Title
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Publication Date
2012

Peer reviewed|Thesis/dissertation

Use of Effective Theories in Nuclear Physics

by

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A dissertation submitted in partial satisfaction of the
requirements for the degree of
Doctor of Philosophy

in

Physics

in the

GRADUATE DIVISION
of the
UNIVERSITY OF CALIFORNIA, BERKELEY

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Spring 2012

Abstract

Use of Effective Theories in Nuclear Physics

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   Approximations are inevitable in solving realistic physics problems, and reliability of calculations depends on evaluation of how much error is associated with the approximations. One method to quantify errors is building effective theories, which organize successive approximations as a power series in some small parameter. We apply effective theories to two problems in nuclear physics.

   One is the calculation of atomic electric dipole moments (EDMs). EDMs are of interest as a probe of CP-violating physics. For atoms, EDM signals can be thought of as departures from Schiff theorem, which states that a neutral system of point-like, nonrelativistic charges that interact only electrostatically has no net EDM. We show how each of the conditions for Schiff theorem are violated in actual atoms by expanding the Breit interaction between the electrons and the nucleus in spherical multipoles. We see that EDM signals arising from violations of the Schiff limit can be organized as a power series in R_N/R_A, the ratio between the spatial sizes of the nucleus and the atom. This ratio is of order 10^{-5}, and the power series in this parameter would have quantifiable errors. We identify the contributions to atomic EDM that correspond to the so-called Schiff moment, and give the general considerations for other contributions that may be of the same order as the Schiff moment in powers of R_N/R_A.

   The other problem is nucleon-nucleon (NN) interaction. The difficulty in describing this basic interaction is the fine tuning that exists between the long-range attraction and short-range repulsion in the NN potential. An effective theory of NN interaction must separate these two length scales. In order to achieve this separation, we introduce a harmonic oscillator (HO) basis, and restrict the calculation to a finite Hilbert space (P-space) of states with energies below some cutoff $\Lambda\hbar\omega$. HO eigenstates contains a length scale, the oscillator length b, which we choose to be 1.7fm, as an intermediate scale. We show that, despite the short-range nature of HO states, restricted wavefunctions contain enough information to reconstruct phase shifts. Projecting wavefunctions into this space throws away both the long-range physics due to the kinetic energy, and the short-range physics due to the strong interactions. We derive an equation in the P-space whose solution is the P-space restriction of the full-space scattering wavefunction, and identify the components of the equation that

treat the long-range and short-range physics, respectively. The long-range information is encoded in what we call the tilde states, which are modification to the HO wavefunction with slower decay as $r \to \infty$. The short-range information is modeled by a contact-gradient expansion, which is essentially a power series in a/b, where a is the length scale associated with the repulsive core in the NN potential. The behavior of the theory is investigated using a toy model of a spherical square well with a repulsive core.

Contents

Acknowledgments

Chapter 1

Atomic Electric Dipole Moment

1.1 Introduction

Ever since the first attempt by Purcell and Ramsey [1], physicists have made successively more precise measurements of electric dipole moments (EDMs). Although EDMs have never been observed in fundamental particles, nucleons, and atoms, this lack of signal has acted as tests of particle physics models. EDM experiments, like other precision measurements, are places where nuclear and atomic experimentalists can make contributions to the search for physics beyond the Standard Model (SM); precision measurements and collider experiments, being sensitive to different aspects of new physics, act as complementary pieces in this quest. Recent developments in cosmology suggest that there are beyond-SM sources of CP violation (CPV), which may be observed in EDM experiments; this acts as added motivation for the experimental effort.

EDM for a non-degenerate system violates CP symmetry. Such a system can be characterized by only one vector—its spin. So its intrinsic EDM, if it exists, must be either aligned or anti-aligned with the spin, and results in a Lagrantian term $d\vec{\sigma} \cdot \vec{E}$, where $\vec{\sigma}$ is the spin of the particle, and $\vec{E}$ is an external electric field. Under time reversal, $\vec{\sigma} \to -\vec{\sigma}$, while $\vec{E}$ is left invariant. Thus, the EDM term violates T symmetry, and by CPT theorem, this implies that CP is also violated. Since σ is an axial vector and E is a polar vector, the EDM term violates P as well.

CP violation has been observed in neutral kaon and B meson systems, and the Standard Model explains this by a phase in the Cabibbo-Kobayashi-Maskawa (CKM) matrix. Another potential source of CPV in the Standard Model is the so-called θ term in QCD. The most stringent limit on the parameter $\bar{\theta}$ comes from neutron EDM experiment and is $\bar{\theta} \lesssim 10^{-10}$. $\bar{\theta}$ can be written as a phase angle and, a priori, can take any value between $-\pi$ and π. Why this is so small is called the strong CP problem. One possible resolution is that the spontaneous breaking of Peccei-Quinn symmetry forces $\bar{\theta}$ to be 0 [2]. If this is correct, then there exist axions, Goldstone bosons corresponding to the broken symmetry [3]. As axions are candidates to be (part of) the dark matter that make up the majority of the matter content of the universe, probing CPV in the strong sector may provide information

for cosmology.

A more intimate connection between CPV and cosmology is the baryon asymmetry of the universe (BAU). From cosmic microwave background (CMB), distribution of galaxies, and supernova measurements of the Hubble constant, the latest estimate of the baryon-to-photon ratio in the energy density of the universe is [4]

$$\eta = (6.19 \pm 0.15) \times 10^{-10}. \tag{1.1}$$

In the standard cosmological picture, this nonzero value cannot be explained simply as the result of some initial condition in the universe, because any initial baryon number density would be vastly diluted by inflation. What is required is a mechanism, called baryogenesis, to produce the BAU after inflation. As Sakharov noted [5], baryogenesis requires (1) baryon number violation, (2) C and CP violation, and (3) non-equilibrium dynamics. The first two conditions are needed to preferentially produce baryons over antibaryons, and the third condition prevents "washout"—in equilibrium, particles with the same mass would end up with the same density. Although the SM does satisfy all three conditions, it is thought that SM processes are insufficient to account for the observed asymmetry, and various models involving physics beyond the SM have been proposed (See [6] for reviews). One class of baryogenesis models, called electroweak (EW) baryogenesis, is the most accessible to experiments. EW baryogenesis posits that baryogenesis occurred during the EW symmetry breaking era, i.e. the new physics for baryogenesis appears at the TeV scale. This is the energy scale that EDM experiments are starting to probe.

Atomic EDMs are intriguing from a number of perspectives. One is that atoms involve electrons as well as hadrons, meaning different sources of CPV can contribute to their EDM. In general, paramagnetic atoms are more sensitive to electron EDMs, whereas diamagnetic atoms are more sensitive to CPV inside the nucleus. Since multiple CPV sources can contribute to the EDM of a particular atom, measurements on multiple atomic species are needed to disentangle each contribution.

One obstacle to measuring atomic EDMs is the shielding effect identified by Schiff [7]. His theorem states that an electrically neutral system cannot have a net EDM if its constituents are non-relativistic and point-like, and interact only electrostatically. The Schiff theorem can be understood at the classical level. A neutral system in an external electric field $\vec{E}_{\text{ext}}$ does not accelerate. This means that at the location of each of the charged particles in the system, the net electric field is 0, i.e. other charges produce an electric field that exactly cancels out the external field. Changing $\vec{E}_{\text{ext}}$ does not alter the situation; charges will be rearranged so that the net field at the location of each of the charged particles will again be 0. Since no particle in the system is experiencing a net electric field, there cannot be any energy shift due to constituent EDMs.

Fortunately, all 3 conditions for the Schiff theorem are only approximately true in atoms. The conditions are evaded as follows: (a) atomic electrons, especially in heavy atoms, cannot be described non-relativistically, (b) the nucleus has finite size, (c) and the constituents have non-electrostatic interactions. The effects of relativistic electrons are especially strong in heavy paramagnetic atoms. One example is ^{205}Tl, whose EDM was interpreted as one of the

best limits on electron EDM [8]. In experiments on diamagnetic atoms, on the other hand, the effects of corrections to (b) and (c) can be more important. Traditionally, CPV effects in the nucleus that are not screened by the electrons have been quantified by the Schiff moment, which is the dipole component of the finite-size correction (e.g. [7, 12]). Following Liu et al. [9], we take a more general approach to the corrections and show that there are other comparable contributions to atomic EDM from nuclear CPV sources, such as those from M_2 and C_3 moments, although these particular contributions vanish for atoms with small total angular momentum or nuclear spin. The smallest current limit for an atomic EDM, from ^{199}Hg, sets limits on various CPV effects in the nucleus [10]. Planned EDM experiments include paramagnetic ^{211}Fr and diamagnetic ^{221}Rn, ^{223}Rn, and ^{225}Ra [11]. The last there are interesting due to the possibility that octupole deformation in the nucleus, which enhances the Schiff moment [12]. The goal for theory is to clarify how a CPV source translates to atomic EDMs, and quantify the errors in the calculation so that experimental results can be evaluated in terms of CPV parameters in the underlying physics.

Our approach to the problem takes the following steps:

1. We express the electron-nucleus interaction using the Breit interaction [13], which is the Coulomb interaction with relativistic corrections, up to $\mathcal{O}(\alpha^2)$. This takes account of the relativistic motion of the electrons as well as the non-electrostatic interactions in the atom.

2. The Breit interaction is separated into 2 parts; one part corresponds to the electron-nucleus interaction in the limit of a point nucleus, and the other part is the correction due to the fact that electrons can penetrate inside the nucleus. This takes account of the finite size of the nucleus.

3. We then expand the interaction in terms of electronic and nuclear multipole operators. Multipole operators describe the charge and current distributions of electrons and the nucleus, and carry definite angular momentum. We see that the multipoles appear with leading factors of the form,

$$\frac{4\pi\alpha}{R_A}\left(\frac{R_N}{R_A}\right)^n \tag{1.2}$$

for some integer n, where R_N is the typical nuclear size, and R_A is the typical atomic size. Since R_N/R_A is of order 10^{-5}, this ratio acts as a small parameter to organize the calculation.

4. The total atomic Hamiltonian H is separated into the unperturbed Hamiltonian H_0 and perturbation V'. We will choose H_0 so that its eigenstate can be written as a tensor product of an electronic wavefunction and a nuclear wavefunction, each having been calculated with no consideration for backreaction.

5. Perturbation theory gives the ground state of the system, $|$g.s. $>$, which is written as

$$|\text{g.s.} >= |0> +|1> +|2> +\ldots, \tag{1.3}$$

where $|0>$ is the unperturbed ground state, and $|n>$ denotes corrections to $|0>$ with n insertions of the perturbation V'. The atomic EDM signal is proportional to

$$< \text{g.s.}|\vec{d}|\text{g.s.} >=< \text{g.s.}|\vec{d_e}|\text{g.s.} > + < \text{g.s.}|\vec{d_N}|\text{g.s.} > \qquad (1.4)$$

where $\vec{d_e}$ and $\vec{d_N}$ are the electronic and nuclear dipole operators.

