached to it.
- A diagram where the i line has a wavy line attached to it.
- A diagram where the k line has a wavy line attached to it.
- A diagram where the i line has a wavy line attached to it, and the j line has a vertex labeled c attached to it.
- A diagram where the i line has a wavy line attached to it, and the a line has a vertex labeled k attached to it.
- A diagram where the k line has a wavy line attached to it, and the a line has a vertex labeled j attached to it.
- A diagram where the i line has a wavy line attached to it, and the j line has a wavy line attached to it.
- A diagram where the i line has a wavy line attached to it, and the a line has a vertex labeled d attached to it.
- A diagram where the k line has a wavy line attached to it, and the a line has a vertex labeled c attached to it.
- A diagram where the i line has a wavy line attached to it, and the a line has a vertex labeled k attached to it.
- A diagram where the k line has a wavy line attached to it, and the a line has a vertex labeled j attached to it.
- A diagram where the i line has a wavy line attached to it, and the j line has a wavy line attached to it.
- A diagram where the i line has a wavy line attached to it, and the a line has a vertex labeled d attached to it.
- A diagram where the k line has a wavy line attached to it, and the a line has a vertex labeled c attached to it.
- A diagram where the i line has a wavy line attached to it, and the a line has a vertex labeled k attached to it.
- A diagram where the k line has a wavy line attached to it, and the a line has a vertex labeled j attached to it.

$$\begin{aligned}
 E_k l_a^{ij} = & \hat{P}(ij) l^i X_a^j - \sum_k l^k X_{ka}^{ij} + \sum_c l_c^{ij} X_a^c - \hat{P}(ij) \sum_k l_a^{ik} X_k^j \\
 & + \frac{1}{2} \sum_{cd} l_a^{kl} X_{kl}^{ij} - \hat{P}(ij) \sum_{kc} l_c^{kj} X_{ak}^{ci} - \frac{1}{2} \sum_{klcd} l_d^{kl} V_{cat}^{ij} t_{kl}^{cd}
 \end{aligned} \tag{4.28}$$

4.3 CC-EOM for 2D Quantum Dots

To examine some properties of the CC-EOM method, it's useful to focus on the results from the two-dimensional quantum dot. This system, also known as an “artificial atom”, consists of a A electrons confined within a 2D harmonic oscillator and subject to the inter-electron Coulomb force. These nanoscale structures are easily tunable which makes them relevant for probing many different quantum phenomena in experiments and theoretical models [?, ?, ?], and because the quantum dot shell structure is similar to those of atomic nuclei, they provide a useful system to test allows experiments to quantify the impact of quantum effects at different levels of correlation [?].

4.3.1 Quantum Dot Formalism

The circular quantum dot system is characterized by a harmonic-oscillator potential of the form $V(r) = m\omega^2 r^2/2$, where ω is its angular frequency, r is the radial distance from the center, and m is the electron mass. The single-particle states can be constructed as

eigenfunctions of the 2D quantum harmonic oscillator,

$$\left(\frac{-\hbar^2}{2m} \nabla^2 + \frac{1}{2} m\omega^2 \hat{r}^2 \right) \phi_{nm\ell\sigma}(\mathbf{r}) = \epsilon_{nm\ell} \phi_{nm\ell\sigma}(\mathbf{r}). \quad (4.29)$$

These states are characterized by a radial quantum number n , a spin quantum number σ , and because this system conserves orbital angular momentum, a corresponding angular momentum projection number, m_ℓ . The energy of each single-particle state $\phi_{nm\ell\sigma}(\mathbf{r})$, independent of the spin quantum projection, is given by,

$$\epsilon_{nm\ell} \equiv (2n + |m_\ell| + 1)\omega. \quad (4.30)$$

In addition to the spin projection σ , they are degenerate with respect to the *shell index* k ,

$$k \equiv 2n + |m_\ell|, \quad (4.31)$$

which labels each shell from zero with a total number of shells, K . The shell structure is illustrated in Fig. 4.1.

The specific form of these basis states can be solved with the cylindrically symmetric Fock–Darwin states $F_{nm\ell}$, which conserve orbital angular momentum $\hat{L}_z \equiv -i\frac{\partial}{\partial\varphi}$. Apart from their spin component, the states are defined as [?],

$$F_{nm\ell}(\rho, \varphi) \equiv \sqrt{\omega} R_{n|m_\ell|}(\sqrt{\omega}\rho) \times \frac{1}{\sqrt{2\pi}} e^{im_\ell\varphi}, \quad \text{where} \quad (4.32)$$

$$R_{nm}(\varrho) \equiv \sqrt{\frac{2 \times n!}{(n+m)!}} e^{-\varrho^2/2} \varrho^m L_n^{(m)}(\varrho^2)$$

and $L_n^{(\alpha)}$ denotes the generalized Laguerre polynomial [?] of degree n and parameter α .

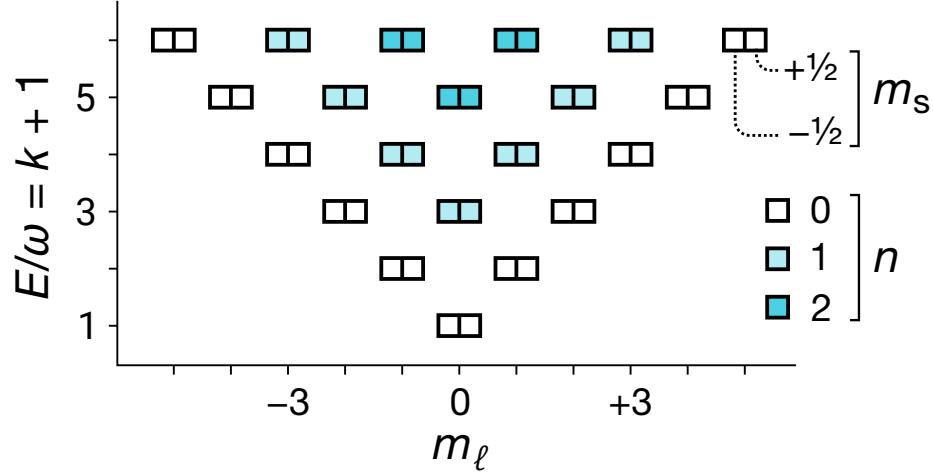


Figure 4.1: The 42 lowest single-particle states (the first 5 shells) in the 2D harmonic oscillator basis. Each box represents a single-particle state arranged by m_ℓ , m_s , and energy, and the up/down arrows indicate the spin of the states. Within each column, the principal quantum number n increases as one traverses upward.

Like the homogeneous electron gas, (see section 3.5), the electrons in the quantum dot interact with each other through the standard Coulomb interaction, $V(\mathbf{r}_1, \mathbf{r}_2) = 1/|\mathbf{r}_1 - \mathbf{r}_2|$, which is expressed in atomic units where $m = c = e = (4\pi\epsilon_0)^{-1} = 1$. With the form of the single-particle wave functions, the second-quantized form of the two-body interaction can be analytically calculated,

$${}^{(2)}H_{rs}^{pq} \equiv \int d\mathbf{r}_1 d\mathbf{r}_2 \phi_p^*(\mathbf{r}_1) \phi_q^*(\mathbf{r}_2) \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} [\phi_r(\mathbf{r}_1) \phi_s(\mathbf{r}_2) - \phi_s(\mathbf{r}_1) \phi_r(\mathbf{r}_2)], \quad (4.33)$$

where $\phi_p(\mathbf{r})$ is shorthand for $\phi_{n_p m_{\ell p} \sigma_p}(\mathbf{r})$.

With these ingredients in hand, the ground state energy, as well as particle-attached and particle-removed energies, can be calculated for different values of the oscillator frequency, ω , and the number of shells, K .

4.3.2 Quantum Dot Results

Here, CC results are shown along with results from the in-medium similarity renormalization group (IM-SRG) method [?] and quasidegenerate perturbation theory (QDPT) [?, ?]. As for all calculations in this work, each method applies a Hartree-Fock transformation of the basis states from Eq. (4.29) before employing the primary many-body method. Before analyzing the EOM results, the ground state energies calculated using HF + IM-SRG(2) and HF + CCSD are show in Fig. 4.11. Also included are results from Møller–Plesset perturbation theory to second order (MP2), DMC [?], and FCI [?] for comparison where available.

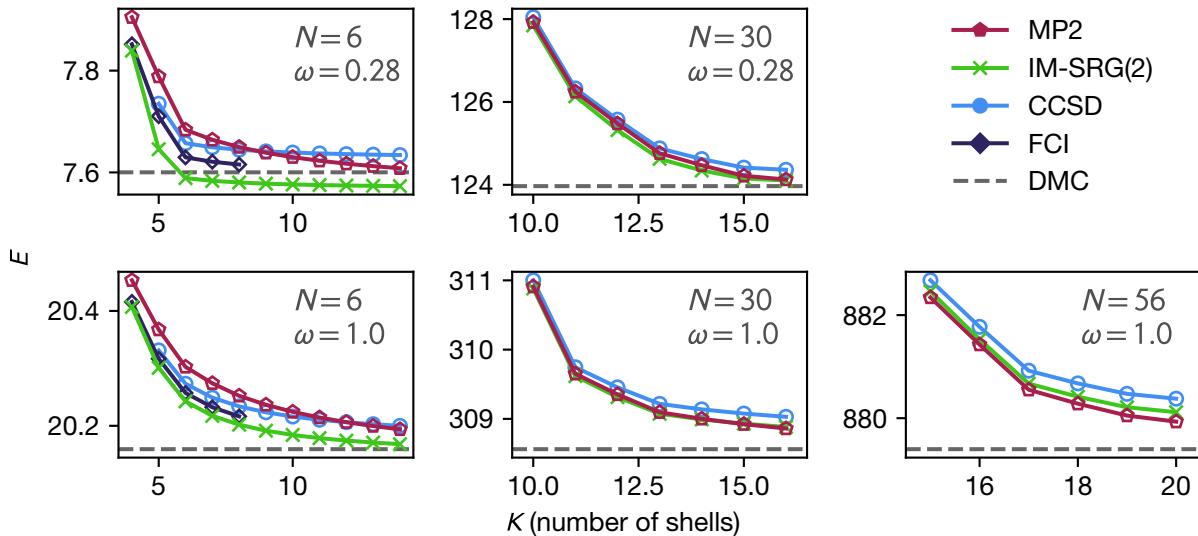


Figure 4.2: Ground state energy (in Hartrees) of quantum dots with N particles and an oscillator frequency of ω calculated with several different methods.

With respect to the number of shells, both IM-SRG(2) and CCSD appear to converge slightly faster than second order perturbation theory (MP2), mainly due to the presence of higher order corrections in IM-SRG(2) and CCSD. However, the main conclusion that can be reached from Fig. 4.11 is the overall convergence between vastly different methods, a hallmark of properly-treated *ab initio* methods.

The results for addition and removal energy calculations are summarized in Fig. 4.3 and

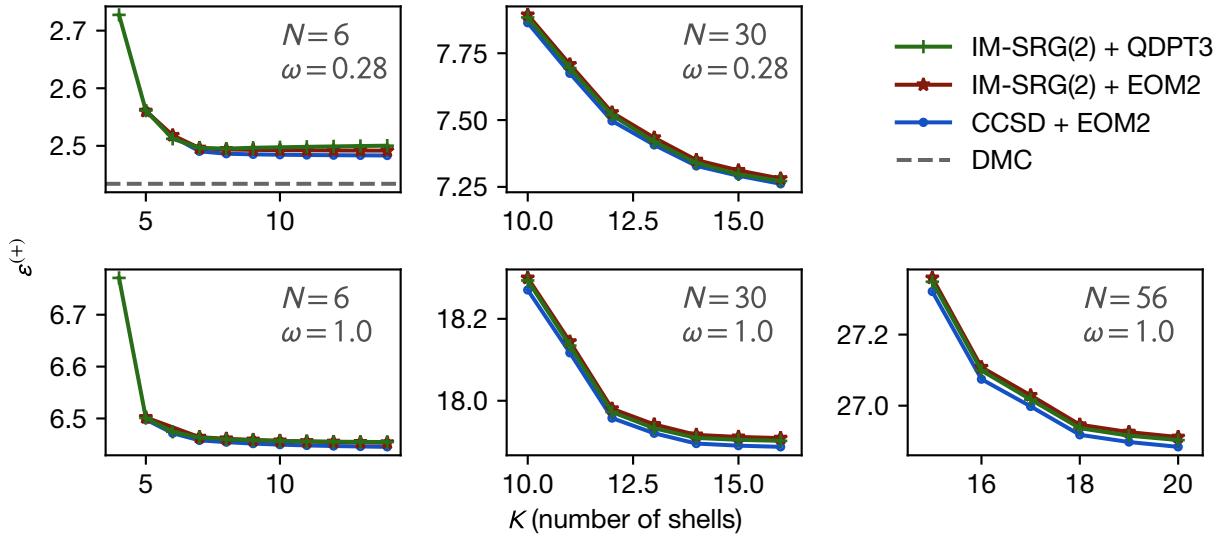


Figure 4.3: Particle-attached energy (in Hartrees) of quantum dots with N particles and an oscillator frequency of ω calculated with several different methods.

Fig. 4.4 respectively.

Not only do these results reaffirm the validity of the CCSD method amongst other *ab initio* methods, they also provide benchmark calculations for the following CC-EOM calculations of open-shell finite nuclei.

4.4 CC-EOM for 2D Quantum Dots

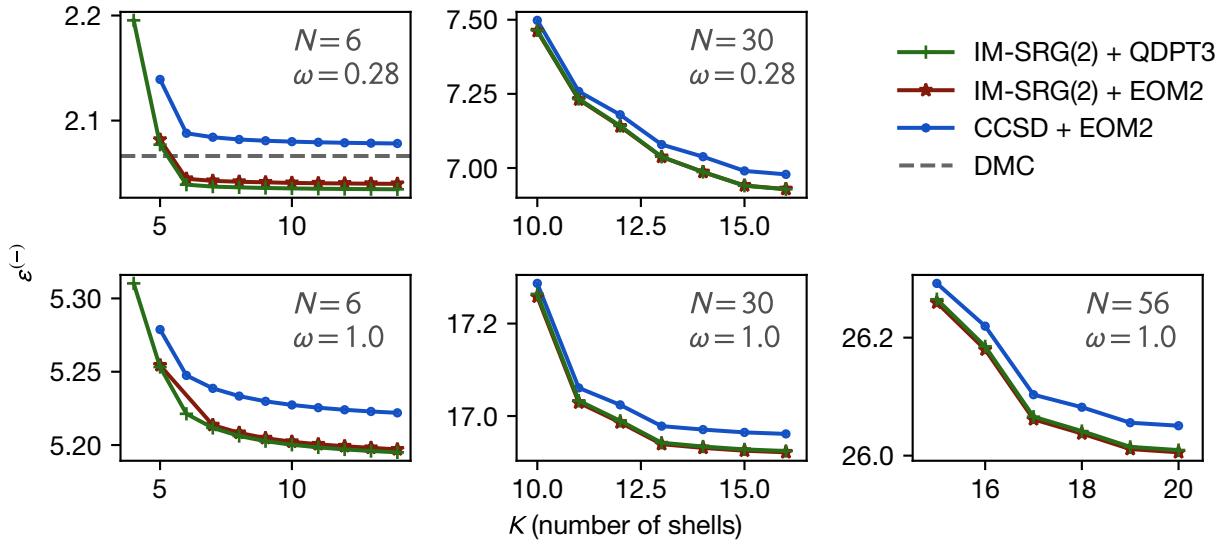


Figure 4.4: Particle-removed energy (in Hartrees) of quantum dots with N particles and an oscillator frequency of ω calculated with several different methods.

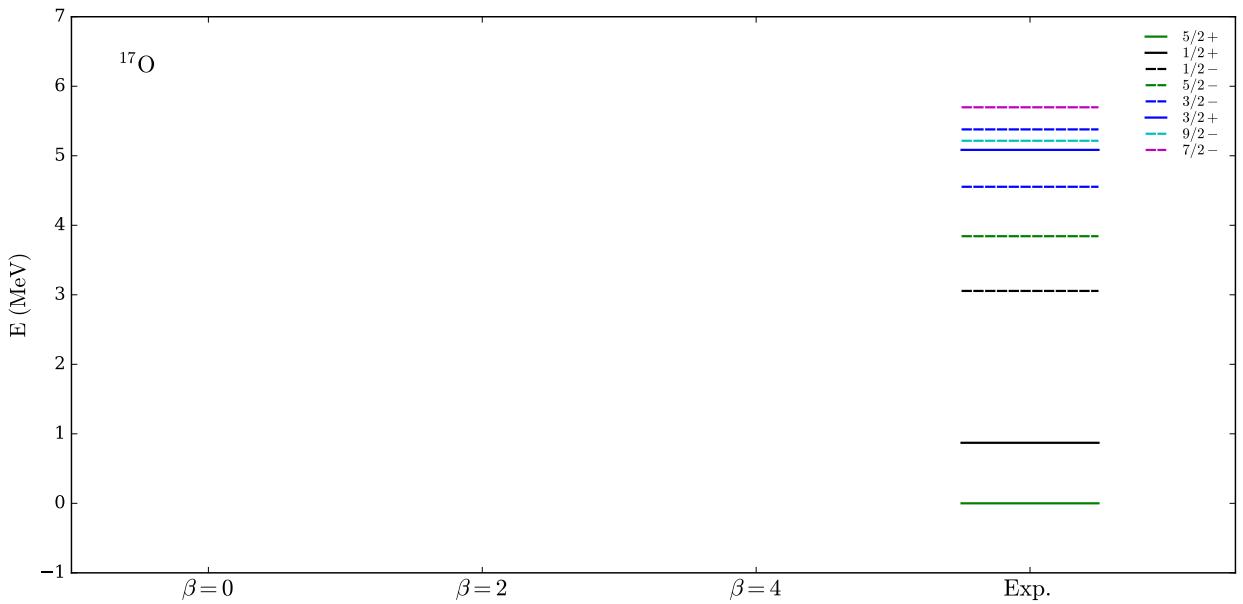


Figure 4.5: CCD energy per electron in Hartrees for the 3D homogeneous electron gas as function of the Wigner-Seitz radius in units of Bohr radii. The calculation used periodic boundary conditions and a basis with 25 shells, resulting in a total of 1238 single-particle states. Also plotted are the variational quantum Monte Carlo (QMC) results from [82].

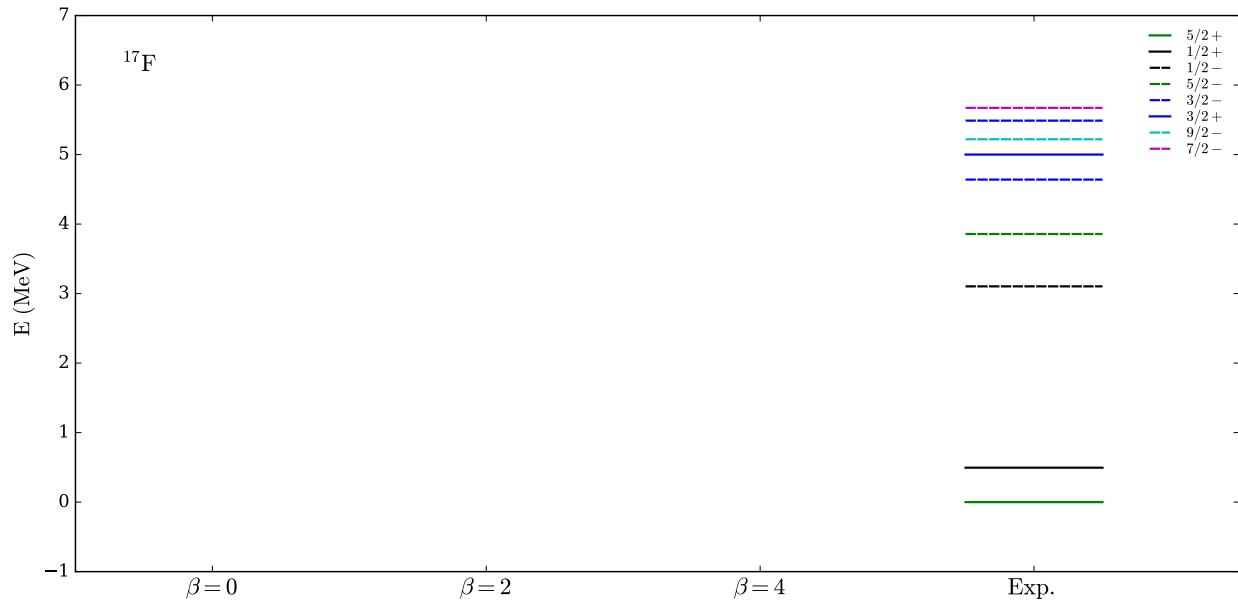


Figure 4.6: CCD energy per electron in Hartrees for the 3D homogeneous electron gas as function of the Wigner-Seitz radius in units of Bohr radii. The calculation used periodic boundary conditions and a basis with 25 shells, resulting in a total of 1238 single-particle states. Also plotted are the variational quantum Monte Carlo (QMC) results from [82].

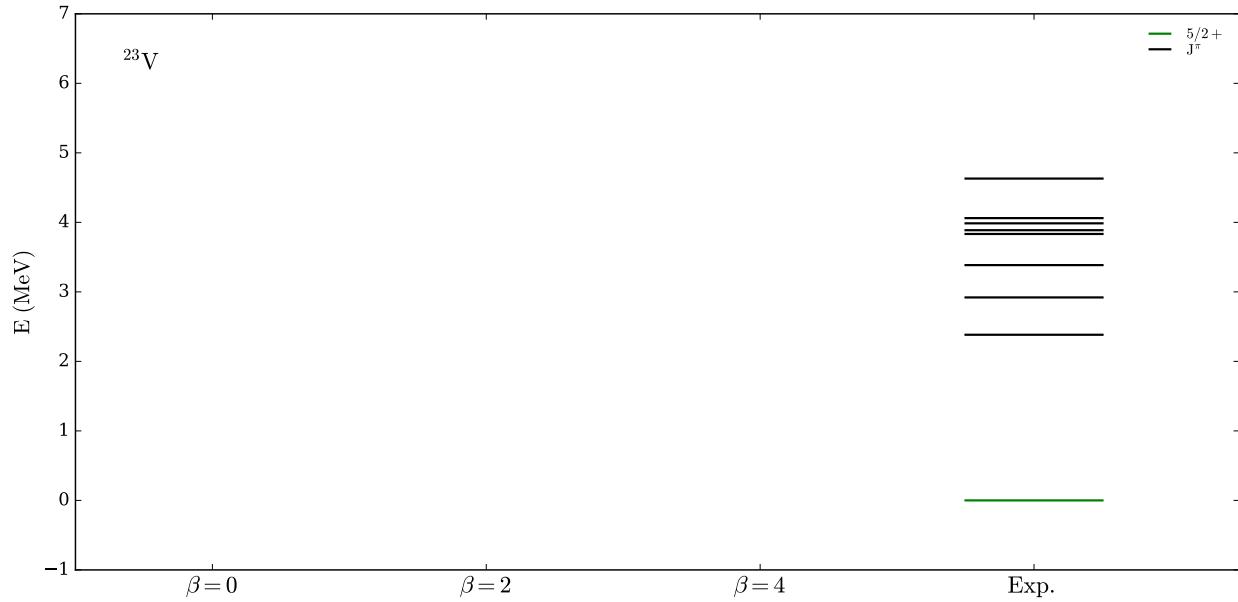


Figure 4.7: CCD energy per electron in Hartrees for the 3D homogeneous electron gas as function of the Wigner-Seitz radius in units of Bohr radii. The calculation used periodic boundary conditions and a basis with 25 shells, resulting in a total of 1238 single-particle states. Also plotted are the variational quantum Monte Carlo (QMC) results from [82].

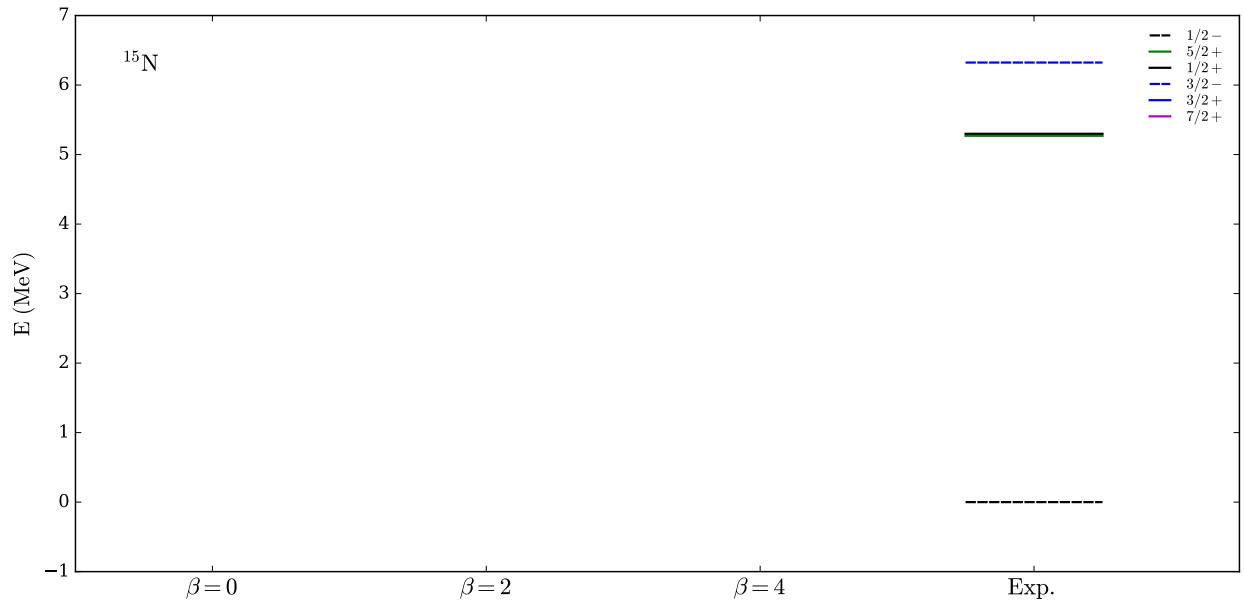


Figure 4.8: CCD energy per electron in Hartrees for the 3D homogeneous electron gas as function of the Wigner-Seitz radius in units of Bohr radii. The calculation used periodic boundary conditions and a basis with 25 shells, resulting in a total of 1238 single-particle states. Also plotted are the variational quantum Monte Carlo (QMC) results from [82].

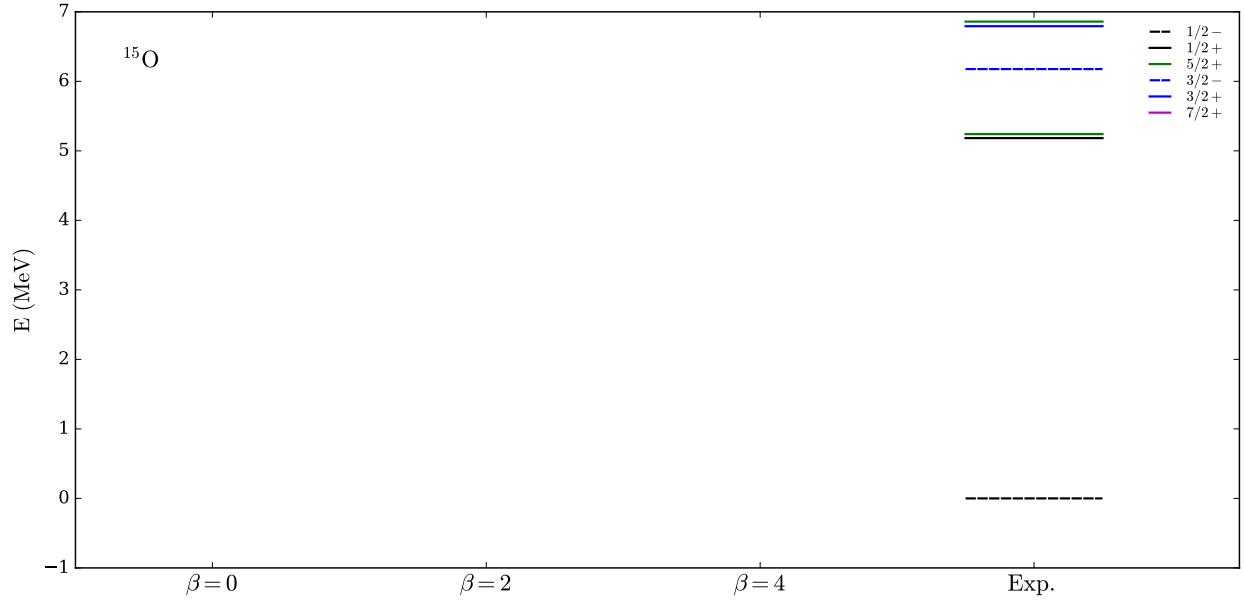


Figure 4.9: CCD energy per electron in Hartrees for the 3D homogeneous electron gas as function of the Wigner-Seitz radius in units of Bohr radii. The calculation used periodic boundary conditions and a basis with 25 shells, resulting in a total of 1238 single-particle states. Also plotted are the variational quantum Monte Carlo (QMC) results from [82].

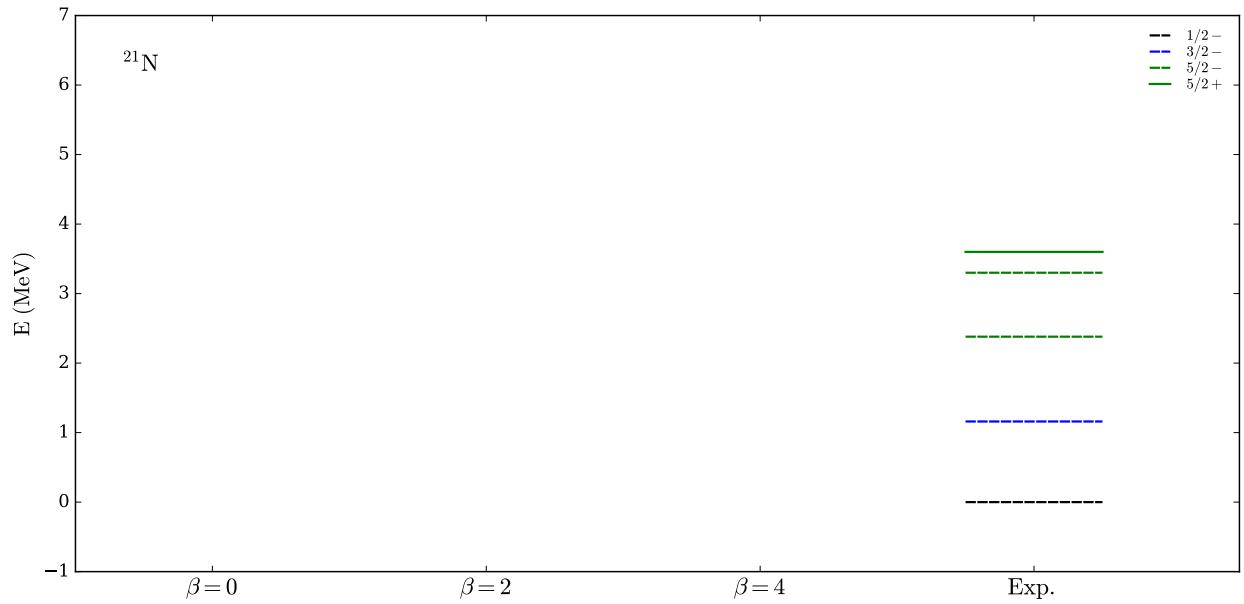


Figure 4.10: CCD energy per electron in Hartrees for the 3D homogeneous electron gas as function of the Wigner-Seitz radius in units of Bohr radii. The calculation used periodic boundary conditions and a basis with 25 shells, resulting in a total of 1238 single-particle states. Also plotted are the variational quantum Monte Carlo (QMC) results from [82].