6. Schiff cancellation is effected by the use of displacement operator, A. The expected leading order (LO) contributions to the EDM from the nuclear dipole operator is seen to cancel out against part of the next-to-leading (NLO) contribution from the electronic dipole operator. To be more specific, polarization of the electron cloud due to the C_1-C_1 interaction between the electron-nucleus interaction produces a signal that is equal and opposite to that of a nuclear EDM. Since the cancellation is demonstrated in terms of operators, we can show that analogous cancellations also take place at higher orders in the perturbative expansion.

7. With Schiff cancellation in place, a general expression for the EDM signal can be given. It is important to ensure that the natural size of the contribution is at least of the same order as the Schiff moment (3 powers of R_N/R_A), and that the term has the right symmetry properties to be an EDM. Up to this point, no reference to a specific atom has been made.

8. Finally, the expression is evaluated by introducing atomic and nuclear wavefunctions.

The rest of this chapter is organized in accordance with these steps. In section 1.2, we define the Hamiltonian, make the separation between the point-nucleus limit and the penetration correction in the electron-nucleus interaction, then expand in terms of spherical multipole operators. In section 1.3, the perturbative scheme is set up, and the relation between matrix elements in the perturbation theory to the atomic EDM signal is given. Section 1.4 demonstrates Schiff cancellation using the displacement operator. In section 1.5 we give the answer that is leading order in perturbation, after discussing how symmetry arguments eliminate many multipoles from contributing.

1.2 The Hamiltonian

The Hamiltonian that we consider for the atom is

$$H = T_e + T_N + V_{ee} + V_{eN} + V_{e\tilde{N}} + V_{NN} + V_{N\tilde{N}}. \qquad (1.5)$$

Subscripts e and N represent electrons and the nucleus, respectively, and $\tilde{N}$ indicates that the interaction with the nucleus violates CP. For our calculation, all of the CPV physics is assumed to come from the nucleus.

Looking at the terms of H in more detail,

$$T_e = -\sum_{i=1}^{Z} \frac{1}{2m_e} \vec{\nabla}_i^2 \tag{1.6}$$

is the total kinetic energy of the atomic electrons, whose coordinates are given by $\vec{x}_i$, with i running from 1 to Z.

$$T_N = -\sum_{i=1}^{A} \frac{1}{2m_N} \vec{\nabla}_{N,i}^2 \tag{1.7}$$

is the kinetic energy of the nucleons.

V_{ee} is the interaction among the electrons, and we write

$$H_e = T_e + V_{ee} - Z\alpha \int d^3x \frac{\rho_e(\vec{x})}{x} \tag{1.8}$$

as the electronic Hamiltonian, taking into account the Coulomb interaction between the point nucleus and the electrons and the two-body interactions among the electrons. Similarly, $H_{NN} + H_{N\tilde{N}}$ is the interaction among the nucleons, and

$$H_N = T_N + V_{NN} + V_{N\tilde{N}} \tag{1.9}$$

is the nuclear Hamiltonian, solved in nuclear structure calculations.

The Breit interaction gives the electron-nucleus interaction in our calculation,

$$V_{eN} + V_{e\tilde{N}} = -\alpha \iint d^3x d^3y \left[\frac{\rho_e(\vec{x})\rho_N(\vec{y})}{|\vec{x}-\vec{y}|} \right.$$
$$\left. -\frac{1}{2} \left(\frac{\vec{j}_e(\vec{x}) \odot \vec{j}_N(\vec{y})}{|\vec{x}-\vec{y}|} + \frac{\vec{j}_e(\vec{x}) \odot (\vec{x}-\vec{y})\, \vec{j}_N(\vec{y}) \odot (\vec{x}-\vec{y})}{|\vec{x}-\vec{y}|^3} \right) \right]. \tag{1.10}$$

ρ_e and ρ_N are the electronic and nuclear charge densities, and $\vec{j}_e$ and $\vec{j}_N$ are the electronic and nuclear current densities (densities for the electrons are defined with a minus sign to take care of the negative charge). The first line is the Coulomb interaction between the nucleus and the electrons, and the second line gives the magnetic and transverse electric interactions due to electromagnetic currents. We use the angular momentum conventions of Varshalovich [14]. Scalar product $\odot$ is defined as

$$A_l \odot B_l \equiv \sum_m (-1)^m A_{lm} B_{l,-m}. \tag{1.11}$$

The Breit interaction can be separated into the electron-nucleus interaction in the point-nucleus limit, and corrections due to electrons penetrating the nucleus (see appendices A.1 and A.2 for the detailed calculation).

$$V_{eN} + V_{e\tilde{N}} = V_{eN}^{\text{point}} + V_{eN}^{\text{pen}}. \tag{1.12}$$

Expanding V_{eN}^{point} and V_{eN}^{pen} in electronic and nuclear multipoles,

$$V_{eN}^{\text{point}} = -\frac{4\pi\alpha}{R_A}\left[\sum_{l=0}^{\infty}\frac{1}{2l+1}\left(\frac{R_N}{R_A}\right)^l C_l^A \odot C_l^N\right.$$
$$+\sum_{l=1}^{\infty}\left\{\frac{1}{2l+1}\left(\frac{R_N}{R_A}\right)^l T_{\text{mag},l}^A \odot T_{\text{mag},l}^N\right.$$
$$+\frac{1}{2l+1}\left(\frac{R_N}{R_A}\right)^{l+1} T_{\text{el},l}^A \odot T_{\text{el},l}'^N$$
$$+\left.\left.\frac{1}{2l+1}\left(\frac{R_N}{R_A}\right)^{l-1} T_{\text{el},l}'^A \odot T_{\text{el},l}^N\right\}\right]. \tag{1.13}$$

and

$$V_{eN}^{\text{pen}} = -\frac{4\pi\alpha}{R_A}\left(\sum_{l=0}^{\infty}\frac{1}{2l+1}\sum_{i=1}^{Z}\left[C_l^{A+}(i) \odot C_l^{N-}(i) - C_l^{A-}(i) \odot C_l^{N+}(i)\right]\right.$$
$$+\sum_{l=1}^{\infty}\left\{\frac{1}{2l+1}\sum_{i=1}^{Z}\left[T_{\text{mag},l}^{A+}(i) \odot T_{\text{mag},l}^{N-}(i) - T_{\text{mag},l}^{A-}(i) \odot T_{\text{mag},l}^{N+}(i)\right]\right.$$
$$+\frac{1}{2l+1}\sum_{i=1}^{Z}\left[T_{\text{el},l}^{A+}(i) \odot T_{\text{el},l}'^{N-}(i) - T_{\text{el},l}^{A-}(i) \odot T_{\text{el},l}'^{N+}(i)\right]$$
$$+\left.\left.\frac{1}{2l+1}\sum_{i=1}^{Z}\left[T_{\text{el},l}'^{A+}(i) \odot T_{\text{el},l}^{N-}(i) - T_{\text{el},l}'^{A-}(i) \odot T_{\text{el},l}^{N+}(i)\right]\right\}\right). \tag{1.14}$$

The multipole operators in the point-nucleus term are the charge multipoles

$$C_{lm}^N \equiv \int d^3y \left(\frac{y}{R_N}\right)^l Y_{lm}(\Omega_y)\rho_N(\vec{y})$$
$$C_{lm}^A \equiv \int d^3x \left(\frac{R_A}{x}\right)^{l+1} Y_{lm}(\Omega_x)\rho_e(\vec{x}) = \sum_{i=1}^{Z}\left(\frac{R_A}{x_i}\right)^{l+1} Y_{lm}(\Omega_i), \tag{1.15}$$

the transverse magnetic multipoles

$$T_{\text{mag},l}^N \equiv \frac{1}{R_N^l}\int d^3y\, y^l \vec{\mathcal{Y}}_{ll1}^m(\Omega_y) \cdot \vec{j}_N(\vec{y}) = \int d^3y \left(\frac{y}{R_N}\right)^l \left[Y_l(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l$$
$$T_{\text{mag},l}^A \equiv R_A^{l+1}\int d^3x\, \frac{1}{x^{l+1}}\vec{\mathcal{Y}}_{ll1}^m(\Omega_x) \cdot \vec{j}_e(\vec{x}) = \sum_{i=1}^{Z}\left(\frac{R_A}{x_i}\right)^{l+1} \left[Y_l(\Omega_i) \otimes \vec{\alpha}_i(\vec{x}_i)\right]_l, \tag{1.16}$$

and the transverse electric multipoles

$$T_{\mathrm{el},l}^{N} \equiv \frac{1}{R_N^{l-1}} \int d^3y \left[\vec{\nabla} \times \left(y^l \vec{\mathcal{Y}}_{ll1}^m(\Omega_y) \right) \right] \cdot \vec{j}_N(\vec{y})$$

$$= i\sqrt{(l+1)(2l+1)} \int d^3y \left(\frac{y}{R_N} \right)^{l-1} \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N(\vec{y}) \right]_l$$

$$T_{\mathrm{el},l}'^{N} \equiv \frac{1}{R_N^{l+1}} \int d^3y \left[\vec{\nabla} \times \left(-\frac{y^{l+2}}{2(2l+3)} \vec{\mathcal{Y}}_{ll1}^m(\Omega_y) \right) \right] \cdot \vec{j}_N(\vec{y})$$

$$= -i \left\{ \frac{1}{2l+3} \sqrt{\frac{l}{2l+1}} \int d^3y \left(\frac{y}{R_N} \right)^{l+1} \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N(\vec{y}) \right]_l \right.$$

$$\left. + \frac{1}{2} \sqrt{\frac{l+1}{2l+1}} \int d^3y \left(\frac{y}{R_N} \right)^{l+1} \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N(\vec{y}) \right]_l \right\}$$

$$T_{\mathrm{el},l}^{A} \equiv R_A^{l+2} \int d^3x \left[\vec{\nabla} \times \left(\frac{1}{x^{l+1}} \vec{\mathcal{Y}}_{ll1}^m(\Omega_x) \right) \right] \cdot \vec{j}_e(\vec{x})$$

$$= -i\sqrt{l(2l+1)} \sum_{i=1}^{Z} \left(\frac{R_A}{x_i} \right)^{l+2} [Y_{l+1}(\Omega_i) \otimes \vec{\alpha}_i(\vec{x}_i)]_l$$

$$T_{\mathrm{el},l}'^{A} \equiv R_A^{l} \int d^3x \left[\vec{\nabla} \times \left(\frac{1}{2(2l-1)} \vec{\mathcal{Y}}_{ll1}^m(\Omega_x) \right) \right] \cdot \vec{j}_e(\vec{x})$$

$$= i \left\{ -\frac{1}{2} \sqrt{\frac{l}{2l+1}} \sum_{i=1}^{Z} \left(\frac{R_A}{x_i} \right)^l [Y_{l+1}(\Omega_i) \otimes \vec{\alpha}_i(\vec{x}_i)]_l \right.$$

$$\left. + \frac{1}{2l-1} \sqrt{\frac{l+1}{2l+1}} \sum_{i=1}^{Z} \left(\frac{R_A}{x_i} \right)^l [Y_{l-1}(\Omega_i) \otimes \vec{\alpha}_i(\vec{x}_i)]_l \right\}. \tag{1.17}$$

The multipoles that appear in V_{eN}^{pen} are

$$C_l^{A+}(i) \odot C_l^{N-}(i) = \left(\frac{x_i}{R_N} \right)^l Y_l(\Omega_i) \odot \int d^3y\, \theta(y-x_i) \left(\frac{R_N}{y} \right)^{l+1} Y_l(\Omega_y) \rho_N(\vec{y})$$

$$C_l^{A-}(i) \odot C_l^{N+}(i) = \left(\frac{R_N}{x_i} \right)^{l+1} Y_l(\Omega_i) \odot \int d^3y\, \theta(y-x_i) \left(\frac{y}{R_N} \right)^l Y_l(\Omega_y) \rho_N(\vec{y}) \tag{1.18}$$

$$T_{\text{mag},l}^{A+}(i) \odot T_{\text{mag},l}^{N-}(i) = \left(\frac{x_i}{R_N}\right)^l \left[Y_l(\Omega_i) \otimes \vec{\alpha}_i\right]_l$$

$$\odot \int d^3y \, \theta(y - x_i) \left(\frac{R_N}{y}\right)^{l+1} \left[Y_l(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l$$

$$T_{\text{mag},l}^{A-}(i) \odot T_{\text{mag},l}^{N+}(i) = \left(\frac{R_N}{x_i}\right)^{l+1} \left[Y_l(\Omega_i) \otimes \vec{\alpha}_i\right]_l$$

$$\odot \int d^3y \, \theta(y - x_i) \left(\frac{y}{R_N}\right)^l \left[Y_l(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l$$

$$T_{\text{el},l}^{A+}(i) \odot T_{\text{el},l}^{'N-}(i) = i\sqrt{(l+1)(2l+1)} \left(\frac{x_i}{R_N}\right)^{l-1} \left[Y_{l-1}(\Omega_i) \otimes \vec{\alpha}_i\right]_l$$

$$\odot \, i \left\{ -\frac{1}{2}\sqrt{\frac{l}{2l+1}} \int d^3y \, \theta(y - x_i) \left(\frac{R_N}{y}\right)^l \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l \right.$$

$$\left. + \frac{1}{2l-1}\sqrt{\frac{l+1}{2l+1}} \int d^3y \, \theta(y - x_i) \left(\frac{R_N}{y}\right)^l \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l \right\}$$

$$T_{\text{el},l}^{A-}(i) \odot T_{\text{el},l}^{'N+}(i) = -i\sqrt{l(2l+1)} \left(\frac{R_N}{x_i}\right)^{l+2} \left[Y_{l+1}(\Omega_i) \otimes \vec{\alpha}_i\right]_l$$

$$\odot \, (-i) \left\{ \frac{1}{2l+3}\sqrt{\frac{l}{2l+1}} \int d^3y \, \theta(y - x_i) \left(\frac{y}{R_N}\right)^{l+1} \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l \right.$$

$$\left. + \frac{1}{2}\sqrt{\frac{l+1}{2l+1}} \int d^3y \, \theta(y - x_i) \left(\frac{y}{R_N}\right)^{l+1} \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l \right\}$$

$$T'^{A+}_{\text{el},l}(i) \odot T^{N-}_{\text{el},l}(i) = - i \left\{ \frac{1}{2l+3} \sqrt{\frac{l}{2l+1}} \left(\frac{x_i}{R_N}\right)^{l+1} [Y_{l+1}(\Omega_i) \otimes \vec{\alpha}_i]_l \right.$$

$$\left. + \frac{1}{2}\sqrt{\frac{l+1}{2l+1}} \left(\frac{x_i}{R_N}\right)^{l+1} [Y_{l-1}(\Omega_i) \otimes \vec{\alpha}_i]_l \right\}$$

$$\odot (-i)\sqrt{l(2l+1)} \int d^3y\, \theta(y-x_i) \left(\frac{R_N}{y}\right)^{l+2} \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l$$

$$T'^{A-}_{\text{el},l}(i) \odot T^{N+}_{\text{el},l}(i) = i \left\{ \frac{1}{2}\sqrt{\frac{l}{2l+1}} \left(\frac{R_N}{x_i}\right)^{l} [Y_{l+1}(\Omega_i) \otimes \vec{\alpha}_i]_l \right.$$

$$\left. + \frac{1}{2l-1}\sqrt{\frac{l+1}{2l+1}} \left(\frac{R_N}{x_i}\right)^{l} [Y_{l-1}(\Omega_i) \otimes \vec{\alpha}_i]_l \right\}$$

$$\odot\, i\sqrt{(l+1)(2l+1)} \int d^3y\, \theta(y-x_i) \left(\frac{y}{R_N}\right)^{l-1} \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l .$$

$$(1.19)$$

Note that the electron coordinates are measured in atomic size R_A, and the nuclear coordinates are measured in nuclear size R_N. Since these length scales have been divided out, one expects the integrals to be of order one. This implies that the leading factors of

$$\frac{4\pi\alpha}{R_A} \left(\frac{R_N}{R_A}\right)^n \tag{1.20}$$

in all these terms correspond to the natural sizes of each operator. Since R_N/R_A is of order 10^{-5}, we use the power of this small number to organize our calculation. Terms with $n \leq 3$ are retained, as that is the order at which the Schiff moment enters. Matrix elements of penetration multipole operators between atomic states can be reproduced by effective operators that have natural sizes. See Appendix A.2 for these effective operators.

1.3 Perturbation Theory

We now set up the perturbation theory by writing the Hamiltonian in 2 parts:

$$H = H_0 + V', \tag{1.21}$$

where

$$H_0 = H_e + H_N. \tag{1.22}$$

The electronic Hamiltonian H_e and the nuclear Hamiltonian H_N were defined in the previous section. The eigenvalue problem for H_0 results in the unperturbed states, which have the form

$$|\alpha_e j_e m_e; \alpha_N j_N m_N> = |\alpha_e j_e m_e>_e \otimes |\alpha_N j_N m_N>_N . \tag{1.23}$$

$|\alpha_e j_e m_e >_e$ and $|\alpha_N j_N m_N >_N$ are eigenstates of H_e and H_N, respectively, with spin quantum numbers specified by j_i and m_i. α_e and α_N represent all other quantum numbers that need to be specified.

Here, note that the last term in the definition of H_e, eq. (1.8), can be rewritten in terms of multipoles as

$$Z\alpha \int d^3x \frac{\rho_e(\vec{x})}{x} = \frac{4\pi\alpha}{R_A} C_0^A \odot < C_0^N >, \tag{1.24}$$

where the expectation value $< C_0^N >$ is taken between the nuclear ground state.

With H_0 specified, the perturbation becomes

$$\begin{aligned} V' &= V_{eN} + V_{e\tilde{N}} + \frac{4\pi\alpha}{R_A} C_0^A \odot < C_0^N > \\ &= V_{eN}'^{\text{point}} + V_{eN}^{\text{pen}}, \end{aligned} \tag{1.25}$$

with the adjustment to the point-nucleus term

$$\begin{aligned} V_{eN}'^{\text{point}} = -\frac{4\pi\alpha}{R_A} &\left[C_0^A \odot \left(C_0^N - < C_0^N > \right) + \sum_{l=1}^{\infty} \frac{1}{2l+1} \left(\frac{R_N}{R_A} \right)^l C_l^A \odot C_l^N \right. \\ &+ \sum_{l=1}^{\infty} \left\{ \frac{1}{2l+1} \left(\frac{R_N}{R_A} \right)^l T_{\text{mag},l}^A \odot T_{\text{mag},l}^N \right. \\ &\left. \left. + \frac{1}{2l+1} \left(\frac{R_N}{R_A} \right)^{l+1} T_{\text{el},l}^A \odot T_{\text{el},l}'^N + \frac{1}{2l+1} \left(\frac{R_N}{R_A} \right)^{l-1} T_{\text{el},l}'^A \odot T_{\text{el},l}^N \right\} \right]. \end{aligned} \tag{1.26}$$

This expression makes clear that nuclear axial charge can contribute to the EDM through $(C_0^N - < C_0^N >)$.

From appendix A.3, the ground state of H is

$$|\text{g.s.} >= |0 > + |1 > + |2 > + |3 > + \ldots, \tag{1.27}$$

where $|0 >$ is the ground state of H_0, and $|n >$ are the perturbative corrections with n insertions of V'.

Explicitly,

$$|1> = \frac{Q_0}{E_0 - H_0} V'|0>$$

$$|2> = \frac{Q_0}{E_0 - H_0} V' \frac{Q_0}{E_0 - H_0} V'|0> - \frac{Q_0}{(E_0 - H_0)^2} V'|0><0|V'|0>$$

$$\begin{aligned}|3> =\ & \frac{Q_0}{E_0 - H_0} V' \frac{Q_0}{E_0 - H_0} V' \frac{Q_0}{E_0 - H_0} V'|0> \\
& - \frac{Q_0}{E_0 - H_0} V' \frac{Q_0}{(E_0 - H_0)^2} V'|0><0|V'|0> \\
& - \frac{Q_0}{(E_0 - H_0)^2} V' \frac{Q_0}{E_0 - H_0} V'|0><0|V'|0> \\
& + \frac{Q_0}{(E_0 - H_0)^3} V'|0> (<0|V'|0>)^2 \\
& - \frac{Q_0}{(E_0 - H_0)^2} V'|0><0|V' \frac{Q_0}{E_0 - H_0} V'|0> \,.\end{aligned}\tag{1.28}$$

Here,

$$Q_0 \equiv \sum_{n \neq 0} |0><0| \tag{1.29}$$

is the projection operator out of the ground state of H_0.

The energy shift experienced by the atom when you turn on the external electric field $\vec{E}_{\text{ext}}$ is (from appendix A.4)

$$\Delta E = \frac{<FM_f|\vec{E}_{\text{ext}} \cdot \vec{F}|FM_i>}{\sqrt{F(F+1)(2F+1)}} <F||\vec{d}||F>, \tag{1.30}$$

where $\vec{F}$ is the total angular momentum of the atom, and

$$\vec{d} = \vec{d}_e + \vec{d}_N = \sum_{i=1}^{Z} \alpha \vec{x}_i - \sqrt{\frac{4\pi}{3}} \alpha R_N C_1^N \tag{1.31}$$

is the EDM operator for the atom. We see that the signal is proportional to the matrix element of $\vec{d}$ between the ground state of H, and this can be expanded perturbatively in the number of V' insertions as follows:

$$\begin{aligned}<\text{g.s.}|\vec{d}|\text{g.s.}> =\ & <0|\vec{d}|0> + \left(<0|\vec{d}|1> + <1|\vec{d}|0>\right) \\
& + \left(<0|\vec{d}|2> + <1|\vec{d}|1> + <2|\vec{d}|0>\right) + \dots. \end{aligned}\tag{1.32}$$

1.4   Schiff Cancellation

The natural size of $\vec{d}_N$ is R_N/R_A times that of $\vec{d}_e$. Hence, $< 0|\vec{d}_e|0 >$ is naively the LO contribution to the atomic EDM. Writing $|0>$ as the tensor product of atomic and nuclear ground states,

$$|0>=|0>_e \otimes|0>_N, \tag{1.33}$$

the ground state expectation value of $\vec{d}_e$ becomes

$$< 0|\vec{d}_e|0 >=< 0|\vec{d}_e|0 >_e \cdot < 0|0 >_N=< 0|\vec{d}_e|0 >_e, \tag{1.34}$$

Since H_e does not contain parity violation, this term vanishes (The term is odd under reversal of all electronic coordinates $\vec{x}_i \to -\vec{x}_i$. See discussion in subsection 1.5.2).