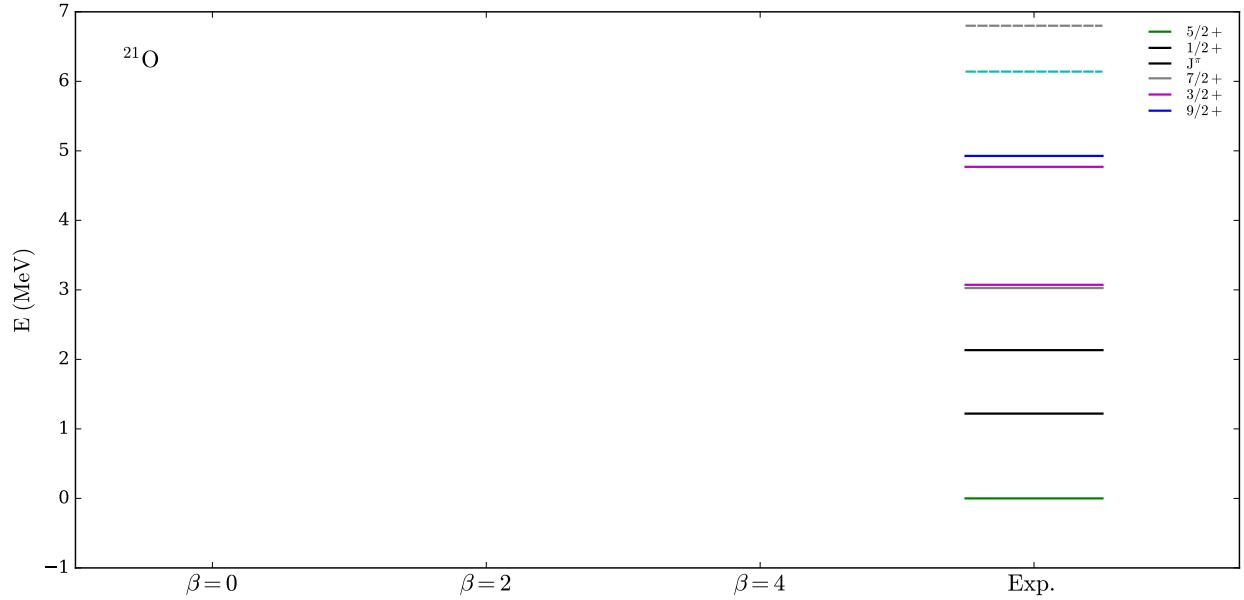


Figure 4.11: CCD energy per electron in Hartrees for the 3D homogeneous electron gas as function of the Wigner-Seitz radius in units of Bohr radii. The calculation used periodic boundary conditions and a basis with 25 shells, resulting in a total of 1238 single-particle states. Also plotted are the variational quantum Monte Carlo (QMC) results from [82].

Chapter 5

Effective Operators

$$\begin{array}{c} \text{Diagram: Two vertical lines with arrows pointing down, labeled } i \text{ and } a, \text{ connected by a dashed horizontal line with a diamond symbol.} \\ = \\ \text{Diagram: Two vertical lines with arrows pointing down, labeled } i \text{ and } a, \text{ connected by a solid horizontal line with a diamond symbol.} \end{array}$$

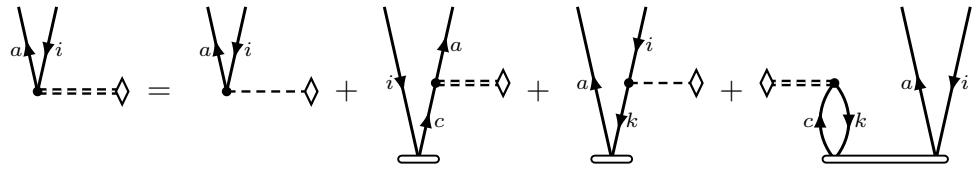
$$\lambda \bar{O}_a^i = \lambda O_a^i \quad (5.1)$$

$$\begin{array}{c} \text{Diagram: Two vertical lines with arrows pointing up, labeled } a \text{ and } b, \text{ connected by a dashed horizontal line with a diamond symbol.} \\ = \\ \text{Diagram: Two vertical lines with arrows pointing up, labeled } a \text{ and } b, \text{ connected by a solid horizontal line with a diamond symbol.} \\ + \\ \text{Diagram: A vertical line with arrow } a \text{ and a diagonal line with arrow } k \text{ meeting at a point connected to a horizontal line with arrow } b, \text{ all connected by a dashed horizontal line with a diamond symbol.} \end{array}$$

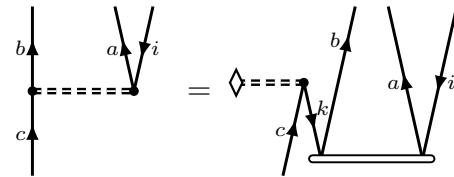
$$\lambda \bar{O}_b^a = \lambda O_b^a - \sum_k \lambda \bar{O}_b^k t_k^a \quad (5.2)$$

$$\begin{array}{c} \text{Diagram: Two vertical lines with arrows pointing down, labeled } j \text{ and } i, \text{ connected by a dashed horizontal line with a diamond symbol.} \\ = \\ \text{Diagram: Two vertical lines with arrows pointing down, labeled } j \text{ and } i, \text{ connected by a solid horizontal line with a diamond symbol.} \\ + \\ \text{Diagram: A vertical line with arrow } j \text{ and a diagonal line with arrow } c \text{ meeting at a point connected to a horizontal line with arrow } i, \text{ all connected by a dashed horizontal line with a diamond symbol.} \end{array}$$

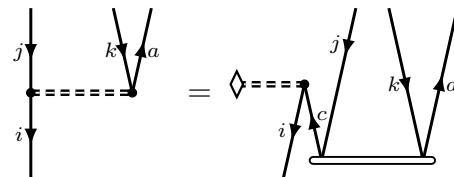
$$\lambda \bar{O}_j^i = \lambda O_j^i + \sum_c \lambda \bar{O}_c^i t_j^c \quad (5.3)$$



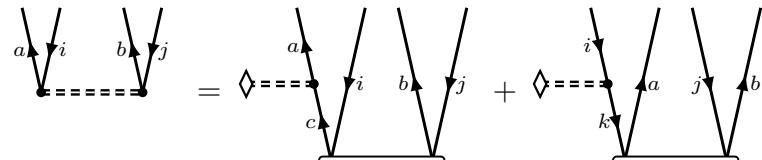
$$\lambda \bar{O}_i^a = \lambda O_i^a + \sum_c \lambda \bar{O}_c^a t_i^c - \sum_k \lambda \bar{O}_i^k t_k^a + \sum_{kc} \lambda \bar{O}_c^k t_{ik}^{ac} \quad (5.4)$$



$$\lambda \bar{O}_{ic}^{ab} = - \sum_k \lambda \bar{O}_c^k t_{ik}^{ab} \quad (5.5)$$



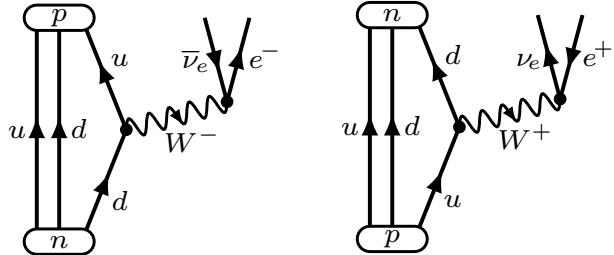
$$\lambda \bar{O}_{jk}^{ia} = \sum_c \lambda \bar{O}_c^i t_{jk}^{ca} \quad (5.6)$$



$$\lambda \bar{O}_{ij}^{ab} = \hat{P}(ab) \sum_c \lambda \bar{O}_c^a t_{ij}^{cb} - \hat{P}(ij) \sum_k \lambda \bar{O}_i^k t_{kj}^{ab} \quad (5.7)$$

Decay Type	$\Delta J = J_F - J_I$	$\pi_F \pi_I$
Fermi	0	+1
Gamow-Teller	1 ($J_F = 0$ or $J_I = 0$)	+1
Gamow-Teller	0,1 ($J_F > 0$ and $J_I > 0$)	+1

Table 5.1: Single-particle states and their quantum numbers and their energies from Eq. (??). The degeneracy for every quantum number p is equal to 2 due to the two possible spin values.

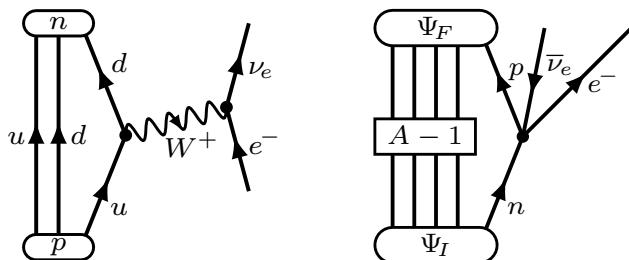


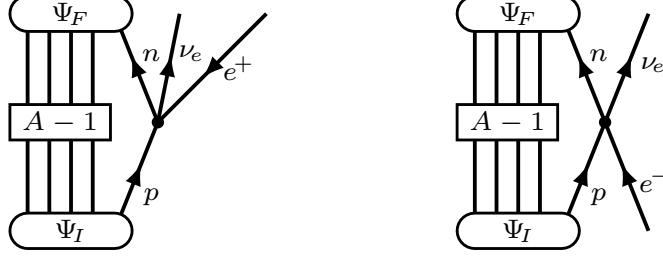
5.1 Beta Decay

$$\beta^- \text{ decay :} \quad n \longrightarrow p + e^- + \bar{\nu}_e \quad (5.8)$$

$$\beta^+ \text{ decay :} \quad p \longrightarrow n + e^+ + \nu_e \quad (5.9)$$

$$\text{Electron capture :} \quad p + e^- \longrightarrow n + \nu_e \quad (5.10)$$





5.2 Sum Rules

$$\begin{aligned}
\sum_F [B_{F-}^{I,F} - B_{F-}^{I,F}] &= \sum_F \left[|\langle F | \sum_i \tau_i^- | I \rangle|^2 - |\langle F | \sum_i \tau_i^+ | I \rangle|^2 \right] \\
&= \sum_F \left[\langle I | \sum_i \tau_i^+ | F \rangle \langle F | \sum_i \tau_i^- | I \rangle - \langle I | \sum_i \tau_i^- | F \rangle \langle F | \sum_i \tau_i^+ | I \rangle \right] \\
&= \langle I | (\sum_i \tau_i^+) (\sum_j \tau_j^-) - (\sum_i \tau_i^-) (\sum_j \tau_j^+) | I \rangle = \langle I | 2 \sum_i \tau_i^z | I \rangle = (N_I - Z_I) \quad (5.11)
\end{aligned}$$

$$\begin{aligned}
\sum_F [B_{GT-}^{I,F} - B_{GT-}^{I,F}] &= \sum_{F,\mu} \left[|\langle F | \sum_i \sigma_i^\mu \tau_i^- | I \rangle|^2 - |\langle F | \sum_i \sigma_i^\mu \tau_i^+ | I \rangle|^2 \right] \\
&= \sum_{F,\mu} \left[\langle I | \sum_i \sigma_i^\mu \tau_i^+ | F \rangle \langle F | \sum_i \sigma_i^\mu \tau_i^- | I \rangle - \langle I | \sum_i \sigma_i^\mu \tau_i^- | F \rangle \langle F | \sum_i \sigma_i^\mu \tau_i^+ | I \rangle \right] \\
&= \sum_\mu \left[\langle I | (\sum_i \sigma_i^\mu \tau_i^+) (\sum_j \sigma_i^\mu \tau_j^-) - (\sum_i \sigma_i^\mu \tau_i^-) (\sum_j \sigma_i^\mu \tau_j^+) | I \rangle \right] \\
&= \sum_\mu \left[\langle I | \sum_i \sigma_{i,\mu}^2 (\tau_{i,+} \tau_{i,-} - \tau_{i,-} \tau_{i,+}) | I \rangle \right] = 3 \langle I | \sum_i (\tau_{i,+} \tau_{i,-} - \tau_{i,-} \tau_{i,+}) | I \rangle \\
&= 3 \langle I | 2 \sum_i \tau_i^z | I \rangle = 3 (N_I - Z_I) \quad (5.12)
\end{aligned}$$

$$\sum_F \left[B_{F_-}^{I,F} - B_{F_-}^{I,F} \right] = (N_I - Z_I) \quad (5.13)$$

$$\sum_F \left[B_{GT_-}^{I,F} - B_{GT_-}^{I,F} \right] = 3(N_I - Z_I) \quad (5.14)$$

5.3 Calculating Beta-Decay Matrix Elements

Fix these!!!!!!