The next terms to be considered are the LO term from the nuclear EDM operator

$$< 0|\vec{d}_N|0 > \tag{1.35}$$

and the NLO terms from the electronic EDM operator

$$< 0|\vec{d}_e|1 > + < 1|\vec{d}_e|0 > . \tag{1.36}$$

It is convenient now to define the displacement operator,

$$A \equiv \frac{1}{Z}\sqrt{\frac{4\pi}{3}} \left(\frac{R_N}{R_A}\right) \sum_{i=1}^{Z} R_A \vec{\nabla}_i \odot C_1^N. \tag{1.37}$$

This operator has two very useful commutator relations. The first relation connects $\vec{d}_e$ and $\vec{d}_N$.

$$\left[\vec{d}_e, A\right] = \left[\sum_i^{Z} \vec{x}_i, \frac{1}{Z}\sqrt{\frac{4\pi}{3}} \left(\frac{R_N}{R_A}\right) \sum_{i=1}^{Z} R_A \vec{\nabla}_i \odot C_1^N\right]$$

$$= -\sqrt{\frac{4\pi}{3}} R_N C_1^N = \vec{d}_N. \tag{1.38}$$

The second relation involves the unperturbed Hamiltonian.

$$[A, H_0] = \frac{1}{Z}\sqrt{\frac{4\pi}{3}} \left(\frac{R_N}{R_A}\right) \left(\left[\sum_{i=1}^{Z} R_A \vec{\nabla}_i \odot C_1^N, H_N\right]\right.$$

$$+ \left[\sum_{i=1}^{Z} R_A \vec{\nabla}_i \odot C_1^N, \left(-\frac{1}{2m_e}\sum_{j=1}^{Z} \vec{\nabla}_j^2\right) + V_{ee}\right]$$

$$+ \left.\left[\sum_{i=1}^{Z} R_A \vec{\nabla}_i \odot C_1^N, -\frac{4\pi\alpha}{R_A}C_0^A \odot < C_0^N >\right]\right) \tag{1.39}$$

The first line will be left as $[A, H_N]$. The second line yields 0, as T_e and V_{ee} are invariant under overall translation of the electron cloud, and therefore commutes with $\sum_i \vec{\nabla}_i$. Finally, evaluating the third line,

$$
\begin{aligned}
&\frac{1}{Z}\sqrt{\frac{4\pi}{3}}\left(\frac{R_N}{R_A}\right)\left[\sum_{i=1}^{Z} R_A \vec{\nabla}_i \odot C_1^N, -\frac{4\pi\alpha}{R_A}C_0^A \odot < C_0^N >\right]\\
&=\frac{1}{Z}\sqrt{\frac{4\pi}{3}}\left(\frac{R_N}{R_A}\right)\times\left(-\frac{4\pi\alpha}{R_A}\right)\left(\frac{Z}{\sqrt{4\pi}}\right)\sum_m (-1)^m \sum_{i=1}^{Z} R_A \nabla_{im}\left[\frac{R_A}{x_i}Y_{00}\right]C_{1,-m}^N\\
&=-\frac{4\pi\alpha}{R_A}\frac{1}{\sqrt{3}}\left(\frac{R_N}{R_A}\right)\sum_m (-1)^m\left(-\frac{1}{\sqrt{3}}\right)C_{1m}^A C_{1,-m}^N\\
&=\frac{4\pi\alpha}{3R_A}\left(\frac{R_N}{R_A}\right)C_1^A \odot C_1^N.
\end{aligned}
\tag{1.40}
$$

This is (-1) times the C_1 term in the electron-nucleus interaction. Combining the results,

$$
[A, H_0] = [A, H_N] + \frac{4\pi\alpha}{3R_A}\left(\frac{R_N}{R_A}\right)C_1^A \odot C_1^N.
\tag{1.41}
$$

Using the form that is most useful, this lets us rewrite the interaction as

$$
V' = [A, E_0 - H_0] + \Delta V,
\tag{1.42}
$$

with

$$
\begin{aligned}
\Delta V \equiv &V' - \frac{4\pi\alpha}{3R_A}\left(\frac{R_N}{R_A}\right)C_1^A \odot C_1^N + [A, H_N]\\
=&-\frac{4\pi\alpha}{R_A}\left[C_0^A \odot (C_0^N - < C_0^N >) + \sum_{l=2}^{\infty}\frac{1}{2l+1}\left(\frac{R_N}{R_A}\right)^l C_l^A \odot C_l^N\right.\\
&+\sum_{l=1}^{\infty}\left\{\frac{1}{2l+1}\left(\frac{R_N}{R_A}\right)^l T_{\text{mag},l}^A \odot T_{\text{mag},l}^N\right.\\
&\left.\left.+\frac{1}{2l+1}\left(\frac{R_N}{R_A}\right)^{l+1}T_{\text{el},l}^A \odot T_{\text{el},l}'^N + \frac{1}{2l+1}\left(\frac{R_N}{R_A}\right)^{l-1}T_{\text{el},l}'^A \odot T_{\text{el},l}^N\right\}\right]\\
&+V_{eN}^{\text{pen}} + [A, H_N].
\end{aligned}
\tag{1.43}
$$

As the C_1 term is now moved to the commutator $[A, E_0 - H_0]$, the sum over the charge-charge interaction in ΔV starts at $l = 2$.

Going back to the NLO expression for the electronic dipole contribution,

$$
\begin{aligned}
&< 0|\vec{d_e}|1 > + < 1|\vec{d_e}|0 > \\
&= \left(< 0|\vec{d_e}\frac{Q_0}{E_0 - H_0}[A, E_0 - H_0]|0 > + < 0|\vec{d_e}\frac{Q_0}{E_0 - H_0}\Delta V|0 > \right) \\
&\quad + \left(< 0|[A, E_0 - H_0]\frac{Q_0}{E_0 - H_0}\vec{d_e}|0 > + < 0|\Delta V\frac{Q_0}{E_0 - H_0}\vec{d_e}|0 > \right) \\
&= - < 0|\vec{d_e}Q_0 A|0 > + < 0|A Q_0\vec{d_e}|0 > \\
&\quad + < 0|\vec{d_e}\frac{Q_0}{E_0 - H_0}\Delta V|0 > + < 0|\vec{d_e}\frac{Q_0}{E_0 - H_0}\Delta V|0 > \\
&= - < 0|[\vec{d_e}, A]|0 > + < 0|\vec{d_e}\frac{Q_0}{E_0 - H_0}\Delta V|0 > + < 0|\vec{d_e}\frac{Q_0}{E_0 - H_0}\Delta V|0 > .
\end{aligned} \tag{1.44}
$$

The first term exactly cancels out $< 0|\vec{d_N}|0 >$. We can now write the LO contribution to the atomic EDM as

$$
\begin{aligned}
d_{\mathrm{LO}} &\equiv < 0|\vec{d_N}|0 > + \left(< 0|\vec{d_e}|1 > + < 1|\vec{d_e}|0 > \right) \\
&= < 0|\vec{d_e}\frac{Q_0}{E_0 - H_0}\Delta V|0 > + < 0|\vec{d_e}\frac{Q_0}{E_0 - H_0}\Delta V|0 > .
\end{aligned} \tag{1.45}
$$

"LO" here is meant as the sum of LO terms involving $\vec{d_N}$ and NLO terms involving $\vec{d_e}$. For higher orders, "N^{n}LO" denotes the sum of N^{n}LO terms involving $\vec{d_N}$ and N^{n+1}LO terms involving $\vec{d_e}$.

1.5 Preliminary Results

1.5.1 General Expression

Through straightforward algebra, the next 2 orders in V' yield the following EDM expressions (to save space, $K = Q_0/(E_0 - H_0)$ is used):

$$
\begin{aligned}
d_{\mathrm{NLO}} &\equiv \left(< 0|\vec{d_N}|1 > + < 1|\vec{d_N}|0 > \right) \\
&\quad + \left(< 0|\vec{d_e}|2 > + < 1|\vec{d_e}|1 > + < 2|\vec{d_e}|0 > \right) \\
&= < 0|\vec{d_e}K\Delta V K\Delta V|0 > + < 0|\Delta V K\vec{d_e}K\Delta V|0 > + < 0|\Delta V K\Delta V K\vec{d_e}|0 > \\
&\quad - \left(< 0|\vec{d_e}K^2\Delta V|0 > + < 0|\Delta V K^2\vec{d_e}|0 > \right) < 0|\Delta V|0 > \\
&\quad + < 0|\vec{d_e}K[A, \Delta V]|0 > + < 0|[A, \Delta V]K\vec{d_e}|0 >
\end{aligned} \tag{1.46}
$$

and

$$d_{\text{NNLO}}$$
$$\equiv \Big(<0|\vec{d}_N|2> + <1|\vec{d}_N|1> + <2|\vec{d}_N|0> \Big)$$
$$+ \Big(<0|\vec{d}_e|3> + <1|\vec{d}_e|2> + <2|\vec{d}_e|1> + <3|\vec{d}_e|0> \Big)$$
$$= <0|\vec{d}_e K \Delta V K \Delta V K \Delta V|0> + <0|\Delta V K \vec{d}_e K \Delta V K \Delta V|0>$$
$$+ <0|\Delta V K \Delta V K \vec{d}_e K \Delta V|0> + <0|\Delta V K \Delta V K \Delta V K \vec{d}_e|0>$$
$$- \Big(<0|\vec{d}_e K \Delta V K^2 \Delta V|0> + <0|\Delta V K \vec{d}_e K^2 \Delta V|0> + <0|\Delta V K \Delta V K^2 \vec{d}_e|0>$$
$$+ <0|\vec{d}_e K^2 \Delta V K \Delta V|0> + <0|\Delta V K^2 \vec{d}_e K \Delta V|0> + <0|\Delta V K^2 \Delta V K \vec{d}_e|0> \Big)$$
$$\times <0|\Delta V|0>$$
$$- \Big(<0|\vec{d}_e K^2 \Delta V|0> + <0|\Delta V K^2 \vec{d}_e|0> \Big) <0|\Delta V K \Delta V|0>$$
$$+ \Big(<0|\vec{d}_e K^3 \Delta V|0> + <0|\Delta V K^3 \vec{d}_e|0> \Big) <0|\Delta V|0>^2$$
$$+ <0|\vec{d}_e K \Delta V K[A, \Delta V]|0> + <0|\vec{d}_e K[A, \Delta V]K \Delta V|0>$$
$$+ <0|\Delta V K \vec{d}_e K[A, \Delta V]|0> + <0|[A, \Delta V]K \vec{d}_e K \Delta V|0>$$
$$+ <0|\Delta V K[A, \Delta V]K \vec{d}_e|0> + <0|[A, \Delta V]K \Delta V K \vec{d}_e|0>$$
$$+ \Big(<0|\vec{d}_e K \Delta V|0> + <0|\Delta V K \vec{d}_e|0> \Big)$$
$$\times (<0|AK \Delta V|0> - <0|\Delta V KA|0>)$$
$$- \Big(<0|\vec{d}_e K^2[A, \Delta V]|0> + <0|[A, \Delta V]K^2 \vec{d}_e|0> \Big) <0|\Delta V|0>$$
$$+ (\text{cont.})$$

(cont.)

$$-\left(<0|\vec{d}_e K^2\Delta V|0> + <0|\Delta V K^2\vec{d}_e|0>\right)<0|[A,\Delta V]|0>$$

$$+\frac{1}{2}\left(<0|\vec{d}_e K\Delta V K[A,[A,E_0-H_0]]|0> + <0|\vec{d}_e K[A,[A,E_0-H_0]]K\Delta V|0>\right.$$

$$+<0|\Delta V K\vec{d}_e K[A,[A,E_0-H_0]]|0> + <0|[A,[A,E_0-H_0]]K\vec{d}_e K\Delta V|0>$$

$$\left. +<0|\Delta V K[A,[A,E_0-H_0]]K\vec{d}_e|0> + <0|[A,[A,E_0-H_0]]K\Delta V K\vec{d}_e|0>\right)$$

$$+\frac{1}{2}\left(<0|\vec{d}_e K[A,[A,\Delta V]]|0> + <0|[A,[A,\Delta V]]K\vec{d}_e|0>\right)$$

$$-\left(<0|\vec{d}_e K\Delta V|0> + <0|\Delta V K\vec{d}_e|0>\right)<0|A^2|0>$$

$$-\frac{1}{2}\left(<0|\vec{d}_e K^2\Delta V|0> + <0|\Delta V K^2\vec{d}_e|0>\right)<0|[A,[A,E_0-H_0]]|0>$$

$$-\frac{1}{2}\left(<0|\vec{d}_e K^2[A,[A,E_0-H_0]]|0> + <0|[A,[A,E_0-H_0]]K^2\vec{d}_e|0>\right)<0|\Delta V|0>$$

$$+\frac{1}{3}\left(<0|\vec{d}_e K[A,[A,[A,E_0-H_0]]]|0> + <0|[A,[A,[A,E_0-H_0]]]K\vec{d}_e|0>\right). \tag{1.47}$$

1.5.2 Symmetry Considerations

There are two symmetry arguments that eliminate many terms from contributing to the EDM. One is the parity operation on the electronic coordinates, and the other is time reversal.

Since we assume that H_e is parity invariant, its ground state, $|0>_e$, has definite parity. Multipole operators also have definite parity when all the electronic coordinates are reversed. An electronic matrix element is an integral of a function of $\vec{x}$ over all space, so if the integrand is odd under the transformation $\vec{x} \to -\vec{x}$, then the matrix element vanishes. For example, consider the case when the ground state is odd under parity. Then under $\vec{x}_i \to -\vec{x}_i$,

$$<0|C_l^A|0>_e \to\; <0|(-1)\cdot(-1)^l C_l^A\cdot(-1)|0>_e$$
$$=(-1)^l<0|C_l^A|0>_e. \tag{1.48}$$

Clearly, this result applies to even parity ground states as well. Since the symmetry transformation here is a symmetry of the theory, l in eq. (1.48) must be even for the term to be nonzero. More generally, for a term in the perturbation series to be nonzero, the total parity of the electronic multipole operators, including $\vec{d}_e$ must be even.

Time reversal requirement is slightly more involved. For an example of how a multipole operator transforms under T, consider C_{lm}^N. It is helpful to use de Forest and Walecka's convention for the charge multipole [15], which is related to C_{lm}^N as

$$C_{lm}'^N \equiv i^l C_{lm}^N. \tag{1.49}$$

Multipole Moment	P	T
C_l	$(-1)^l$	$(-1)^l$
$T_{\mathrm{mag},l}$	$(-1)^{l+1}$	$(-1)^{l+1}$
$T_{\mathrm{el},l}, T'_{\mathrm{el},l}$	$(-1)^l$	$(-1)^{l+1}$

Table 1.1: Phases acquired by multipoles under symmetry operations.

The action of time reversal operator T on the nuclear charge operator ρ_N is

$$T\rho_N(\vec{y})T^{-1} = (-1)^s \rho_N(\vec{y}), \tag{1.50}$$

where $s = 0$ if the charge conserves T, and $s = 1$ if it violates T (we assume $s = 0$ for the electronic charge operator). T operator also takes complex conjugates of any number. Using these properties,

$$
\begin{aligned}
TC'^N_{lm}T^{-1} &= Ti^lT^{-1}\int d^3y \left(\frac{y}{R_A}\right)^l TY_{lm}(\Omega_y)T^{-1}T\rho_N(\vec{y})T^{-1} \\
&= (-i)^l \int d^3y \left(\frac{y}{R_A}\right)^l (-1)^m Y_{l,-m}(\Omega_y)(-1)^s \rho_N(\vec{y}) \\
&= (-1)^{l+m+s} C'^N_{l,-m}.
\end{aligned}
\tag{1.51}
$$

Now, consider a reduced matrix element of C'^N_{lm}.

$$
\begin{aligned}
&< J_f ||C'^N_l|| J_i > \\
&= \left[(-1)^{J_f-M_f} \begin{pmatrix} J_f & l & J_i \\ -M_f & M & M_i \end{pmatrix}\right]^{-1} < J_f M_f | T^{-1}TC'^N_{lm}T^{-1}T | J_i M_i > \\
&= \left[(-1)^{J_f-M_f} \begin{pmatrix} J_f & l & J_i \\ -M_f & M & M_i \end{pmatrix}\right]^{-1} (-1)^{l+M+J_i+M_i+s} < J_f, -M_f | C'^N_{l,-m} | J_i, -M_i >^* \\
&= \left[(-1)^{J_f-M_f} \begin{pmatrix} J_f & l & J_i \\ -M_f & M & M_i \end{pmatrix}\right]^{-1} (-1)^{J_i+M_i+s} < J_i, -M_i | C'^N_{lm} | J_f, -M_f > \\
&= (-1)^{J_i-J_f+l+s} < J_i ||C'^N_l|| J_f > .
\end{aligned}
\tag{1.52}
$$

In the special case of a nuclear moment, $J_f = J_i$, and the matrix element vanishes if $l + s$ is odd. This is the expected result that even-l charge multipoles conserve T, and odd-l multipoles violate T. Analogous calculations show that $T_{\mathrm{mag},l}$ and $T_{\mathrm{el},l}$ transform with the opposite sign. These results are summarized in Table 1.1.

T (or, equivalently, CP) violation is a very small effect. It follows that any term that includes more than one instance of T violation would be vanishingly small. This forbids, for example, terms with two or more T-violating nuclear moments to contribute.

1.5.3 LO contributions

From section 1.4, the leading order contributions in ΔV to the atomic EDM is,

$$d_{\text{LO}} = < 0|\vec{d}_e \frac{Q_0}{E_0 - H_0}\Delta V|0> + < 0|\vec{d}_e \frac{Q_0}{E_0 - H_0}\Delta V|0>. \tag{1.53}$$

Since $\vec{d}_e$ cannot cause a nuclear excitation, these terms involve ground state moments of the nuclear multipoles that appear in ΔV. Also, the electronic multipole in ΔV must be odd under parity, so that the combined parity with $\vec{d}_e$ becomes even. From these considerations, the only multipoles in ΔV that can contribute up to $(R_N/R_A)^3$ order are C_3 and $T_{\text{mag},2}$ in the point-nucleus interaction, and C_1 in the penetration term.

We evaluate the C_3 contribution.

$$-\frac{4\pi\alpha}{7R_A}\left(\frac{R_N}{R_A}\right)^3\left(<0||\vec{d}_e\frac{Q_0}{E_0 - H_0}\left[C_3^A \odot C_3^N\right]||0> +\right.$$

$$\left. + <0||\left[C_3^A \odot C_3^N\right]\frac{Q_0}{E_0 - H_0}\vec{d}_e||0>\right)$$

$$= -\frac{4\pi\alpha}{7R_A}\left(\frac{R_N}{R_A}\right)^3\frac{1}{\begin{pmatrix} F & 1 & F \\ -F & 0 & F \end{pmatrix}}\sum_{n'\neq0}\frac{1}{E_0 - E_{n'}}$$

$$\left(<\alpha_e j_e \alpha_N j_N|\vec{d}_{e,10}|\alpha'_e j'_e \alpha_N j_N><\alpha'_e j'_e \alpha_N j_N|C_3^A \odot C_3^N|\alpha_e j_e \alpha_N j_N>\right.$$

$$\left. + <\alpha_e j_e \alpha_N j_N|C_3^A \odot C_3^N|\alpha'_e j'_e \alpha_N j_N><\alpha'_e j'_e \alpha_N j_N|\vec{d}_{e,10}|\alpha_e j_e \alpha_N j_N>\right)$$

$$= -\frac{4\pi\alpha}{7R_A}\left(\frac{R_N}{R_A}\right)^3\frac{1}{\begin{pmatrix} F & 1 & F \\ -F & 0 & F \end{pmatrix}}\sum_{n'\neq0}\frac{1}{E_0 - E_{n'}}[F]<FF10|FF>$$

$$\left[-\begin{Bmatrix} j'_e & j_N & F \\ F & 1 & j_e \end{Bmatrix}\begin{Bmatrix} j'_e & j_e & 3 \\ j_N & j_N & F \end{Bmatrix}\right.$$

$$\times <\alpha_e j_e||\vec{d}_e||\alpha'_e j'_e><\alpha'_e j'_e||C_3^A||\alpha_e j_e><\alpha_N j_N||C_3^N||\alpha_N j_N>$$

$$-\begin{Bmatrix} j_e & j'_e & 3 \\ j_N & j_N & F \end{Bmatrix}\begin{Bmatrix} j_e & j_N & F \\ F & 1 & j'_e \end{Bmatrix}$$

$$\left.\times <\alpha_e j_e||C_3^A||\alpha'_e j'_e><\alpha'_e j'_e||\vec{d}_e||\alpha_e j_e><\alpha_N j_N||C_3^N||\alpha_N j_N>\right]$$

$$= \frac{4\pi\alpha}{7R_A}\left(\frac{R_N}{R_A}\right)^3[F]^2\sum_{n'}\frac{1}{E_0 - E_{n'}}\begin{Bmatrix} j'_e & j_N & F \\ F & 1 & j_e \end{Bmatrix}\begin{Bmatrix} j'_e & j_e & 3 \\ j_N & j_N & F \end{Bmatrix}$$

$$\times <\alpha_N j_N||C_3^N||\alpha_N j_N>$$

$$\times (<\alpha_e j_e||\vec{d}_e||\alpha'_e j'_e><\alpha'_e j'_e||C_3^A||\alpha_e j_e> + <\alpha_e j_e||C_3^A||\alpha'_e j'_e><\alpha'_e j'_e||\vec{d}_e||\alpha_e j_e>),$$

$$\tag{1.54}$$

where n' stands for the state $|\alpha'_e j'_e \alpha_N j_N >$.