$$\mathcal{M}_F \equiv \langle \mathcal{F} \| \tau \| \mathcal{I} \rangle = \delta_{J_{\mathcal{F}} J_{\mathcal{I}}} \sum_{pq} \langle p \| \hat{\tau} \| q \rangle \langle \mathcal{F} \| \hat{a}_p^\dagger \hat{a}_q \| \mathcal{I} \rangle \quad (5.15)$$

$$\mathcal{M}_{GT} \equiv \langle \mathcal{F} \| \sigma \tau \| \mathcal{I} \rangle = \sum_{pq} \langle p \| \hat{\sigma} \hat{\tau} \| q \rangle \langle \mathcal{F} \| \hat{a}_p^\dagger \hat{a}_q \| \mathcal{I} \rangle \quad (5.16)$$

$$\langle p \| \hat{\tau} \| q \rangle = \delta_{n_p n_q} \delta_{l_p l_q} \delta_{j_p j_q} \delta_{t_p t_q \pm 1} \hat{j}_p \quad (5.17)$$

$$\langle p \| \hat{\sigma} \hat{\tau} \| q \rangle = \sqrt{6} \delta_{n_p n_q} \delta_{l_p l_q} \delta_{t_p t_q \pm 1} \hat{j}_p \hat{j}_q (-1)^{l_p + j_p + \frac{3}{2}} \begin{Bmatrix} \frac{1}{2} & \frac{1}{2} & 1 \\ j_q & j_p & l_p \end{Bmatrix} \quad (5.18)$$

$$B_F \equiv \frac{g_V^2}{\hat{J}_{\mathcal{I}}^2} |\mathcal{M}_F|^2 \quad (5.19)$$

$$B_F = \frac{g_V^2}{\hat{J}_{\mathcal{I}}^2} \sum_{\mathcal{F}\mathcal{I}} \frac{\langle \hat{L}^{\mathcal{I}} | \tau | \hat{R}^{\mathcal{F}} \rangle \langle \hat{L}^{\mathcal{F}} | \tau | \hat{R}^{\mathcal{I}} \rangle}{\langle J_{\mathcal{I}} M_{\mathcal{I}}; \lambda - \mu | J_{\mathcal{F}} M_{\mathcal{F}} \rangle^2} \quad (5.20)$$

$$B_{GT} \equiv \frac{g_A^2}{\hat{J}_{\mathcal{I}}^2} |\mathcal{M}_{GT}|^2 \quad (5.21)$$

$$B_{GT} = \frac{g_A^2}{\hat{J}_{\mathcal{I}}^2} \sum_{\mathcal{F}\mathcal{I}} \frac{\langle \hat{L}^{\mathcal{I}} | \sigma \tau | \hat{R}^{\mathcal{F}} \rangle \langle \hat{L}^{\mathcal{F}} | \sigma \tau | \hat{R}^{\mathcal{I}} \rangle}{\langle J_{\mathcal{I}} M_{\mathcal{I}}; \lambda - \mu | J_{\mathcal{F}} M_{\mathcal{F}} \rangle^2} \quad (5.22)$$

$$B_{\beta-} = \langle \Phi_0 | \hat{L}^n (\hat{\beta}^-)^\dagger \hat{R}^p | \Phi_0 \rangle \langle \Phi_0 | \hat{L}^p \hat{\beta}^- \hat{R}^n | \Phi_0 \rangle \quad (5.23)$$

$$B_{\beta-} = \langle \Phi_0 | \hat{L}_p (\hat{\beta}^-)^\dagger \hat{R}_n | \Phi_0 \rangle \langle \Phi_0 | \hat{L}_n \hat{\beta}^- \hat{R}_p | \Phi_0 \rangle \quad (5.24)$$

$$B_{\beta-} = \langle \Phi_0 | \hat{L}_p^n (\hat{\beta}^-)^\dagger \hat{R}_0 | \Phi_0 \rangle \langle \Phi_0 | \hat{L}_0 \hat{\beta}^- \hat{R}_p^n | \Phi_0 \rangle \quad (5.25)$$

$$B_{\beta-} = \langle \Phi_0 | \hat{L}_0 (\hat{\beta}^-)^\dagger \hat{R}_n^p | \Phi_0 \rangle \langle \Phi_0 | \hat{L}_n^p \hat{\beta}^- \hat{R}_0 | \Phi_0 \rangle \quad (5.26)$$

$$B_{\beta+} = \langle \Phi_0 | \hat{L}^p (\hat{\beta}^+)^\dagger \hat{R}^n | \Phi_0 \rangle \langle \Phi_0 | \hat{L}^n \hat{\beta}^+ \hat{R}^p | \Phi_0 \rangle \quad (5.27)$$

$$B_{\beta+} = \langle \Phi_0 | \hat{L}_n (\hat{\beta}^+)^\dagger \hat{R}_p | \Phi_0 \rangle \langle \Phi_0 | \hat{L}_p \hat{\beta}^+ \hat{R}_n | \Phi_0 \rangle \quad (5.28)$$

$$B_{\beta+} = \langle \Phi_0 | \hat{L}_n^p (\hat{\beta}^+)^\dagger \hat{R}_0 | \Phi_0 \rangle \langle \Phi_0 | \hat{L}_0 \hat{\beta}^+ \hat{R}_n^p | \Phi_0 \rangle \quad (5.29)$$

$$B_{\beta+} = \langle \Phi_0 | \hat{L}_0 (\hat{\beta}^+)^\dagger \hat{R}_p^n | \Phi_0 \rangle \langle \Phi_0 | \hat{L}_p^n \hat{\beta}^+ \hat{R}_0 | \Phi_0 \rangle \quad (5.30)$$

Quenching [57, 123, 75, 94]

Chapter 6

Conclusions and Perspectives

Chapter 7

Instructions

If you want to use this class, it's probably a good idea to use the source code of this example document is as a starting point.

7.1 Preamble

The document class may be declared using `\documentclass[<type>]{msudissertation}`, where `<type>` is either `dissertation` (default) or `thesis`. The class is based on the `book` class and thus inherits all its structural conventions. The WikiBooks has more information about this: https://en.wikibooks.org/wiki/LaTeX/Document_Structure.

Afterward, you can load your packages. Among those, you may find the following packages useful:

- `\usepackage{hyperref}`: provides hyperlinks (`\url`) and PDF bookmarks
- `\usepackage{pdflscape}`: provides `\begin{landscape} ... \end{landscape}`
- `\usepackage{titling}`: provides `\thetitle`, `\theauthor`, and `\thedate`

Refer to their official documentation on CTAN for more details.

To set the title, author, degree program, and date, include the following commands in your preamble:

- `\title{<title>}`

- `\author{<name>}`
- `\def\thegreeprogram{<subject>---<degree>}`
- `\date{<year>}` (optional) Per university guidelines, the date must be contain only a 4-digit year. If omitted, it defaults to the current year, which could be undesirable if you want to reproduce the document years later.

The rest of the document is divided into three major parts, preceded the special markers `\frontmatter`, `\mainmatter`, and `\appendix` respectively.

7.2 Front matter

In the front matter, certain chapter names have been endowed with special meanings, e.g. `\chapter{Abstract}` or `\chapter{Copyright}`. This is what allows them to have unique formatting. They must be spelled and capitalized exactly as written in the source code of this example, unless otherwise specified. Given that the `\chapter` command has been imbued with some rather fragile (read: hacky) logic, try not to sneeze on them too hard.

The front matter in this example is a lot more packed than your typical dissertation or thesis, because a lot of the chapters are optional and have been filled with placeholder text. They can be safely deleted if neither you nor the university guidelines require them.

7.3 Main matter

The main matter is the most uninteresting part of this template, because it's almost the same as your vanilla `book` class. Just write `\chapter{<chapter>}` and `\section{<section>}` like you normally do.

Chapter 8

Appendix

You can have as many appendices as you want, or none at all. If you do have *at least one*, use the `\appendix` macro to create a cover page with the correct grammatical number depending on how many you have, and adjusts the table of contents according to university guidelines. After this macro, all uses of `\chapter{<chapter>}` will create an appendix chapter instead of a regular chapter. Do *not* use `\appendix` if you have no appendices at all.

At the very end, there is the mandatory bibliography. Here, I'm assuming you want to use the traditional `natbib`. If so, start by selecting a `\bibliographystyle{<style>}`. If you don't like calling it "Bibliography", you can pick a more suitable title using the syntax `\renewcommand{\bibname}{<title>}` as long as it conforms to university guidelines. Afterward, you can write `\bibliography{<name>}` where `<name>` is the path to the `.bib` database without the file extension.

And that's it! The remaining part of this document is full of placeholder text so you can stop reading now.

Chapter 9

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 Pellentesque ac consequat leo. Nulla vel aliquet ex. Nulla non faucibus sapien, eu porttitor ligula:

$$I = \int_{-\infty}^{\infty} \frac{x}{1 + e^{-x^2/2}} dx \quad (9.1)$$

 Class aptent taciti sociosqu ad litora torquent per conubia nostra, per inceptos himenaeos. In odio metus, maximus sed dictum vel, rhoncus id ante. Phasellus viverra sodales neque ut pulvinar.

 Sed neque metus, elementum at risus in, imperdiet iaculis dolor. Vivamus quis libero quis dui finibus imperdiet. Nunc mollis odio eget nibh volutpat interdum. Praesent non felis in sem luctus gravida.

9.1 Pellentesque Scelerisque

 Pellentesque scelerisque justo in pellentesque iaculis. Nunc in elementum tortor. Vestibulum tincidunt lacus ac libero gravida, sit amet rhoncus risus iaculis. Sed rhoncus nisl ac felis



Figure 9.1: Lorem ipsum dolor sit amet, consectetur adipiscing elit. Ut et leo non tortor viverra sodales.

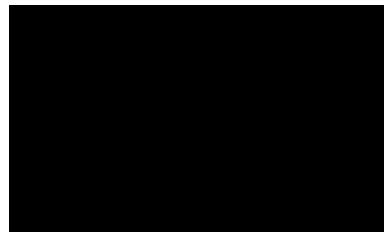


Figure 9.2: Ut condimentum odio orci, a varius sapien vehicula quis.

auctor efficitur. Nulla vel lorem bibendum, laoreet risus vel, euismod odio. Sed a justo augue. Sed sed viverra magna. Etiam vestibulum tellus ut consequat ultricies. Pellentesque mattis odio ac ipsum luctus, ac laoreet ex efficitur. Sed tincidunt, est id viverra ullamcorper, orci dolor accumsan erat, id accumsan tortor est quis ante. Integer pretium elit a ligula tristique imperdiet.

In vitae elit massa. Morbi fermentum arcu quis tristique pretium. Suspendisse potenti. Suspendisse at nisi nec sem dignissim mattis finibus ac sem. Aenean lobortis volutpat turpis, a lacinia ante pellentesque eget. Vivamus fermentum tempus viverra. Sed sollicitudin cursus rutrum. [35]

9.1.1 Fusce Convallis

Fusce convallis placerat porta. Nam porta euismod justo, nec efficitur erat tincidunt non. Proin viverra sagittis nibh, ac rhoncus nibh condimentum in. Nunc tristique augue quis ante

dapibus, sit amet porta purus molestie. Pellentesque sagittis tincidunt eros, eget fermentum nibh viverra dapibus. Phasellus id dignissim sapien. Mauris eget interdum sapien, in hendrerit ante.

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9.2 Curabitur

Curabitur dignissim orci quis orci blandit blandit.¹ Cras cursus tellus quam, sed lacinia nisi scelerisque posuere. Vestibulum sed lacinia orci, id sagittis lorem. Etiam lacinia elementum tortor, ut rutrum tellus laoreet sit amet. Etiam dignissim sagittis sapien in dignissim. Vestibulum ante ipsum primis in faucibus orci luctus et ultrices posuere cubilia Curae; Nullam blandit est vel diam hendrerit posuere. Maecenas nec arcu eu dui semper porttitor.

¹Aenean a semper dolor.

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O	X	
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Table 9.1: Nulla suscipit ultricies massa at sagittis.

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APPENDICES

$$E\left(\hat{T}, \hat{L}\right) = \langle \Phi_0 | \hat{L} e^{-\hat{T}} \hat{H} e^{\hat{T}} | \Phi_0 \rangle = \langle \Phi_0 | \hat{L} \bar{H} | \Phi_0 \rangle \quad (2)$$

$$\hat{L} \equiv \lambda_0 + \hat{L}_1 + \hat{L}_2 + \cdots + \hat{L}_N \quad (3)$$

$$\hat{L}_k \equiv \left(\frac{1}{k!} \right)^2 \sum_{\substack{a_1 \dots a_k \\ i_1 \dots i_k}} \lambda_{a_1 \dots a_k}^{i_1 \dots i_k} \left\{ \hat{i}_1^\dagger \dots \hat{i}_k^\dagger \hat{a}_k \dots \hat{a}_1 \right\}, \quad (4)$$

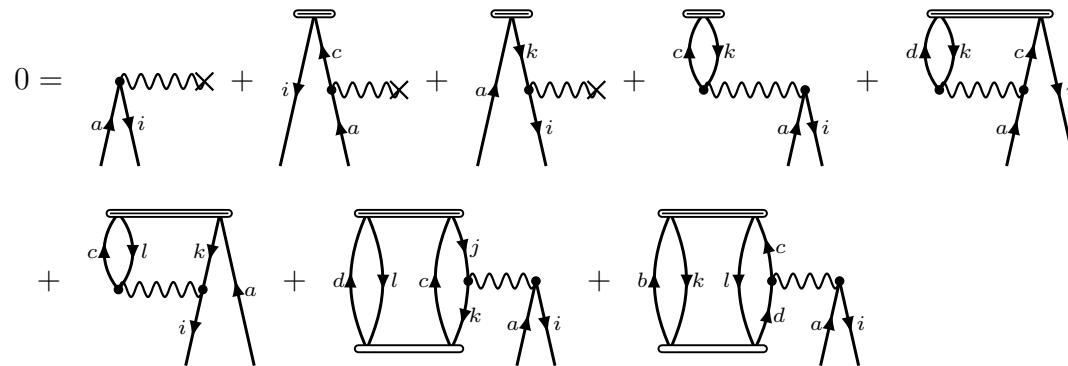
$$\begin{aligned} \langle \Phi_0 | \hat{L} \left[\bar{H}, \left\{ \hat{a}^\dagger \hat{i} \right\} \right] | \Phi_0 \rangle &= 0, \\ \langle \Phi_0 | \hat{L} \left[\bar{H}, \left\{ \hat{a}^\dagger \hat{b}^\dagger \hat{j} \hat{i} \right\} \right] | \Phi_0 \rangle &= 0. \end{aligned} \quad (5)$$

$$\langle \Phi_0 | \hat{L} \bar{H} | \Phi_i^a \rangle = \omega \langle \Phi_0 | \hat{L} | \Phi_i^a \rangle, \quad (6)$$

$$\langle \Phi_0 | \hat{L} \bar{H} | \Phi_{ij}^{ab} \rangle = \omega \langle \Phi_0 | \hat{L} | \Phi_{ij}^{ab} \rangle.$$

$$\begin{aligned} \Delta E_{CCSD} &= \text{Diagram 1} + \text{Diagram 2} + \text{Diagram 3} \\ &= \frac{1}{4} \sum_{klcd} V_{cd}^{kl} t_{kl}^{cd} + \frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_k^c t_l^d + \sum_{\substack{k \\ c}} \bar{f}_c^k \bar{t}_{k'}^c \end{aligned} \quad (7)$$

$$\begin{aligned}
0 = & \quad \text{Diagram 1} + \text{Diagram 2} + \text{Diagram 3} + \text{Diagram 4} + \text{Diagram 5} \\
& + \text{Diagram 6} + \text{Diagram 7} + \text{Diagram 8} \\
= & X_a^i + \sum_c \lambda_c^i X_a^c - \sum_k \lambda_a^k X_k^i + \sum_{kc} \lambda_c^k X_{ak}^{ic} + \frac{1}{2} \sum_{kcd} \lambda_{cd}^{ik} X_{ak}^{cd} \\
& - \frac{1}{2} \sum_{klc} \lambda_{ac}^{kl} X_{kl}^{ic} - \frac{1}{2} \sum_{jklcd} \lambda_{cd}^{jl} X_{aj}^{ik} t_{kl}^{cd} + \frac{1}{2} \sum_{klbcd} \lambda_{bc}^{kl} X_{ad}^{ic} t_{kl}^{bd}
\end{aligned} \tag{8}$$


 A series of eight Feynman-like diagrams representing different terms in the equation.
 1. A wavy line with vertices labeled 'a' and 'i'.
 2. A wavy line with vertices labeled 'i' and 'a'.
 3. A wavy line with vertices labeled 'a' and 'i', with a curved arrow above it pointing from 'a' to 'i'.
 4. A wavy line with vertices labeled 'a' and 'i', with a curved arrow above it pointing from 'i' to 'a'.
 5. A wavy line with vertices labeled 'a' and 'i', with two curved arrows above it, one pointing from 'c' to 'k' and another from 'k' to 'c'.
 6. A wavy line with vertices labeled 'a' and 'i', with two curved arrows above it, one pointing from 'c' to 'l' and another from 'l' to 'c'.
 7. A wavy line with vertices labeled 'a' and 'i', with three curved arrows above it, one from 'd' to 'l', one from 'l' to 'c', and one from 'c' to 'k'.
 8. A wavy line with vertices labeled 'a' and 'i', with four curved arrows above it, one from 'd' to 'k', one from 'k' to 'l', one from 'l' to 'c', and one from 'c' to 'd'.

$$\begin{aligned}
0 = & \quad \text{Diagram 1} + \text{Diagram 2} \times \cancel{\text{Diagram 3}} + \text{Diagram 4} \times \cancel{\text{Diagram 5}} + \text{Diagram 6} \\
& + \text{Diagram 7} \times \cancel{\text{Diagram 8}} + \text{Diagram 9} \times \cancel{\text{Diagram 10}} + \text{Diagram 11} + \text{Diagram 12} \\
& + \text{Diagram 13} + \text{Diagram 14} \times \cancel{\text{Diagram 15}} + \text{Diagram 16} + \text{Diagram 17} \\
= & V_{ab}^{ij} + \hat{P}(ab|ij) \lambda_a^i X_b^j - \hat{P}(ab) \sum_k \lambda_a^k X_{kb}^{ij} + \hat{P}(ij) \sum_c \lambda_c^i X_{ab}^{cj} \\
& + \hat{P}(ab) \sum_c \lambda_{ac}^{ij} X_b^c - \hat{P}(ij) \sum_k \lambda_{ab}^{ik} X_k^j + \frac{1}{2} \sum_{cd} \lambda_{cd}^{ij} X_{ab}^{cd} + \frac{1}{2} \sum_{kl} \lambda_{ab}^{kl} X_{kl}^{ij} \\
& - \hat{P}(ab|ij) \sum_{kc} \lambda_{ac}^{kj} X_{kb}^{ic} - \hat{P}(ab) \frac{1}{2} \sum_{klcd} \lambda_{ad}^{kl} V_{cb}^{ij} t_{kl}^{cd} - \hat{P}(ij) \frac{1}{2} \sum_{kled} \lambda_{cd}^{il} V_{ab}^{kj} t_{kl}^{cd} \quad (9)
\end{aligned}$$

The diagrams are Feynman-like graphs representing terms in a field theory. They consist of vertices labeled with letters (a, b, c, d, i, j, k, l) and lines representing fields. Some lines are dashed, some are solid, and some have arrows indicating direction. A crossed-out diagram indicates it is zero. The equations show how these diagrams combine to form the terms in equation (9).

Appendix A

CCSD Diagrams

The following diagrams and their corresponding algebraic expressions comprise the different contributions to the CCSD cluster amplitudes *without* directly building the effective Hamiltonian, \bar{H} . The boxed diagrams are automatically zero in a Hartree-Fock basis. The contributions to the CCSD singles equation, (3.7), are given by Eqs. (A.1)-(A.8).

$$\hat{f}_N \hat{t}_1 |\Phi_0\rangle_c = \boxed{\begin{array}{c} \text{Diagram: } a \downarrow i \\ \text{Diagram: } a \downarrow i \end{array}} + \begin{array}{c} \text{Diagram: } a \downarrow i \\ \text{Diagram: } a \downarrow k \end{array} + \begin{array}{c} \text{Diagram: } i \downarrow a \\ \text{Diagram: } i \downarrow c \end{array} \\ = [f_i^a] - \sum_k f_i^k t_k^a + \sum_c f_c^a t_i^c \quad (\text{A.1})$$

$$\hat{V}_N \hat{t}_1 |\Phi_0\rangle_c = \begin{array}{c} \text{Diagram: } a \downarrow i \\ \text{Diagram: } c \circlearrowleft k \end{array} \\ = - \sum_{kc} V_{ic}^{ka} t_k^c \quad (\text{A.2})$$

$$\begin{aligned}
\hat{f}_N \hat{t}_2 |\Phi_0\rangle_c &= \text{Diagram} \\
&= \sum_{[kc]} f_c^k t_{ki}^{ac} \quad (\text{A.3})
\end{aligned}$$

$$\begin{aligned}
\hat{V}_N \hat{t}_2 |\Phi_0\rangle_c &= \text{Diagram} + \text{Diagram} \\
&= \frac{1}{2} \sum_{kcd} V_{cd}^{ka} t_{ki}^{cd} + \frac{1}{2} \sum_{klc} V_{ic}^{kl} t_{kl}^{ca} \quad (\text{A.4})
\end{aligned}$$

$$\begin{aligned}
\hat{f}_N \hat{t}_1^2 |\Phi_0\rangle_c &= \text{Diagram} \\
&= \sum_{[kcd]} \bar{f}_d^l \bar{t}_l^a \bar{t}_i^d \quad (\text{A.5})
\end{aligned}$$

$$\begin{aligned}
\hat{V}_N \hat{t}_1^2 |\Phi_0\rangle_c &= \text{Diagram} + \text{Diagram} \\
&= \sum_{kcd} V_{cd}^{ka} t_k^c t_i^d + \sum_{klc} V_{ic}^{kl} t_k^c t_l^a \quad (\text{A.6})
\end{aligned}$$

$$\begin{aligned}
\hat{V}_N \hat{t}_1 \hat{t}_2 |\Phi_0\rangle_c &= \text{Diagram 1} + \text{Diagram 2} + \text{Diagram 3} \\
&= -\frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_{ki}^{cd} t_l^a - \frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_{kl}^{ca} t_i^d + \sum_{klcd} V_{cd}^{kl} t_{il}^{ad} t_k^c
\end{aligned} \tag{A.7}$$

$$\begin{aligned}
\hat{V}_N \hat{t}_1^3 |\Phi_0\rangle_c &= \text{Diagram 1} \\
&= - \sum_{klcd} V_{cd}^{kl} t_k^c t_i^d t_l^a
\end{aligned} \tag{A.8}$$

The contributions to the CCSD doubles equation, (3.7), are given by Eqs. (A.9)-(A.19).

$$\begin{aligned}
\hat{V}_N |\Phi_0\rangle_c &= \text{Diagram 1} \\
&= V_{ij}^{ab}
\end{aligned} \tag{A.9}$$

$$\begin{aligned}
\hat{f}_N \hat{t}_2 |\Phi_0\rangle_c &= \text{Diagram 1} + \text{Diagram 2} \\
&= \hat{P}(ab) \sum_c f_c^b t_{ij}^{ac} - \hat{P}(ij) \sum_k f_j^k t_{ik}^{ab}
\end{aligned} \tag{A.10}$$

$$\hat{V}_N \hat{t}_1 |\Phi_0\rangle_c = \begin{array}{c} \text{Diagram showing two terms:} \\ \text{Term 1: } \text{Two vertical lines } a \text{ and } b \text{ meet at a point } i. \text{ From } i, \text{ a dashed line } k \text{ goes down to a base.} \\ \text{Term 2: } \text{Two vertical lines } i \text{ and } j \text{ meet at a point } a. \text{ From } a, \text{ a dashed line } c \text{ goes down to a base.} \end{array} + \hat{P}(ab) \sum_k V_{ij}^{kb} t_k^a + \hat{P}(ij) \sum_c V_{cj}^{ab} t_i^c \quad (\text{A.11})$$

$$\hat{V}_N \hat{t}_2 |\Phi_0\rangle_c = \begin{array}{c} \text{Diagram showing three terms:} \\ \text{Term 1: } \text{Two vertical lines } a \text{ and } b \text{ meet at a point } i. \text{ From } i, \text{ a dashed line } l \text{ goes down to a point } j. \\ \text{Term 2: } \text{Two vertical lines } i \text{ and } j \text{ meet at a point } a. \text{ From } a, \text{ a dashed line } c \text{ goes down to a point } d. \\ \text{Term 3: } \text{Two vertical lines } a \text{ and } b \text{ meet at a point } i. \text{ From } i, \text{ a dashed line } k \text{ goes down to a point } c. \end{array} + \frac{1}{2} \sum_{kl} V_{ij}^{kl} t_{kl}^{ab} + \frac{1}{2} \sum_{cd} V_{cd}^{ab} t_{ij}^{cd} - \hat{P}(ij|ab) \sum_{kc} V_{ic}^{kb} t_{kj}^{ac} \quad (\text{A.12})$$

$$\hat{V}_N \hat{t}_1^2 |\Phi_0\rangle_c = \begin{array}{c} \text{Diagram showing three terms:} \\ \text{Term 1: } \text{Two vertical lines } a \text{ and } b \text{ meet at a point } i. \text{ From } i, \text{ a dashed line } l \text{ goes down to a point } j. \\ \text{Term 2: } \text{Two vertical lines } i \text{ and } j \text{ meet at a point } a. \text{ From } a, \text{ a dashed line } c \text{ goes down to a point } d. \\ \text{Term 3: } \text{Two vertical lines } a \text{ and } b \text{ meet at a point } i. \text{ From } i, \text{ a dashed line } k \text{ goes down to a point } c. \end{array} + \sum_{kl} V_{ij}^{kl} t_k^a t_l^b + \sum_{cd} V_{cd}^{ab} t_i^c t_j^d - \hat{P}(ij|ab) \sum_{kc} V_{ic}^{kb} t_k^a t_j^c \quad (\text{A.13})$$

$$\begin{aligned}
\hat{V}_N \hat{t}_2^2 |\Phi_0\rangle_c &= \text{Diagram 1} + \text{Diagram 2} + \text{Diagram 3} \\
&\quad + \text{Diagram 4} \\
&= \frac{1}{4} \sum_{klcd} V_{cd}^{kl} t_{kl}^{ab} t_{ij}^{cd} + \hat{P}(ab) \sum_{klcd} V_{cd}^{kl} t_{lj}^{ac} t_{ki}^{bd} - \hat{P}(ij) \frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_{lj}^{ab} t_{ki}^{cd} \\
&\quad - \hat{P}(ab) \frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_{ij}^{db} t_{kl}^{ca} \tag{A.14}
\end{aligned}$$

$$\begin{aligned}
\hat{f}_N \hat{t}_1 \hat{t}_2 |\Phi_0\rangle_c &= \boxed{\text{Diagram 5}} + \boxed{\text{Diagram 6}} \\
&= -[\hat{P}(ab) \sum_{kc} \bar{f}_c^k \bar{t}_k^a \bar{t}_{ij}^{cb}] - [\hat{P}(ij) \sum_{kc} \bar{f}_c^k \bar{t}_i^c \bar{t}_{kj}^{ab}] \tag{A.15}
\end{aligned}$$

$$\begin{aligned}
\hat{V}_N \hat{t}_1 \hat{t}_2 |\Phi_0\rangle_c &= \text{Diagram 1} + \text{Diagram 2} + \text{Diagram 3} \\
&\quad + \text{Diagram 4} + \text{Diagram 5} + \text{Diagram 6} \\
&= \hat{P}(ij|ab) \sum_{kcd} V_{cd}^{ka} t_{jk}^{bc} t_i^d - \hat{P}(ij|ab) \sum_{klc} V_{ci}^{kl} t_{jk}^{bc} t_l^a - \hat{P}(ab) \frac{1}{2} \sum_{kcd} V_{cd}^{kb} t_{ij}^{cd} t_k^a \\
&\quad + \hat{P}(ij) \frac{1}{2} \sum_{klc} V_{cj}^{kl} t_{kl}^{ab} t_i^c + \hat{P}(ab) \sum_{kcd} V_{cd}^{ka} t_k^c t_{ij}^{db} - \hat{P}(ij) \sum_{klc} V_{ci}^{kl} t_k^c t_{lj}^{ab} \quad (\text{A.16})
\end{aligned}$$

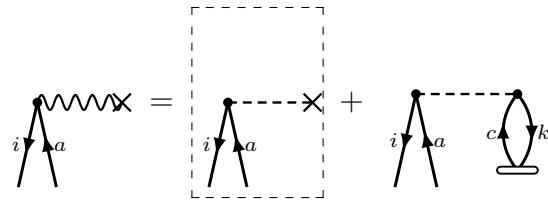
$$\begin{aligned}
\hat{V}_N \hat{t}_1^3 |\Phi_0\rangle_c &= \text{Diagram 7} + \text{Diagram 8} \\
&= -\hat{P}(ij|ab) \sum_{kcd} V_{cd}^{kb} t_k^a t_i^c t_j^d + \hat{P}(ij|ab) \sum_{klc} V_{cj}^{kl} t_i^c t_k^a t_l^b \quad (\text{A.17})
\end{aligned}$$

$$\begin{aligned}
\hat{V}_N \hat{t}_1^2 \hat{t}_2 |\Phi_0\rangle_c &= \text{Diagram 1} + \text{Diagram 2} + \text{Diagram 3} \\
&\quad + \text{Diagram 4} + \text{Diagram 5} \\
&= \frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_{kl}^{ab} t_i^c t_j^d + \frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_{ij}^{cd} t_k^a t_l^b + \hat{P}(ij|ab) \sum_{klcd} V_{cd}^{kl} t_{lj}^{ac} t_k^b t_i^d \\
&\quad - \hat{P}(ij) \frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_{lj}^{ab} t_k^c t_i^d - \hat{P}(ab) \frac{1}{2} \sum_{klcd} V_{cd}^{kl} t_{ij}^{db} t_k^c t_l^a
\end{aligned} \tag{A.18}$$

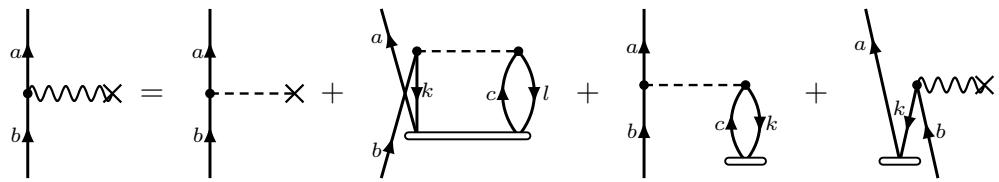
$$\begin{aligned}
\hat{V}_N \hat{t}_1^4 |\Phi_0\rangle_c &= \text{Diagram 6} \\
&= \sum_{klcd} V_{cd}^{kl} t_k^a t_l^b t_i^c t_j^d
\end{aligned} \tag{A.19}$$

The following diagrams and their corresponding algebraic expressions comprise the different components of the CCSD effective Hamiltonian, \bar{H} . Because some components are used as intermediates to build other components, they must be built in the order written. Some intermediate components overcount some diagrams which motivates the need for further intermediates, denoted by X' , X'' , and X''' . The boxed diagrams are automatically zero in a Hartree-Fock basis. The one-body components to the CCSD effective Hamiltonian,

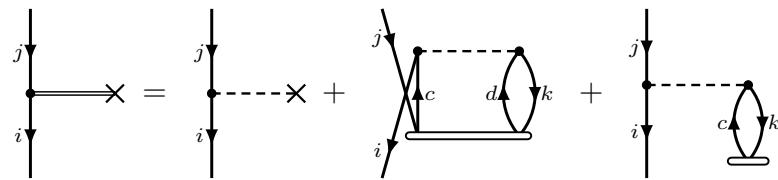
Eq. (3.5), are given by Eqs. (A.20)-(A.23).