The other two terms can be calculated by analogy, as only the multipolarity of C_3 entered in the calculation above. The $T_{\text{mag},2}$ term becomes

$$
-\frac{4\pi\alpha}{5R_A}\left(\frac{R_N}{R_A}\right)^2 \left(< 0||\vec{d_e}\frac{Q_0}{E_0 - H_0}\left[T^A_{\text{mag},2} \odot T^N_{\text{mag},2}\right]||0 > + \right.
$$
$$
\left. + < 0||\left[T^A_{\text{mag},2} \odot T^N_{\text{mag},2}\right]\frac{Q_0}{E_0 - H_0}\vec{d_e}||0 > \right)
$$
$$
=\frac{4\pi\alpha}{5R_A}\left(\frac{R_N}{R_A}\right)^2 [F]^2 \sum_{n'}\frac{1}{E_0 - E_{n'}}\left\{\begin{array}{ccc} j'_e & j_N & F \\ F & 1 & j_e \end{array}\right\}\left\{\begin{array}{ccc} j'_e & j_e & 2 \\ j_N & j_N & F \end{array}\right\}
$$
$$
\times < \alpha_N j_N||T^N_{\text{mag},2}||\alpha_N j_N >
$$
$$
\times \left(< \alpha_e j_e||\vec{d_e}||\alpha'_e j'_e >< \alpha'_e j'_e||T^A_{\text{mag},2}||\alpha_e j_e > \right.
$$
$$
\left. + < \alpha_e j_e||T^A_{\text{mag},2}||\alpha'_e j'_e >< \alpha'_e j'_e||\vec{d_e}||\alpha_e j_e > \right). \tag{1.55}
$$

From Appendix A.2, the effective operator for the C_1 penetration term is

$$
V_{\text{eff}}(C^A_1 \odot C^N_1) =\frac{4\pi\alpha}{R_A}\frac{1}{10}\sqrt{\frac{4\pi}{3}}\left(\frac{R_N}{R_A}\right)^3\left[R^4_A\sum_{i=1}^{Z}\left(\overleftarrow{\nabla}_{i,m}\delta^3(\vec{x_i}) + \delta^3(\vec{x_i})\overrightarrow{\nabla}_{i,m}\right)\right]
$$
$$
\odot \int d^3y\left(\frac{y}{R_N}\right)^3\rho_N(\vec{y})Y_1(\Omega_y). \tag{1.56}
$$

Again by analogy, the C_1 penetration contribution becomes

$$
\left(< 0||\vec{d_e}\frac{Q_0}{E_0 - H_0}V_{eff}(C^A_1 \odot C^N_1)||0 > + < 0||V_{eff}(C^A_1 \odot C^N_1)\frac{Q_0}{E_0 - H_0}\vec{d_e}||0 > \right)
$$
$$
=-\frac{4\pi\alpha}{10R_A}\left(\frac{R_N}{R_A}\right)^3 [F]^2 \sum_{n'}\frac{1}{E_0 - E_{n'}}\left\{\begin{array}{ccc} j'_e & j_N & F \\ F & 1 & j_e \end{array}\right\}\left\{\begin{array}{ccc} j'_e & j_e & 1 \\ j_N & j_N & F \end{array}\right\}
$$
$$
\times < \alpha_N j_N||\int d^3y\left(\frac{y}{R_N}\right)^3 Y_1(\Omega_y)\rho_N(\vec{y})||\alpha_N j_N >
$$
$$
\times \left[< \alpha_e j_e||\vec{d_e}||\alpha'_e j'_e >< \alpha'_e j'_e||R^4_A\sum_i\left(\overleftarrow{\nabla}_i\delta^3(\vec{x_i}) + \delta^3(\vec{x_i})\overrightarrow{\nabla}_i\right)||\alpha_e j_e > \right.
$$
$$
\left. + < \alpha_e j_e||R^4_A\sum_i\left(\overleftarrow{\nabla}_i\delta^3(\vec{x_i}) + \delta^3(\vec{x_i})\overrightarrow{\nabla}_i\right)||\alpha'_e j'_e >< \alpha'_e j'_e||\vec{d_e}||\alpha_e j_e > \right]. \tag{1.57}
$$

The usual Schiff moment is given by

$$
\vec{S} = \frac{e}{10}\left(< y^2\vec{y} > -\frac{5}{3Z} < y^2 >< \vec{y} > \right), \tag{1.58}
$$

which was corrected in [9] to

$$\vec{S} = \frac{e}{10}\left[<y^2\vec{y}> - \frac{5}{3Z}\left\langle y^2\left(1 - \frac{4\sqrt{\pi}}{5}Y_2(\hat{y})\right)\otimes\vec{y}\right\rangle_1\right].$$ (1.59)

Inspection of the C_1 penetration term (1.57) shows that it can be identified with the first term in eq. (1.59). In our formalism, the other 2 terms come from $[A,\Delta V]$, with C_0 penetration and C_2 penetration terms in ΔV. These contributions appear at the next order in perturbation theory. Note, however, that the C_3 contribution is of the same order in R_N/R_A as the Schiff moment, and the M_2 term is actually larger in terms of this power counting.

There are restrictions on the nuclear and atomic spins, from angular momentum considerations:

- For the C_1 contribution, there is no restriction on atomic spin, as $\vec{d}_e$ and the electronic C_1 operator can couple to form a scalar. The nuclear spin does have to be $1/2$ or greater in order to have a $J = 1$ moment.

- For the M_2 contribution, j_e must be at least $1/2$, and j_N must be at least 1.

- For the C_3 contribution, j_e must be at least 1, and j_N must be at least $3/2$.

1.5.4 NLO Summary and Conclusions

For d_{NLO}, possible combinations of operators that can contribute to eq. (1.46), up to $(R_N/R_A)^3$ order, are: without a nuclear excitation,

- $[A, C_2^A \odot C_2^N]$

- $[A, C_0^A \odot C_0^N]$ penetration

- $[A, C_2^A \odot C_2^N]$ penetration

- $[A, M_1^A \odot M_1^N]$

- $M_1^A \odot M_1^N$ and $M_2^A \odot M_2^N$

and with a nuclear excitation, it has to be a combination of

- $[A, H_N]$ or

- $T_{\mathrm{el},1}^{'A} \odot T_{\mathrm{el},1}^N$

and

- $M_1^A \odot M_1^N$,

- $T_{\text{el},2}'^{A} \odot T_{\text{el},2}^{N}$, or

- $C_2^A \odot C_2^N$.

Since the typical nuclear excitation energy is larger than the typical atomic excitation energy by a factor of approximately R_A/R_N, the energy denominator with a nuclear excitation counts as one more power of R_N/R_A. The first two contributions, as mentioned earlier, completes the Schiff moment.

Some of the other terms at this order can be identified with terms first considered in [9]. In addition, using the Breit interaction has resulted in contributions from transverse electric moments, and power counting based on the ratio R_N/R_A ensures consistent treatment of the terms. The NLO and NNLO expressions after the implementation of Schiff cancellation, (1.46, 1.47), is also a new result.

The effects of $(C_0^N - < C_0^N >)$ are yet to be considered. Since this operator is not suppressed by powers of R_N/R_A, it can potentially result in a relatively large EDM signal. Also, higher order terms involving the C_1 interaction can also be large in terms of power counting, and may require special treatment. These are the next steps in this project, along with exhausting all possible terms at NNLO and above.

Chapter 2

Harmonic-Oscillator-Based Effective Theory

2.1   Introduction

One of the difficulties in building an effective theory (ET) of the nucleus is that the nuclear potential is finely tuned. The shallow binding of the deuteron and the long scattering lengths in nucleon-nucleon (NN) scattering are very sensitive to small changes in the NN potential. The potential is attractive at long range and highly repulsive in the short range, and a successful ET of nucleon interactions must respect how nuclear observables result from the interplay between these components to the potential.

The use of harmonic-oscillator (HO) basis is one way to disentangle the short- and the long-range physics. Oscillator length, b, gives HO eigenstates a characteristic length scale, and if we choose b to have an intermediate scale, then the short-range physics and the long-range physics can be treated separately. Effectiveness of the traditional shell model approach to nuclear physics was partly due to this separation of the length scales. Another reason for shell model's success is the center-of-mass separability in the basis of HO Slater determinants, i.e. if all states having up to N quanta are in the model space, then the effective Hamiltonian can be separated into the center-of-mass motion terms and relative motion terms. Despite these virtues of shell model, its approximations are uncontrolled, compromising its reliability. Our goal is to formulate a harmonic-oscillator-based effective theory (HOBET) that takes advantage of the scale separation and the computational machinery in the HO basis, while maintaining the systematic order-by-order approximation of effective theory (ET).

The final product should be similar to an effective field theory: a HOBET that contains all the operators allowed by the symmetries as interactions. The strengths of each interaction, in the end, should be determined by directly matching to experimental results, e.g. scattering phase shifts. This avoids reference to any sort of high-momentum NN potential, which would be an unnecessary intermediate step between QCD and the final ET.

The first step in building a HOBET is to introduce a cutoff Λ in the HO basis, including

all HO states with Λ energy quanta or less in the allowed (P-) space. The point of using an ET is to restrict all of the calculations to this finite Hilbert space, rather than carrying out an expensive calculation in the full, infinite space. As restricting a wavefunction into P-space strips out the long-range part of the wavefunction, it may be expected that the information about the phase shift would be lost in the process. We show, using a toy model of a double square well, that the longest-range behavior of the restricted wavefunction contains sufficient information to recover the phase shift. To make the potential a model of the NN interaction, it is attractive at long range, and has a repulsive core in the short range, and has a bound state at the deuteron binding energy.

Then we derive an equation, which we call the master equation for the HOBET, that resembles the Lippmann-Schwinger equation, but only acts on states in the P-space. The contributions from states in the excluded (Q-) space are encoded in effective operators, which will depend on the energy E of the system as well as the cutoff Λ. For the double square well, it is possible to solve the Schrödinger equation (or the Lippmann-Schwinger equation) analytically, and the Green's function representation of $1/(E-H)$ in coordinate space can be written down in closed form. This allows us to verify that solving the master equation reproduces the P-space projection of the exact, full-space wavefunction.

In the actual NN potential problem, this is not a desirable path to take. Calculating the Green's function $1/(E-H)$ for the NN potential is computationally expensive, and does not clarify what roles the short-range and long-range parts of the potential play in determining the observables. As in [17], we define the tilde state

$$|\tilde{\alpha}> \equiv \frac{E}{E - QT + i\epsilon}|\alpha> .$$ (2.1)

The tilde state modifies the long-range behavior of the "edge" states, the states with maximum nodal quantum number n for given angular momentum l. Instead of the $e^{-r^2/2}$ tail common to HO eigenstates, the tilde state tails off much more slowly, as e^{ikr}/kr, where $k = \sqrt{2E/\hbar\omega}$. Examination shows that the expression here is the analytical continuation of the tilde state in [17].

The tilde state modification to the edge state wavefunction has the effect of bringing in long-range physics into P-space. The short-range physics is modeled by replacing the short-range part of the effective potential by a contact-gradient series. This is equivalent to expanding around the intermediate momentum scale $q \sim 1/b$, using a small parameter (a/b), where a is the length scale associated with the short-range repulsion of the NN interaction. For HOBET to be successful, each successive order in the contact-gradient expansion must produce better approximation to the actual interaction. We show the results of some of the studies that tested this using the toy model.