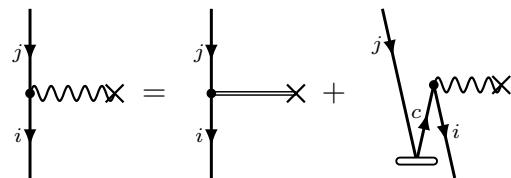
$$X_a^i = f_a^i + \sum_{kc} V_{ac}^{ik} t_k^c \quad (\text{A.20})$$



$$X_b^a = f_b^a - \frac{1}{2} \sum_{klc} V_{bc}^{kl} t_{kl}^{ac} + \sum_{kc} V_{cb}^{ka} t_k^c - \sum_k X_b^k t_k^a \quad (\text{A.21})$$

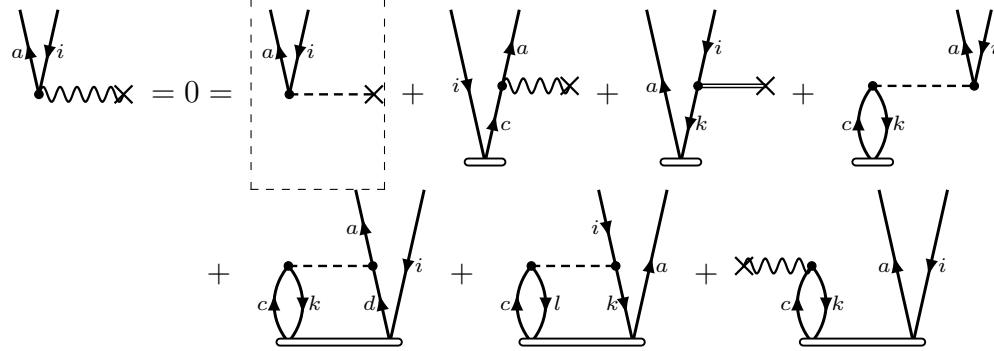


$$X_j^i = f_j^i + \frac{1}{2} \sum_{kcd} V_{cd}^{ik} t_{jk}^{cd} + \sum_{kc} V_{jc}^{ik} t_k^c \quad (\text{A.22})$$



$$X_j^i = X_j^{\prime i} + \sum_c X_c^i t_j^c \quad (\text{A.23})$$

Once the one-body components have been constructed, the pseudo-linear form for the CCSD singles equation, Eq. (3.6), can be evaluated,



$$\begin{aligned}
X_i^a = 0 = & \left[f_i^a \right] + \sum_c X_c^a t_i^c - \sum_k X'_i^k t_k^a + \sum_{kc} V_{ci}^{ka} t_k^c \\
& + \frac{1}{2} \sum_{bcd} V_{cd}^{ka} t_{ki}^{cd} - \frac{1}{2} \sum_{klc} V_{ic}^{kl} t_{kl}^{ac} + \sum_{kc} X_c^k t_{ik}^{ac}.
\end{aligned} \tag{A.24}$$

During a CC iteration, it's possible to use these updated singles amplitudes \hat{T}_1 when evaluating the doubles amplitudes \hat{T}_2 in the same iteration, which can accelerate the convergence.

The two-body components to the CCSD effective Hamiltonian, Eq. (3.5), are given by Eqs. (A.25)-(A.39).

$$X_{bc}^{ia} = V_{bc}^{ia} - \frac{1}{2} \sum_k V_{bc}^{ik} t_k^a \tag{A.25}$$

The diagram shows a vertical line with vertices a (up) and c (down). A wavy line connects a to a vertex b , which then connects to a vertex i . This is equated to the sum of two terms: a vertical line with vertices a (up) and c (down), a dashed line from a to b , and a vertex i ; and a vertical line with vertices a (up) and c (down), a dashed line from a to k , a wavy line from k to c , and a vertex i .

$$X_{bc}^{ia} = V_{bc}^{ia} - \sum_k V_{bc}^{ik} t_k^a \quad (\text{A.26})$$

The diagram shows a vertical line with vertices k (up) and i (down). A horizontal line connects k to a vertex j , which then connects to a vertex a . This is equated to the sum of two terms: a vertical line with vertices k (up) and i (down), a dashed line from k to j , and a vertex a ; and a vertical line with vertices k (up) and i (down), a dashed line from k to c , a wavy line from c to i , a horizontal line from c to j , and a vertex a .

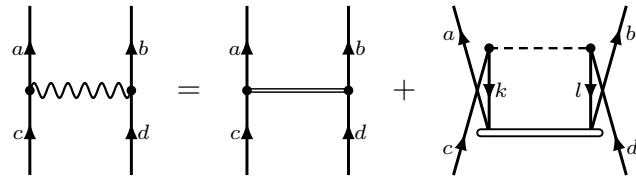
$$X'_{ka}^{ij} = V_{ka}^{ij} + \frac{1}{2} \sum_c V_{cat}^{ij} t_k^c \quad (\text{A.27})$$

The diagram shows a vertical line with vertices k (up) and i (down). A wavy line connects k to a vertex j , which then connects to a vertex a . This is equated to the sum of two terms: a vertical line with vertices k (up) and i (down), a dashed line from k to j , and a vertex a ; and a vertical line with vertices k (up) and i (down), a dashed line from k to c , a wavy line from c to i , a horizontal line from c to j , and a vertex a .

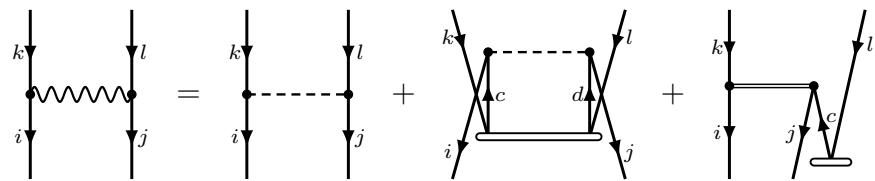
$$X'_{ka}^{ij} = V_{ka}^{ij} + \sum_c V_{cat}^{ij} t_k^c \quad (\text{A.28})$$

The diagram shows a vertical line with vertices a (up) and c (down) connected by a horizontal line to a vertical line with vertices b (up) and d (down). This is equated to the sum of two terms: a vertical line with vertices a (up) and c (down), a dashed line from a to b , and a vertical line with vertices b (up) and d (down); and a vertical line with vertices a (up) and c (down), a dashed line from a to k , a wavy line from k to c , a horizontal line from k to b , and a vertical line with vertices b (up) and d (down).

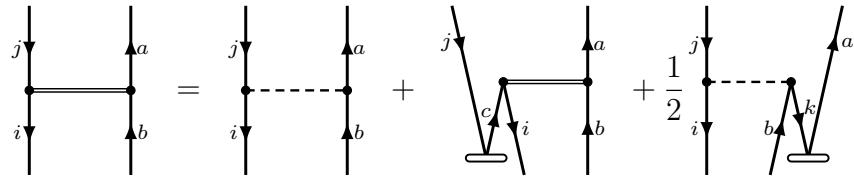
$$X'_{cd}^{ab} = V_{cd}^{ab} - \hat{P}(ab) \sum_k X'_{cd}^{kb} t_k^a \quad (\text{A.29})$$



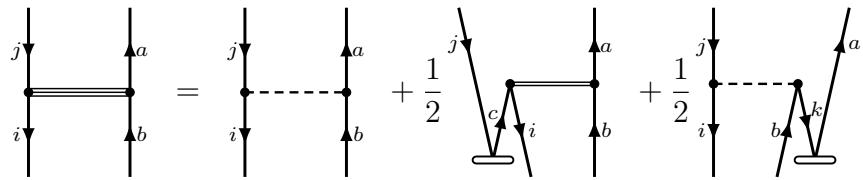
$$X_{cd}^{ab} = X'_{cd}^{ab} + \frac{1}{2} \sum_{kl} V_{cd}^{kl} t_{kl}^{ab} \quad (\text{A.30})$$



$$X_{kl}^{ij} = V_{kl}^{ij} + \frac{1}{2} \sum_{cd} V_{cd}^{ij} t_{kl}^{cd} + \hat{P}(kl) \sum_c X'_{kc}^{ij} t_l^c \quad (\text{A.31})$$



$$X'_{jb}^{ia} = V_{jb}^{ia} + \sum_c X'_{cb}^{ia} t_j^c - \frac{1}{2} \sum_k V_{jb}^{ik} t_k^a \quad (\text{A.32})$$



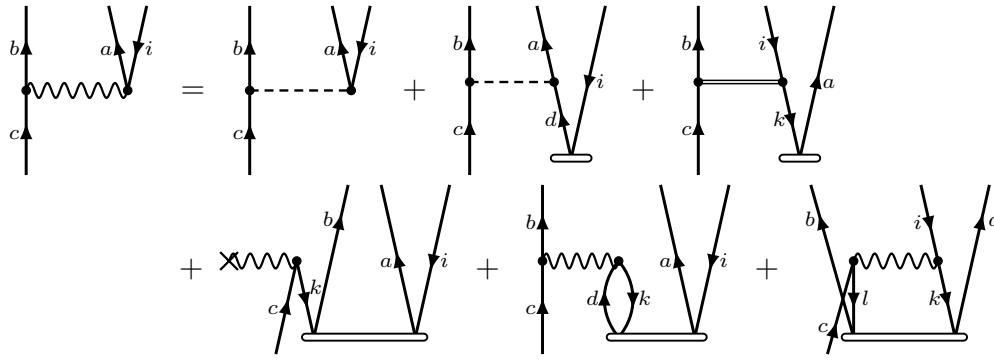
$$X''_{jb}^{ia} = V_{jb}^{ia} + \frac{1}{2} \sum_c X'_{cb}^{ia} t_j^c - \frac{1}{2} \sum_k V_{jb}^{ik} t_k^a \quad (\text{A.33})$$

$$X'''_{jb}^{ia} = V_{jb}^{ia} + \frac{1}{2} \sum_c X_{cb}^{ia} t_j^c - \sum_k V_{jb}^{ik} t_k^a \quad (\text{A.34})$$

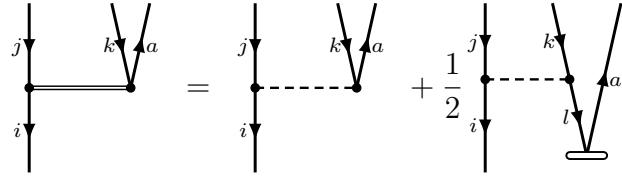
$$X_{jb}^{ia} = X'''_{jb}^{ia} - \left(\frac{1}{2}\right) \sum_{kc} V_{cb}^{ik} t_{jk}^{ca} + \frac{1}{2} \sum_c X_{cb}^{ia} t_j^c \quad (\text{A.35})$$

The factor of $\left(\frac{1}{2}\right)$ is applied when solving the CCSD equations but omitted when applying the effective Hamiltonian in post-CC methods.

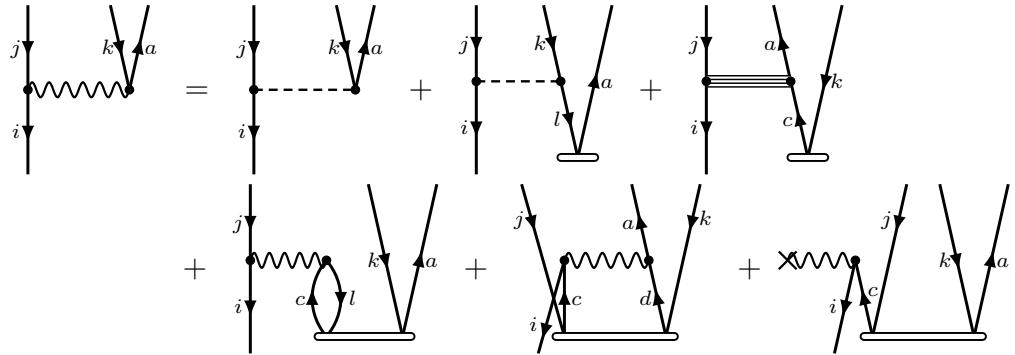
$$X'_{ic}^{ab} = V_{ic}^{ab} + \frac{1}{2} \sum_d V_{dc}^{ab} t_i^d - \hat{P}(ab) \sum_k X''_{ic}^{kb} t_k^a \quad (\text{A.36})$$



$$\begin{aligned}
 X_{ic}^{ab} = & V_{ic}^{ab} + \sum_d V_{dc}^{ab} t_i^d - \hat{P}(ab) \sum_k X'_{ic}^{kb} t_k^a \\
 & - \sum_k X_c^k t_{ik}^{ab} + \hat{P}(ab) \sum_{kd} X_{dc}^{kb} t_{ik}^{ad} + \frac{1}{2} \sum_{kl} X_{ic}^{kl} t_{kl}^{ab}
 \end{aligned} \tag{A.37}$$



$$X_{jk}^{ia} = V_{jk}^{ia} - \frac{1}{2} \sum_l V_{jk}^{il} t_l^a \tag{A.38}$$



$$\begin{aligned}
 X_{jk}^{ia} = & V_{jk}^{ia} - \sum_l V_{jk}^{il} t_l^a + \hat{P}(jk) \sum_c X'''_{jc}^{ia} t_k^c \\
 & + \hat{P}(jk) \sum_{lc} X_{jc}^{il} t_{lk}^{ca} + \frac{1}{2} \sum_{cd} X_{cd}^{ia} t_{jk}^{cd} + \sum_c X_c^i t_{jk}^{ca}
 \end{aligned} \tag{A.39}$$

$$\begin{aligned}
& \text{Diagram 1: } a \downarrow i \quad b \downarrow j = 0 = \\
& \quad \text{Diagram 2: } a \downarrow i \quad b \downarrow j + \text{Diagram 3: } a \downarrow i \quad b \downarrow j + \text{Diagram 4: } a \downarrow i \quad b \downarrow j \\
& + \text{Diagram 5: } a \downarrow i \quad b \downarrow j + \text{Diagram 6: } a \downarrow i \quad b \downarrow j + \text{Diagram 7: } a \downarrow i \quad b \downarrow j + \text{Diagram 8: } a \downarrow i \quad b \downarrow j \\
X_{ij}^{ab} &= 0 = V_{ij}^{ab} + \hat{P}(ab) \sum_c X_c^a t_{ij}^{cb} - \hat{P}(ij) \sum_k X_i^k t_{kj}^{ab} \\
&+ \frac{1}{2} \sum_{cd} X'_{cd}^{ab} t_{ij}^{cd} + \frac{1}{2} \sum_{kl} X_{ij}^{kl} t_{kl}^{ab} - \hat{P}(ab|ij) \sum_{kc} X_{ic}^{kb} t_{kj}^{ac} - \hat{P}(ab) \sum_k X'_{ij}^{kb} t_k^a + \hat{P}(ij) \sum_c X'_{ic}^{ab} t_j^c
\end{aligned} \tag{A.40}$$

Appendix B

Computational Implementation

The sums involved in building the CC effective Hamiltonian, solving the CC equations, solving the EOM-CC equations, and building effective operators can all be reformulated as matrix-matrix multiplications and thus performed with efficient LAPACK and BLAS routines. To take advantage of this efficiency, the various cluster amplitudes and matrix elements must be grouped into structures with similar index organization so that summed indices map to the same states and matrix elements. An additional benefit to these structures is that angular-momentum-coupling coefficients are automatically removed by summing over Clebsch-Gordon coefficients ($\sum_{m_1 m_2} C_{m_1 m_2 M}^{j_1 j_2 J} C_{m_1 m_2 M'}^{j_1 j_2 J'} = \delta_{JJ'} \delta_{MM'}$).

Channel Definitions

Each matrix structure is separated into different symmetry channels for different permutations of its indices. The channels are denoted as $\Sigma_{\vec{\xi}}$, where $\vec{\xi}$ represents the relevant quantum numbers of a certain channel. There are four different channel types that are relevant for the structures used in this work.

The direct two-body channel categorizes the vector sum of two single-particle-state quantum numbers, $\Sigma_{\vec{\xi}_1}$,

$$\vec{\xi}_{pq} = \vec{\xi}_p + \vec{\xi}_q \quad \longrightarrow \quad |pq\rangle \in \Sigma_{\vec{\xi}_1 = \vec{\xi}_{pq}}. \quad (\text{B.1})$$

The cross two-body channel categorizes the vector difference of two single-particle-state

quantum numbers or, equivalently, the vector sum of a single-particle state and a time-reversed single-particle state, $\Sigma_{\vec{\xi}_2}$,

$$\vec{\xi}_{p\bar{q}} = \vec{\xi}_p - \vec{\xi}_q = \vec{\xi}_p + \vec{\xi}_{\bar{q}} \quad \longrightarrow \quad |p\bar{q}\rangle \in \Sigma_{\vec{\xi}_2}. \quad (\text{B.2})$$

The one-body channel categorizes single-particle states by their vector quantum numbers,

$$\Sigma_{\vec{\xi}_3},$$

$$\vec{\xi}_p \quad \longrightarrow \quad |p\rangle \in \Sigma_{\vec{\xi}_3}. \quad (\text{B.3})$$

The cross three-body state categorizes the vector difference between the quantum numbers of a direct two-body state and a single-particle state or, equivalently, the vector sum of the quantum numbers of a two-body direct state and a time-reversed single-particle state, $\Sigma_{\vec{\xi}_3}$,

$$\vec{\xi}_{pq\bar{r}} = \vec{\xi}_p + \vec{\xi}_q - \vec{\xi}_{\bar{r}} = \vec{\xi}_p + \vec{\xi}_q + \vec{\xi}_{\bar{r}} \quad \longrightarrow \quad |pq\bar{r}\rangle \in \Sigma_{\vec{\xi}_3=\vec{\xi}_{pq\bar{r}}}$$

Structure Definitions

Different matrix structures are indexed by their channel type: 1 for direct channels, 2 for cross channels, and 3 for one-/three-body channels. Matrices with more than one structure of the same type, there is an additional index that depends on the specific permutation involved.