2.2 Formalism

First, we choose the oscillator length b. b is a free parameter in the theory, but we wish to choose a value that is longer than the length scale of the repulsive core, and shorter than

the length scale associated with small deuteron binding and large scattering lengths. As in [17], we use $b = 1.7$fm, roughly the range of the tail in the NN potential. The dependence of the theory on this parameter is a topic for future investigation.

Once b is chosen, the usual three-dimensional harmonic oscillator (HO) Hamiltonian can be solved, and the set of HO eigenstates is the basis that we will conduct our calculations. Each HO state is labeled by the nodal quantum number n and angular momentum l. Our convention is that the lowest lying states have the nodal quantum number $n = 1$. The energy eigenvalues are given by

$$H|nl> = \hbar\omega(2n + l - 1/2)|nl>, \tag{2.2}$$

and we will write the coordinate space wavefunction as

$$< r|nl> = R_{nl}(r)Y_{lm}(\Omega). \tag{2.3}$$

The form of this wavefunction is given in appendix B.1.

Next, we pick the cutoff for the ET, parametrized by an integer Λ. States with energies up to $(\Lambda + 3/2)\hbar\omega$ are included in the model space ($3/2\hbar\omega$ is the ground state energy). Projection operator into the model space (P-space) can be written

$$P = \sum_{nl}^{2n+l\leq\Lambda} |nl><nl|. \tag{2.4}$$

Projection operator out of the P-space, and into the Q-space, is then

$$Q = 1 - P. \tag{2.5}$$

For the time being, we use $\Lambda = 8$, but observing the changes in the ET as this parameter (much like b) is adjusted is one way to investigate the properties of this theory.

2.3   Phase Shift

In HOBET, only the P-space projection of wavefunctions appear explicitly. As HO wavefunctions have Gaussian tails that quickly goes to 0, one may wonder if projecting a scattering wavefunction into a finite space of HO states would destroy the information about the asymptotic behavior. Before embarking on ET calculations, we will demonstrate that P-space projection preserves enough information for us to recover the phase shift.

Let

$$< r|\psi_l> = \psi_l(r)Y_{lm}(\Omega) \tag{2.6}$$

be the partial wave with angular momentum l for the full-space solution of the Schrödinger equation, $H|\psi> = E|\psi>$, and

$$< r|P|\psi_l> = \psi_l^{(P)}(r)Y_{lm}(\Omega) \tag{2.7}$$

be its P-space projection.

Consider integrals of the form

$$\int_0^\infty r^2 dr \; r^{2m+l} e^{-r^2/2} R_{nl}(r), \tag{2.8}$$

where m is a non-negative integer. These integrals vanish when $m < n$. This allows us to identify the longest-range information in $\psi_l(r)$ that remains intact after P-space projection. To see this, explicitly expand $\psi_l(r)$ in HO basis.

$$\psi_l(r) = \sum_{n=1}^\infty <r|nl><nl|\psi_l> = \sum_{n=1}^\infty c_{n,\psi} R_{nl}(r), \tag{2.9}$$

with the definition $c_{n,\psi} \equiv <nl|\psi_l>$. Then the P-space projection is

$$\psi_l^{(P)}(r) = \sum_{n=1}^\infty <r|nl><nl|P|\psi_l> = \sum_{n=1}^{n_{\max}} c_{n,\psi} R_{nl}(r), \tag{2.10}$$

where $n_{\max}$ is the largest integer n that satisfies $2n + l \le \Lambda$.

Using the fact that the integral (2.8) vanishes for $m < n_{\max}$,

$$\begin{aligned}
I(m) &\equiv \int_0^\infty r^2 dr \; r^{2m+l} e^{-r^2/2} \psi_l(r) \\
&= \sum_{n=1}^\infty \int_0^\infty r^2 dr \; r^{2m+l} e^{-r^2/2} R_{nl}(r) \\
&= \sum_{n=1}^{n_{\max}} \int_0^\infty r^2 dr \; r^{2m+l} e^{-r^2/2} R_{nl}(r) \\
&= \int_0^\infty r^2 dr \; r^{2m+l} e^{-r^2/2} \psi_l^{(P)}(r).
\end{aligned} \tag{2.11}$$

Evaluating the integral using the projected wavefunction yields the same answer as using the full wavefunction. In particular, $I(n_{\max})$ and $I(n_{\max} - 1)$ are the integrals that capture the two longest-range behaviors of $\psi_l(r)$ that is preserved in the P-space projection.

If these moments receive most of their weights from points outside the strong core, then their values are only sensitive to the asymptotic part of the wavefunction, and contain information about the phase shift. To test this, we replace the true wavefunction by its asymptotic form,

$$\psi_l(r) \to A \left(\cos \delta_l j_l(kr) - \sin \delta_l n_l(kr) \right), \tag{2.12}$$

where $j_l(r)$ and $n_l(r)$ are the usual spherical Bessel functions, and $k \equiv \sqrt{2E/\hbar\omega}$; the test is whether this replacement affects the long-range moments or not.

Making the replacement,

$$I(m) \rightarrow A \int_0^\infty r^2 dr \, r^{2m+l} e^{-r^2/2} \left(\cos \delta_l j_l(kr) - \sin \delta_l n_l(kr) \right). \tag{2.13}$$

Integrals involving the spherical Bessel functions can be evaluated:

$$I_j(m) \equiv \int_0^\infty r^2 dr \, r^{2m+l} e^{-r^2/2} j_l(kr)$$

$$= 2^m k^l e^{-k^2/2} \sqrt{\frac{\pi}{2}} \frac{\Gamma(m+l+3/2)}{\Gamma(l+3/2)} \, {}_1F_1(-m; l+3/2; k^2/2) \tag{2.14}$$

and

$$I_n(m) \equiv \int_0^\infty r^2 dr \, r^{2m+l} e^{-r^2/2} n_l(kr)$$

$$= -\frac{2^{m+l} e^{-k^2/2}}{\sqrt{\pi} k^{l+1}} \Gamma(m+1) \Gamma(l+1/2) {}_1F_1(-l-m-1/2; 1/2-l; k^2/2), \tag{2.15}$$

where ${}_1F_1(a; b; c)$ is the confluent hypergeometric function, as defined in [18]. Having evaluated the integrals, we can write

$$I(n_{\max}) \rightarrow A \left(\cos \delta_l I_j(n_{\max}) - \sin \delta_l I_n(n_{\max}) \right)$$
$$I(n_{\max} - 1) \rightarrow A \left(\cos \delta_l I_j(n_{\max} - 1) - \sin \delta_l I_n(n_{\max} - 1) \right). \tag{2.16}$$

If the replacement of the wavefunction by its asymptotic piece has only small effects on the values of the moments, we can solve for $\cot \delta_l$ to obtain

$$k \cot \delta_l \approx k \frac{I_n(n_{\max}) I(n_{\max} - 1) - I_n(n_{\max} - 1) I(n_{\max})}{I_j(n_{\max}) I(n_{\max} - 1) - I_j(n_{\max} - 1) I(n_{\max})}. \tag{2.17}$$

We need 2 integrals $I(n_{\max})$ and $I(n_{\max} - 1)$ so that the arbitrary constant factor A can be removed. Once we have $P|\psi >$ as a function of k, It is straightforward to expand the right-hand side in power series of k, and this can be compared to the true effective range expansion (the left-hand side).

We introduce a toy model for the NN potential for this comparison. It is a mildly attractive square well with a strongly repulsive core. Specifically, we use the potential

$$V(r) = \begin{cases} V_1, & r < a_1 \\ -V_2, & a_1 < r < a_2 \\ 0, & r > a_2, \end{cases} \tag{2.18}$$

where $a_1 = 0.25 \text{fm}$, $a_2 = 1.7 \text{fm}$, $V_1 = 40 \times 54.531 \text{MeV}$, and $V_2 = 54.531 \text{MeV}$ (note the minus sign in the definition of V).

The lengths were chosen to match the typical length scales in the NN potential, and the strength of the potential was chosen so that the potential has a bound state at the deuteron binding energy, $E = -2.22452\text{MeV}$. Although the deuteron wavefunction contains $l = 2$ components, the model should be sufficient for capturing the relevant features of the NN potential. The square well is useful because it is solvable analytically; scattering wavefunctions can be written in closed form using standard functions, with explicit dependence on k.

By analytically solving the Schrödinger equation (or the equivalent Lippmann-Schwinger equation) analytically, we obtain the exact expression for the phase shift in the $^1S_0$ channel, δ_0. Then, expanding $k \cot \delta_0$ in powers of k gives the usual effective range expansion,

$$k \cot \delta_0 = -0.457721 + 0.323155k^2 + 1.15663 \times 10^{-2}k^4 + 4.23022 \times 10^{-4}k^6 + \ldots. \quad (2.19)$$

The test is to project the exact scattering wavefunction (written as a function of k) into P-space, then use the procedure described above to find the approximate effective range expansion, and compare with the exact result. Numerical calculation on Mathematica gives the series

$$k \cot \delta_0 \approx -0.457725 + 0.323136k^2 + 1.15749 \times 10^{-2}k^4 + 4.22876 \times 10^{-4}k^6 + \ldots. \quad (2.20)$$

The fractional errors in the approximated coefficients of k^{2n} are $1.0 \times 10^{-5} : 5.9 \times 10^{-6} : 7.4 \times 10^{-4} : 3.5 \times 10^{-4}$. This shows that the long-range moments are insensitive to the interior parts of the wavefunction and that the P-space projection of the scattering wavefunction does contain sufficient information for reconstructing the asymptotic behavior with high precision.

2.4   Master Equation

The equation that we use to find $P|\psi>$ is inspired by both the Bloch-Horowitz equation and the Lippmann-Schwinger equation.

Bloch-Horowitz equation for an eigenvalue problem $H|\psi>= E|\psi>$ is given by

$$H_{\text{eff}}P|\psi>= EP|\psi>, \quad (2.21)$$

where the energy-dependent effective Hamiltonian H_{eff} is defined as

$$\begin{aligned} H_{\text{eff}} &\equiv H + HQ\frac{1}{E - QH}QH \\ &= \frac{E}{E - TQ}\left[T - \frac{TQT}{E} + V + V\frac{1}{E - QH}QV\right]\frac{E}{E - QT}. \end{aligned} \quad (2.22)$$

The form in the second line was derived in one of the appendices of [19]. This equation prescribes a method to obtain the P-space restriction of an eigenstate of H, using effective operators in P-space that implicitly take account of the Q-space effects.

The Lippmann-Schwinger equation is

$$|\psi> = |\phi> + \frac{1}{E-T+i\epsilon}V|\psi> . \tag{2.23}$$

This is an equation that is typically solved self-consistently for $|\psi>$; the equation picks out the eigenstate of H that is asymptotically an unperturbed incoming wave plus an outgoing wave that has been altered by the potential. $(+i\epsilon)$ in the denominator ensures that the scattering term has only an outgoing wave component in the $r \to \infty$ limit.