For a one-body operator $A_q^p \left\{ \hat{a}_p^\dagger \hat{a}_q \right\}$, there is a direct-channel matrix element and a

cross-channel matrix element,

$$\mathbf{A}_1 = A_q^p \quad \mathbf{A}_2 = A^{p\bar{q}} \quad (\text{B.5})$$

For a two-body operator $A_{rs}^{pq} \left\{ \hat{a}_p^\dagger \hat{a}_q^\dagger \hat{a}_s \hat{a}_r \right\}$, there is a direct-channel matrix element, four cross-channel matrix elements, and four one-channel matrix elements,

$$\begin{aligned} \mathbf{A}_1 &= A_{r\bar{s}}^{pq} \\ \mathbf{A}_{21} &= A_{r\bar{q}}^{p\bar{s}} \quad \mathbf{A}_{22} = A_{s\bar{p}}^{q\bar{r}} \quad \mathbf{A}_{23} = A_{s\bar{q}}^{p\bar{r}} \quad \mathbf{A}_{24} = A_{r\bar{p}}^{q\bar{s}} \\ \mathbf{A}_{31} &= A_{rs\bar{q}}^p \quad \mathbf{A}_{32} = A_{rs\bar{p}}^q \quad \mathbf{A}_{33} = A_r^{pq\bar{s}} \quad \mathbf{A}_{34} = A_s^{pq\bar{r}} \end{aligned} \quad (\text{B.6})$$

For an EOM operator of the form $A_r^{pq} \left\{ \hat{a}_p^\dagger \hat{a}_q^\dagger \hat{a}_r \right\}$, there is a direct-channel matrix element, a one-channel matrix element, and two cross-channel matrix elements,

$$\begin{aligned} \mathbf{A}_1 &= A_r^{pq} \quad \mathbf{A}_3 = A^{pq\bar{r}} \\ \mathbf{A}_{21} &= A_{r\bar{q}}^p \quad \mathbf{A}_{22} = A_{r\bar{p}}^q \end{aligned} \quad (\text{B.7})$$

EOM operators of the form $A_{qr}^p \left\{ \hat{a}_p^\dagger \hat{a}_r \hat{a}_q \right\}$ has similar structures,

$$\begin{aligned} \mathbf{A}_1 &= A_{qr}^p \quad \mathbf{A}_3 = A_{qr\bar{p}} \\ \mathbf{A}_{21} &= A_q^{p\bar{r}} \quad \mathbf{A}_{22} = A_r^{p\bar{q}} \end{aligned} \quad (\text{B.8})$$

\bar{H} , Matrix Form

$$\begin{aligned} X_a^i &= \lceil \bar{f}_a^i \rceil + V_{c\bar{k}}^{i\bar{a}} t^{c\bar{k}} \\ \mathbf{X}_2^{hp} &\leftarrow \lceil \bar{\mathbf{f}}_2^{hp} \rceil + \mathbf{V}_{23}^{hhpp} \cdot \mathbf{t}_2 \end{aligned} \quad (\text{B.9})$$

$$\begin{aligned} X_b^a &= f_b^a - \frac{1}{2} t_{kl\bar{c}}^a V_b^{kl\bar{c}} + V_{c\bar{k}}^{a\bar{b}} t^{c\bar{k}} - t_k^a X_b^k \\ \mathbf{X}_3^{pp} &\leftarrow -\frac{1}{2} \mathbf{t}_{31} \cdot \mathbf{V}_{33}^{hhpp} - \mathbf{t}_3 \cdot \mathbf{X}_3^{hp} \\ \mathbf{X}_2^{pp} &\leftarrow \mathbf{f}_2^{pp} + \mathbf{V}_{24}^{hPPP} \cdot \mathbf{t}_2 \end{aligned} \quad (\text{B.10})$$

$$\begin{aligned} X_j'^i &= f_j^i + \frac{1}{2} V_{cd\bar{k}}^i t_j^{cd\bar{k}} + V_{c\bar{k}}^{i\bar{j}} t^{c\bar{k}} \\ \mathbf{X}'_3^{hh} &\leftarrow \frac{1}{2} \mathbf{V}_{31}^{hhpp} \cdot \mathbf{t}_{33} \\ \mathbf{X}'_2^{hh} &\leftarrow \mathbf{f}_2^{hh} + \mathbf{V}_{23}^{hhhp} \cdot \mathbf{t}_2 \end{aligned} \quad (\text{B.11})$$

$$\begin{aligned} X_j^i &= f_j^i + \frac{1}{2} V_{cd\bar{k}}^i t_j^{cd\bar{k}} + V_{c\bar{k}}^{i\bar{j}} t^{c\bar{k}} + X_c^i t_j^c \\ \mathbf{X}_3^{hh} &\leftarrow \frac{1}{2} \mathbf{V}_{31}^{hhpp} \cdot \mathbf{t}_{33} + \mathbf{X}_3^{hp} \cdot \mathbf{t}_3 \\ \mathbf{X}'_2^{hh} &\leftarrow \mathbf{f}_2^{hh} + \mathbf{V}_{23}^{hhhp} \cdot \mathbf{t}_2 \end{aligned} \quad (\text{B.12})$$

$$\begin{aligned}
X_i^a &= \lceil f_i^a \rceil + X_c^a t_i^c - t_k^a X_i'^k - V_{ck}^{a\bar{i}} t^{c\bar{k}} + \frac{1}{2} V_{cd\bar{k}}^a t_i^{cd\bar{k}} - \frac{1}{2} t_{kl\bar{c}}^a V_i^{kl\bar{c}} + t_{k\bar{c}}^{a\bar{i}} X^{k\bar{c}} \\
\mathbf{X}_3^{ph} &\leftarrow \mathbf{X}_3^{pp} \cdot \mathbf{t}_3 - \mathbf{t}_3 \cdot \mathbf{X}_3'^{hh} + \frac{1}{2} \mathbf{V}_{32}^{hppp} \cdot \mathbf{t}_{34} - \frac{1}{2} \mathbf{t}_{31} \cdot \mathbf{V}_{33}^{hhhp} \\
\mathbf{X}_2^{ph} &\leftarrow \lceil \mathbf{f}_2^{ph} \rceil - \mathbf{V}_{22}^{hphp} \cdot \mathbf{t}_2 + \mathbf{t}_{23} \cdot \mathbf{X}_2^{hp}
\end{aligned} \tag{B.13}$$

$$\begin{aligned}
X_{bc}'^{ia} &= V_{bc}^{ia} - \frac{1}{2} t_k^a V_{bc\bar{i}}^k \\
\mathbf{X}_{32}'^{hppp} &\leftarrow \mathbf{V}_{32}^{hppp} - \frac{1}{2} \mathbf{t}_3 \cdot \mathbf{V}_{32}^{hhpp}
\end{aligned} \tag{B.14}$$

$$\begin{aligned}
X_{bc}^{ia} &= V_{bc}^{ia} - t_k^a V_{bc\bar{i}}^k \\
\mathbf{X}_{32}^{hppp} &\leftarrow \mathbf{V}_{32}^{hppp} - \mathbf{t}_3 \cdot \mathbf{V}_{32}^{hhpp}
\end{aligned} \tag{B.15}$$

$$\begin{aligned}
X_{ka}'^{ij} &= V_{ka}^{ij} + \frac{1}{2} V_c^{ij\bar{a}} t_k^c \\
\mathbf{X}_{33}'^{hhhp} &\leftarrow \mathbf{V}_{ka}^{ij} + \frac{1}{2} \mathbf{V}_{33}^{hhpp} \cdot \mathbf{t}_3
\end{aligned} \tag{B.16}$$

$$\begin{aligned}
X_{ka}^{ij} &= V_{ka}^{ij} + V_c^{ij\bar{a}} t_k^c \\
\mathbf{X}_{33}^{hhhp} &\leftarrow \mathbf{V}_{ka}^{ij} + \mathbf{V}_{33}^{hhpp} \cdot \mathbf{t}_3
\end{aligned} \tag{B.17}$$

$$\begin{aligned}
X'_{cd}^{ab} &= V_{cd}^{ab} - \hat{P}(ab) t_k^a X'_{cd\bar{b}}^{lk} \\
\mathbf{X}'_1^{pppp} &\longleftarrow \mathbf{V}_1^{pppp} \\
\mathbf{X}'_{3(2)}^{pppp} &\longleftarrow \mp \mathbf{t}_3 \cdot \mathbf{X}'_{31}^{hppp}
\end{aligned} \tag{B.18}$$

$$\begin{aligned}
X_{cd}^{ab} &= X'_{cd}^{ab} + \frac{1}{2} t_{kl}^{ab} V_{cd}^{kl} \\
\mathbf{X}_1^{pppp} &\longleftarrow \mathbf{X}'_1^{pppp} + \frac{1}{2} \mathbf{t}_1 \cdot \mathbf{V}_1^{happ}
\end{aligned} \tag{B.19}$$

$$\begin{aligned}
X_{kl}^{ij} &= V_{kl}^{ij} + \frac{1}{2} V_{cd}^{ij} t_{kl}^{cd} + \hat{P}(kl) X_c^{ij\bar{k}} t_l^c \\
\mathbf{X}_1^{hhhh} &\longleftarrow \mathbf{V}_1^{hhhh} + \frac{1}{2} \mathbf{V}_1^{happ} \cdot \mathbf{t}_1 \\
\mathbf{X}_{3(4)}^{hhhh} &\longleftarrow \mp \mathbf{X}'_{34}^{hhhp} \cdot \mathbf{t}_3
\end{aligned} \tag{B.20}$$

$$\begin{aligned}
X_{jb}^{ia} &= V_{jb}^{ia} + X_c^{i\bar{a}\bar{b}} t_j^c - \frac{1}{2} t_k^a V_{j\bar{b}\bar{i}}^k \\
\mathbf{X}'_{21}^{hphp} &\longleftarrow \mathbf{V}_{21}^{hphp} \\
\mathbf{X}'_{33}^{hphp} &\longleftarrow \mathbf{X}'_{33}^{hphp} \cdot \mathbf{t}_3 \\
\mathbf{X}'_{32}^{hphp} &\longleftarrow -\frac{1}{2} \mathbf{t}_3 \cdot \mathbf{V}_{32}^{hphp}
\end{aligned} \tag{B.21}$$

$$\begin{aligned}
X''_{j\bar{b}}^{ia} &= V_{j\bar{b}}^{ia} + \frac{1}{2} X_c'^{i\bar{a}\bar{b}} t_j^c - \frac{1}{2} t_k^a V_{j\bar{b}\bar{a}}^k \\
\mathbf{X}''_{2_1}^{hphp} &\leftarrow \mathbf{V}_{2_1}^{hphp} \\
\mathbf{X}''_{3_3}^{hphp} &\leftarrow \frac{1}{2} \mathbf{X}'_{3_3}^{hppp} \cdot \mathbf{t}_3 \\
\mathbf{X}''_{3_2}^{hphp} &\leftarrow -\frac{1}{2} \mathbf{t}_3 \cdot \mathbf{V}_{3_2}^{hhhp} \tag{B.22}
\end{aligned}$$

$$\begin{aligned}
X'''_{j\bar{b}}^{ia} &= V_{j\bar{b}}^{ia} + \frac{1}{2} X_c'^{i\bar{a}\bar{b}} t_j^c - t_k^a V_{j\bar{b}\bar{a}}^k \\
\mathbf{X}'''_{2_1}^{hphp} &\leftarrow \mathbf{V}_{2_1}^{hphp} \\
\mathbf{X}'''_{3_3}^{hphp} &\leftarrow \frac{1}{2} \mathbf{X}'_{3_3}^{hppp} \cdot \mathbf{t}_3 \\
\mathbf{X}'''_{3_2}^{hphp} &\leftarrow -\mathbf{t}_3 \cdot \mathbf{V}_{3_2}^{hhhp} \tag{B.23}
\end{aligned}$$

$$\begin{aligned}
X_{j\bar{b}}^{ia} &= V_{j\bar{b}}^{ia} + X_c'^{i\bar{a}\bar{b}} t_j^c - t_k^a V_{j\bar{b}\bar{a}}^k - \left(\frac{1}{2}\right) V_{c\bar{k}}^{i\bar{b}} t_{j\bar{a}}^{c\bar{k}} \\
\mathbf{X}_{2_1}^{hphp} &\leftarrow \mathbf{V}_{2_1}^{hphp} - \left(\frac{1}{2}\right) \mathbf{V}_{2_1}^{hhhp} \mathbf{t}_{2_1} \\
\mathbf{X}_{3_3}^{hphp} &\leftarrow \mathbf{X}_{3_3}^{hppp} \cdot \mathbf{t}_3 \\
\mathbf{X}_{3_2}^{hphp} &\leftarrow -\mathbf{t}_3 \cdot \mathbf{V}_{3_2}^{hhhp} \tag{B.24}
\end{aligned}$$

$$\begin{aligned}
X'_{ic}^{ab} &= V_{ic}^{ab} + \frac{1}{2} V_d^{ab\bar{c}} t_i^d - \hat{P}(ab) t_k^a X''_{ic\bar{b}}^{ik} \\
\mathbf{X}'_{3_3}^{pphp} &\leftarrow \mathbf{V}_{3_3}^{pphp} + \frac{1}{2} \mathbf{V}_{3_3}^{pppp} \cdot \mathbf{t}_3 \\
\mathbf{X}'_{3_{1(2)}}^{pphp} &\leftarrow \mp \mathbf{t}_3 \cdot \mathbf{X}''_{3_1}^{hphp} \tag{B.25}
\end{aligned}$$

$$\begin{aligned}
X_{ic}^{ab} &= V_{ic}^{ab} + V_d^{ab\bar{c}} \mathbf{t}_i^d - \hat{P}(ab) t_k^a X_{ic\bar{b}}'^k - t_k^{ab\bar{i}} X_c^k + \hat{P}(ab) t_{k\bar{d}}^{a\bar{i}} X_{c\bar{b}}^{k\bar{d}} + \frac{1}{2} t_{kl}^{ab} X_{ic}^{kl} \\
\mathbf{X}_{33}^{pphp} &\leftarrow \mathbf{V}_{33}^{pphp} + \mathbf{V}_{33}^{pppp} \cdot \mathbf{t}_3 \\
\mathbf{X}_{31(2)}^{pphp} &\leftarrow \mp \mathbf{t}_3 \cdot \mathbf{X}_{31}^{hphp} \\
\mathbf{X}_{34}^{pphp} &\leftarrow -\mathbf{t}_{34} \cdot \mathbf{X}_3^{hp} \\
\mathbf{X}_{22(3)}^{pphp} &\leftarrow \mp \mathbf{t}_{23} \cdot \mathbf{X}_{23}^{hphp} \\
\mathbf{X}_1^{pphp} &\leftarrow \frac{1}{2} \mathbf{t}_1 \cdot \mathbf{X}_1^{hhhp} \tag{B.26}
\end{aligned}$$

$$\begin{aligned}
X_{jk}'^{ia} &= V_{jk}^{ia} - \frac{1}{2} t_l^a V_{jk\bar{i}}^l \\
\mathbf{X}_{32}^{hphh} &\leftarrow \mathbf{V}_{32}^{hphh} - \frac{1}{2} \mathbf{t}_3 \cdot \mathbf{V}_{32}^{hhhh} \tag{B.27}
\end{aligned}$$

$$\begin{aligned}
X_{jk}^{ia} &= V_{jk}^{ia} - t_l^a V_{jk\bar{i}}^l + \hat{P}(jk) X_{c\bar{l}}'''^{ia\bar{j}} t_k^c + \hat{P}(jk) X_{c\bar{l}}^{i\bar{j}} t_{k\bar{a}}^{c\bar{l}} + \frac{1}{2} X_{cd}^{ia} t_{jk}^{cd} + X_{cd}^i t_{jk}^c \\
\mathbf{X}_{32}^{hphh} &\leftarrow \mathbf{V}_{32}^{hphh} - \mathbf{t}_3 \cdot \mathbf{V}_{32}^{hhhh} \\
\mathbf{X}_{33(4)}^{hphh} &\leftarrow \mp \mathbf{X}_{34}'''^{hphp} \cdot \mathbf{t}_3 \\
\mathbf{X}_{21(3)}^{hphh} &\leftarrow \mp \mathbf{X}_{23}^{hhhp} \cdot \mathbf{t}_{23} \\
\mathbf{X}_1^{hphh} &\leftarrow \frac{1}{2} \mathbf{X}_1^{hphp} \cdot \mathbf{t}_1 \\
\mathbf{X}_{31}^{hphh} &\leftarrow \mathbf{X}_3^{hp} \cdot \mathbf{t}_{31} \tag{B.28}
\end{aligned}$$

$$\begin{aligned}
X_{ij}^{ab} &= V_{ij}^{ab} + \hat{P}(ab) X_c^a t_{ij\bar{b}}^c - \hat{P}(ij) t_k^{ab\bar{j}} X_i^k + \frac{1}{2} X_{cd}^{tab} t_{ij}^{cd} + \frac{1}{2} t_{kl}^{ab} X_{ij}^{kl} \\
&\quad - \hat{P}(ab|ij) t_{k\bar{c}}^{a\bar{j}} X_{i\bar{b}}^{k\bar{c}} - \hat{P}(ab) t_k^a X_{ij\bar{b}}^{ik} + \hat{P}(ij) X_c^{ab\bar{i}} t_j^c \\
\mathbf{X}_1^{pphh} &\leftarrow \mathbf{V}_1^{pphh} + \frac{1}{2} \mathbf{X}'_1^{pppp} \cdot \mathbf{t}_1 + \frac{1}{2} \mathbf{t}_1 \cdot \mathbf{X}_1^{hhhh} \\
\mathbf{X}_{3_{1(2)}}^{pphh} &\leftarrow \pm \mathbf{X}_3^{pp} \cdot \mathbf{t}_{3_1} \mp \mathbf{t}_3 \mathbf{X}'_{3_1}^{hpjh} \\
\mathbf{X}_{3_{3(4)}}^{pphh} &\leftarrow \mp \mathbf{t}_{3_3} \cdot \mathbf{X}_3^{hh} \pm \mathbf{X}'_{3_4}^{pphp} \mathbf{t}_3 \\
\mathbf{X}_{2_{1(2)}}^{pphh} &\leftarrow -\mathbf{t}_{2_1} \cdot \mathbf{X}_{2_1}^{hpjh} \\
\mathbf{X}_{2_{3(4)}}^{pphh} &\leftarrow \mathbf{t}_{2_1} \cdot \mathbf{X}_{2_1}^{hpjh} \tag{B.29}
\end{aligned}$$

\hat{R}_k^+ , Matrix Form

$$\begin{aligned}
\omega_k r^a &= X_c^a r^c + r_{k\bar{c}}^a X^{k\bar{c}} - \frac{1}{2} X_{cd\bar{k}}^a r^{cd\bar{k}} \\
\omega_k \mathbf{r} &\leftarrow \mathbf{X}_3^{pp} \cdot \mathbf{r} + \mathbf{r}_{2_1} \cdot \mathbf{X}_2^{hp} - \frac{1}{2} \mathbf{X}_{3_2}^{hppp} \cdot \mathbf{r}_3 \tag{B.30}
\end{aligned}$$

$$\begin{aligned}
\omega_k r_i^{ab} &= -X_c^{ab\bar{i}} r^c + \hat{P}(ab) X_c^b r_{i\bar{a}}^c - r_k^{ab} X_i^k + \frac{1}{2} X_{cd}^{ab} r_i^{cd} - \hat{P}(ab) r_{k\bar{c}}^b X_{i\bar{a}}^{k\bar{c}} - \frac{1}{2} \mathbf{t}_k^{ab\bar{i}} V_{cd\bar{l}}^k r^{cd\bar{l}} \\
\omega_k \mathbf{r}_3 &\leftarrow -\mathbf{X}_{3_4}^{pphp} \cdot \mathbf{r} - \frac{1}{2} \mathbf{t}_{3_3} \cdot \mathbf{V}_{3_1}^{hhpp} \cdot \mathbf{r}_3 \\
\omega_k \mathbf{r}_{2_{1(2)}} &\leftarrow \mp \mathbf{X}_3^{pp} \cdot \mathbf{r}_{2_2} \pm \mathbf{r}_{2_2} \cdot \mathbf{X}_{2_1}^{hpjh} \\
\omega_k \mathbf{r}_1 &\leftarrow -\mathbf{r}_1 \cdot \mathbf{X}_3^{hh} + \frac{1}{2} \mathbf{X}_1^{pppp} \cdot \mathbf{r}_1 \tag{B.31}
\end{aligned}$$

\hat{L}_k^+ , Matrix Form

The difference for the left eigenproblem is that disconnected term is computed as an outer product rather than with matrix-matrix multiplication.

$$\begin{aligned} E_k l_a &= l_c X_a^c - \frac{1}{2} l_{cd\bar{k}} X_a^{cd\bar{k}} \\ E_k \mathbf{l} &\leftarrow \mathbf{l} \cdot \mathbf{X}_3^{pp} - \frac{1}{2} \mathbf{l}_3 \cdot \mathbf{X}_{3_4}^{pphp} \end{aligned} \quad (\text{B.32})$$

$$\begin{aligned} E_k l_{ab}^i &= \hat{P}(ab) l_a X^{i\bar{b}} - l_c X_{ab\bar{i}}^c - X_k^i l_{ab}^k + \hat{P}(ab) l_c^{i\bar{a}} X_b^c + \frac{1}{2} l_{cd}^i X_{ab}^{cd} \\ &\quad - \hat{P}(ab) X_{k\bar{c}}^{i\bar{a}} l_b^{k\bar{c}} - \frac{1}{2} l_{cd\bar{l}} t_k^{cd\bar{l}} V_{ab\bar{i}}^k \\ E_k l_3 &\leftarrow -\mathbf{l} \cdot \mathbf{X}_{3_2}^{happ} - \frac{1}{2} \mathbf{l}_3 \cdot \mathbf{t}_{3_3} \cdot \mathbf{V}_{3_1}^{hhpp} \\ E_k \mathbf{l}_{2_{1(2)}} &\leftarrow \pm \mathbf{l} \otimes \mathbf{X}_2^{hp} \mp \mathbf{l}_{2_2} \cdot \mathbf{X}_3^{pp} \pm \mathbf{X}_{2_1}^{hphp} \cdot \mathbf{l}_{2_2} \\ E_k \mathbf{l}_1 &\leftarrow -\mathbf{X}_3^{hh} \cdot \mathbf{l}_1 + \frac{1}{2} \mathbf{l}_1 \cdot \mathbf{X}_1^{pppp} \end{aligned} \quad (\text{B.33})$$

\hat{R}_k^- , Matrix Form

$$\begin{aligned} \omega_k r_i &= -r_k X_i^k + X_{c\bar{k}} r_i^{c\bar{k}} - \frac{1}{2} r_{kl\bar{c}} X_i^{kl\bar{c}} \\ \omega_k \mathbf{r} &\leftarrow -\mathbf{r} \cdot \mathbf{X}_3^{hh} + \mathbf{X}_{2'}^{hp} \cdot \mathbf{r}_{2_1} - \frac{1}{2} \mathbf{r}_3 \cdot \mathbf{X}_{3_3}^{hhhp} \end{aligned} \quad (\text{B.34})$$

$$\begin{aligned}
\omega_k r_{ij}^a &= -r_k X_{ij\bar{a}}^k - \hat{P}(ij) r_k^{a\bar{i}} X_j^k + X_c^a r_{ij}^c + \frac{1}{2} r_{kl}^a X_{ij}^{kl} - \hat{P}(ij) X_{c\bar{k}}^{a\bar{i}} r_j^{c\bar{k}} - \frac{1}{2} r_{kl\bar{d}} V_c^{kl\bar{d}} t_{ij\bar{a}}^c \\
\omega_k \mathbf{r}_3 &\leftarrow -\mathbf{r} \cdot \mathbf{X}_{31}^{hphh} - \frac{1}{2} \mathbf{r}_3 \cdot \mathbf{V}_{33}^{hhpp} \cdot \mathbf{t}_{31} \\
\omega_k \mathbf{r}_{21(2)} &\leftarrow \pm \mathbf{r}_{22} \cdot \mathbf{X}_3^{hh} \pm \mathbf{X}_{22}^{hphp} \cdot \mathbf{r}_{22} \\
\omega_k \mathbf{r}_1 &\leftarrow \mathbf{X}_3^{pp} \cdot \mathbf{r}_1 + \frac{1}{2} \mathbf{r}_1 \cdot \mathbf{X}_1^{hhhh}
\end{aligned} \tag{B.35}$$

\hat{L}_k^- , Matrix Form

Again, the disconnected term is computed as an outer product rather than with matrix-matrix multiplication.

$$\begin{aligned}
E_k l^i &= -X_k^i l^k - \frac{1}{2} X_{kl\bar{c}}^i l^{kl\bar{c}} \\
E_k \mathbf{l} &\leftarrow -\mathbf{X}_3^{HH} \cdot \mathbf{l} - \frac{1}{2} \mathbf{X}_{31}^{hphh} \cdot \mathbf{l}_3
\end{aligned} \tag{B.36}$$

$$\begin{aligned}
E_k l_a^{ij} &= \hat{P}(ij) l^i X_{a\bar{j}} - X_k^{ij\bar{a}} l^k + l_c^{ij} X_a^c - \hat{P}(ij) X_k^j l_{a\bar{i}}^k + \frac{1}{2} X_{kl}^{ij} l_a^{kl} \\
&\quad - \hat{P}(ij) l_{c\bar{k}}^j X_{a\bar{i}}^{c\bar{k}} - \frac{1}{2} V_c^{ij\bar{a}} t_{kl\bar{d}}^c l^{kl\bar{d}} \\
E_k l_3 &\leftarrow -\mathbf{X}_{33}^{hhhp} \cdot \mathbf{l} - \frac{1}{2} \mathbf{V}_{33}^{hhpp} \cdot \mathbf{t}_{31} \cdot \mathbf{l}_3 \\
E_k \mathbf{l}_{21(2)} &\leftarrow \pm \mathbf{l} \otimes \mathbf{X}_{2'}^{hp} \pm \mathbf{X}_3^{hh} \cdot \mathbf{l}_{22} \pm \mathbf{l}_{22} \cdot \mathbf{X}_{22}^{hphp} \\
E_k \mathbf{l}_1 &\leftarrow \mathbf{l}_1 \cdot \mathbf{X}_3^{pp} + \frac{1}{2} \mathbf{X}_1^{hhhh} \cdot \mathbf{l}_1
\end{aligned} \tag{B.37}$$

Appendix C

Angular Momentum Coupling

Before deriving useful equations for J-scheme angular momentum coupling, it's necessary to list some shorthand notations and definitions:

$$\hat{p} \equiv \sqrt{2j_p + 1} \quad (\text{C.1})$$

$$\sum_{\{m\}} \equiv \text{sum over all } m \quad (\text{C.2})$$

Clebsch-Gordan coefficients:

$$\langle pq|J\rangle \equiv \langle j_p m_p; j_q m_q | JM \rangle \quad (\text{C.3})$$

$$\langle p\bar{q}|J\rangle \equiv \langle j_p, m_p; j_q, -m_q | JM \rangle (-1)^{(q-m_q)} \quad (\text{C.4})$$

$$\sum_{JM} \langle pq|J\rangle \langle p'q'|J\rangle = \delta_{m_p m_{p'}} \delta_{m_q m_{q'}} \quad (\text{C.5})$$

$$\sum_{m_p m_q} \langle pq|J\rangle \langle pq|J'\rangle = \delta_{JJ'} \delta_{MM'} \quad (\text{C.6})$$

Two-body J-scheme matrix elements in terms of M-scheme matrix elements:

$$X_{rsJ}^{pqJ} = \sum_{\{m\}} X_{rmrsm_s}^{pmpqmq} \langle pq|J\rangle \langle rs|J\rangle \quad (\text{C.7})$$

$$X_{r\bar{q}J'}^{p\bar{s}J'} = \sum_{\{m\}} X_{rmrsm_s}^{pmpqmq} \langle p\bar{s}|J'\rangle \langle r\bar{q}|J'\rangle \quad (\text{C.8})$$

$$X_{rsJ\bar{q}}^p = \sum_{\{m\}} X_{rmrsm_s}^{pmpqmq} \langle rs|J\rangle \langle J\bar{q}|p\rangle \quad (\text{C.9})$$

$$X_r^{pqJ\bar{s}} = \sum_{\{m\}} X_{rmrsm_s}^{pmpqmq} \langle pq|J\rangle \langle J\bar{s}|r\rangle \quad (\text{C.10})$$

$$(\text{C.11})$$

Two-body M-scheme matrix elements in terms of J-scheme matrix elements:

$$X_{rmrsm_s}^{pmpqmq} = \sum_{JM} X_{rsJ}^{pqJ} \langle pq|J\rangle \langle rs|J\rangle \quad (\text{C.12})$$

$$= \sum_{J'M'} X_{r\bar{q}J'}^{p\bar{s}J'} \langle p\bar{s}|J'\rangle \langle r\bar{q}|J'\rangle \quad (\text{C.13})$$

$$= \sum_{JM} X_{rsJ\bar{q}}^p \langle rs|J\rangle \langle J\bar{q}|p\rangle \quad (\text{C.14})$$

$$= \sum_{JM} X_r^{pqJ\bar{s}} \langle pq|J\rangle \langle J\bar{s}|r\rangle \quad (\text{C.15})$$

$$(\text{C.16})$$

$${^\lambda R_i^{abJ}} = {^\lambda \mathbb{R}_i^{abJ}} \frac{\langle \bar{Ji} | \lambda \rangle}{\hat{\lambda}} (-1)^{2\lambda} \quad (\text{C.17})$$

$${}^{\lambda}\mathbb{R}_i^{abJ} = \sum_{J'} {}^{\lambda}\mathbb{R}_{i\bar{b}J'\hat{J}\hat{J}'}^a \begin{Bmatrix} a & b & J \\ i & \lambda & J' \end{Bmatrix} (-1)^{\lambda-a+J'} \quad (\text{C.18})$$

$${}^{\lambda}\mathbb{R}_{ijJ}^a = \sum_{J'} {}^{\lambda}\mathbb{R}_i^{a\bar{j}J'} \begin{Bmatrix} i & j & J \\ a & \lambda & J' \end{Bmatrix} (-1)^{\lambda+a-J} \quad (\text{C.19})$$

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