In order to derive the master equation, we start with a rewritten version of the Lippmann-Schwinger equation:

$$|\psi> = |\phi> + \frac{1}{E-H+i\epsilon}V|\phi> . \tag{2.24}$$

Projecting this into P space,

$$P|\psi> = P|\phi> + P\frac{1}{E-H+i\epsilon}V|\phi> . \tag{2.25}$$

Using the relation

$$\frac{1}{E-H+i\epsilon} = \frac{1}{E-HQ+i\epsilon} + \frac{1}{E-H+i\epsilon}HP\frac{1}{E-HQ+i\epsilon}$$
$$= \left(1 + \frac{1}{E-H+i\epsilon}HP\right)\frac{1}{E-HQ+i\epsilon}, \tag{2.26}$$

the scattered wave (the last term) in eq. (2.25) can be written as

$$P\frac{1}{E-H+i\epsilon}V|\phi> = P\left(1 + \frac{1}{E-H+i\epsilon}HP\right)\frac{1}{E-HQ+i\epsilon}V|\phi>$$
$$= PMP\frac{1}{E-HQ+i\epsilon}V|\phi>, \tag{2.27}$$

where

$$M \equiv 1 + M_{pp} + M_{pp}^2 + M_{pp}^3 + \cdots = \frac{1}{1-M_{pp}} \tag{2.28}$$

with

$$M_{pp} \equiv P\frac{1}{E-HQ+i\epsilon}HP. \tag{2.29}$$

M_{pp} is suggestive of the Bloch-Horowitz effective Hamiltonian, eq. (2.22), and it allows a

useful rewrite:

$$
\begin{aligned}
M_{pp} &\equiv P\frac{1}{E-HQ+i\epsilon}HP \\
&= PH\frac{1}{E-QH+i\epsilon}P \\
&= PT\frac{1}{E-QH+i\epsilon}P + PV\frac{1}{E-QH+i\epsilon}P \\
&= PT\left(\frac{1}{E-QT+i\epsilon}+\frac{1}{E-QT+i\epsilon}QV\frac{1}{E-QH+i\epsilon}\right)P + PV\frac{1}{E-QH+i\epsilon}P \\
&= PT\frac{1}{E-QT+i\epsilon}P + P\left(T\frac{1}{E-QT+i\epsilon}Q+1\right)V\frac{1}{E-QH+i\epsilon}P \\
&= PT\frac{1}{E-QT+i\epsilon}P + P\frac{E}{E-TQ+i\epsilon}V\frac{1}{E-QH+i\epsilon}P \\
&= PT\frac{1}{E-QT+i\epsilon}P + P\frac{E}{E-TQ+i\epsilon}\left(V+V\frac{1}{E-QH+i\epsilon}QV\right)\frac{1}{E-QT+i\epsilon}P.
\end{aligned}
$$

$$(2.30)$$

Rewriting the last part of the scattered wave,

$$
\begin{aligned}
\frac{1}{E-HQ+i\epsilon}V &= \frac{1}{E-TQ+i\epsilon}V + \frac{1}{E-TQ+i\epsilon}VQ\frac{1}{E-HQ+i\epsilon}V \\
&= \frac{1}{E-TQ+i\epsilon}\left(V+V\frac{1}{E-QH+i\epsilon}QV\right).
\end{aligned}
$$

$$(2.31)$$

Putting these ingredients together, we get the master equation,

$$
P|\psi> = P|\phi> + PMP\frac{1}{E-TQ+i\epsilon}\left(V+V\frac{1}{E-QH+i\epsilon}QV\right)|\phi>.
$$

$$(2.32)$$

2.5 Tilde State

In order to evaluate the matrix elements in the master equation, we need an expression for the tilde state,

$$
|\tilde{\alpha}> \equiv \frac{E}{E-QT+i\epsilon}|\alpha>.
$$

$$(2.33)$$

For non-edge states (states that are not at the boundary between the P-space and the Q-space), $QT|\alpha>=0$, and so

$$
|\tilde{\alpha}> = |\alpha>.
$$

$$(2.34)$$

For an edge state, the tilde state can be expanded in HO basis as

$$
\frac{E}{E-QT+i\epsilon}|nl> = \sum_{i=0}^{\infty}\tilde{g}_i(k^2;n,l)|n+i,l>.
$$

$$(2.35)$$

In [17], an iterative scheme to find the coefficients $\tilde{g}_i$ was given for $E < 0$ ($k^2 < 0$). However, this scheme is not useful in the $E > 0$ case, because $\tilde{g}_i$ does not vanish quickly as $i \to \infty$.

Instead of the iteration method, we should adapt the Green's function method, which is also given in [17], to the $E > 0$ case. Key observation is that

$$(E - T)|\tilde{\alpha}> = P \left[E - T \frac{E}{E - QT + i\epsilon} \right] P|\alpha>$$

$$= \left[P \frac{1}{E - T + i\epsilon} P \right]^{-1} |\alpha>, \tag{2.36}$$

so the tilde state can be written

$$|\tilde{\alpha}> = \frac{1}{E - T + i\epsilon} \left[P \frac{1}{E - T + i\epsilon} P \right]^{-1} |\alpha>. \tag{2.37}$$

It is easier to evaluate $1/(E - T + i\epsilon)$ in momentum space than in coordinate space, since T is diagonal in momentum space. First, the momentum space expression for this operator is (detailed derivations are given in the appendix B.2)

$$<p'|\frac{1}{E - T + i\epsilon}|p> = \frac{2}{\hbar\omega} \frac{1}{k^2 - p^2 + i\epsilon} (2\pi)^3 \delta^3(\vec{p} - \vec{p'}). \tag{2.38}$$

As is well known, HO wavefunctions are Fourier transforms of themselves, but up to a constant that depends on the normalization convention:

$$<p|nlm> = \int d^3r <p|r><r|nlm>$$

$$= \int d^3r\, e^{-i\vec{p}\cdot\vec{r}} \sqrt{\frac{2(n-1)!}{\Gamma(n+l+1/2)}} L_{n-1}^{l+1/2}(r^2) r^l e^{-r^2/2} Y_{lm}(\Omega)$$

$$= (2\pi)^{3/2}(-i)^l(-1)^{n-1} \sqrt{\frac{2(n-1)!}{\Gamma(n+l+1/2)}} p^l e^{-p^2/2} L_{n-1}^{l+1/2}(p^2) Y_{lm}(\Gamma_p). \tag{2.39}$$

Expanding the Laguerre polynomials,

$$<n'lm|\frac{1}{E - T + i\epsilon}|nlm>$$

$$= \frac{2}{\hbar\omega} \sum_{m=0}^{n-1} \sum_{m'=0}^{n'-1} \frac{(-1)^{m+m'+n+n'} \sqrt{4(n-1)!(n'-1)!\Gamma(n+l+1/2)\Gamma(n'+l+1/2)}}{m!m'!(n-1-m)!(n'-1-m')!\Gamma(l+m+3/2)\Gamma(l+m'+3/2)} \times$$

$$\times \int_0^\infty \frac{dp\, p^{2l+2m+2m'+2} e^{-p^2}}{k^2 - p^2 + i\epsilon}. \tag{2.40}$$

The last integral can be turned into a principal value integral, then evaluated:

$$
\int_0^\infty \frac{dp\, p^{2N} e^{-p^2}}{k^2 - p^2 + i\epsilon}
$$
$$
= -\frac{1}{2}\left[\mathcal{P}\int_0^\infty du \frac{u^{N-1/2} e^{-u}}{u - k^2} + i\pi k^{2N-1} e^{-k^2}\right]
$$
$$
= \frac{e^{-k^2}}{2}\left[(N-1/2)(-k^2)^{N-1/2}\Gamma(N-1/2)\gamma(1/2-N,-k^2) - i\pi k^{2N-1}\right], \tag{2.41}
$$

where $\gamma(a,z) = \Gamma(a) - \Gamma(a,z)$ is the lower incomplete gamma function. The matrix elements of $1/(E - T + i\epsilon)$ in the HO basis are

$$
< n'lm|\frac{1}{E - T + i\epsilon}|nlm > \tag{2.42}
$$
$$
= \frac{1}{\hbar\omega}\sum_{m=0}^{n-1}\sum_{m'=0}^{n'-1}\frac{(-1)^{m+m'+n+n'+1}\sqrt{4(n-1)!(n'-1)!\Gamma(n+l+1/2)\Gamma(n'+l+1/2)}}{m!m'!(n-1-m)!(n'-1-m')!\Gamma(l+m+3/2)\Gamma(l+m'+3/2)} \tag{2.43}
$$
$$
\times e^{-k^2}\left[(N-1/2)(-k^2)^{N-1/2}\Gamma(N-1/2)\gamma(1/2-N,-k^2) - i\pi k^{2N-1}\right], \tag{2.44}
$$

where $N \equiv l + m + m' + 1$.

Now that the matrix elements of $1/(E - T + i\epsilon)$ in the HO basis are known,

$$
\left[P\frac{1}{E - T + i\epsilon}P\right]^{-1} \tag{2.45}
$$

can be calculated by matrix inversion. In practice, we expand the answers as power series in k and keep only up to $\mathcal{O}(k^6)$, as that is the highest order we consider for the effective range expansion. Here, define a ket $|\alpha_{nlm} >$ and its radial wavefunction $R_{nl}^\alpha(r)$ as follows:

$$
|\alpha_{nlm} > \equiv \left[P\frac{1}{E - T + i\epsilon}P\right]^{-1}|nlm >
$$
$$
R_{nl}^\alpha(r)Y_{lm}(\Omega) = < r|\alpha_{nlm} > . \tag{2.46}
$$

This function acts as the driving term for the Green's function equation (2.37). The action of the operator $1/(E - T + i\epsilon)$ is given by the integration kernel

$$
g_l^{(0)}(r_1, r_2) = -ik j_l(kr_<)h_l^{(1)}(kr_>), \tag{2.47}
$$

where $h_l^{(1)}$ is the spherical Hankel function of the first kind. This was constructed from 2 solutions to the equation $(E - T)|\phi > = 0$ (the Helmholtz equation, in coordinate space), which satisfy the boundary conditions. $j_l(kr)$ is regular at the origin, and $h_l^{(1)}(kr)$ asymptotes to an outgoing wave as $r \to \infty$.

Eq. (2.37) turns into the following equation to find the tilde state:

$$< r|\tilde{\alpha} >=\tilde{R}_{nl}^{\alpha}Y_{lm}(\Omega)$$
$$= -ikY_{lm}(\Omega)\left[\int_0^r r'^2 dr' j_l(kr')h_l^{(1)}(kr)R_{nl}^{\alpha}(r')\right.$$
$$\left. + \int_r^{\infty} r'^2 dr' j_l(kr)h_l^{(1)}(kr')R_{nl}^{\alpha}(r')\right]. \tag{2.48}$$

For negative k^2, the integral in eq. (2.40) can be done without taking the principal part, and it is

$$\int_0^{\infty} \frac{dp\, p^{2N}e^{-p^2}}{k^2 - p^2 + i\epsilon} = -\frac{e^{-k^2}}{2}(-k^2)^{N-1/2}\Gamma(N+1/2)\Gamma(1/2 - N, -k^2). \tag{2.49}$$

Numerical tests have confirmed that using this expression reproduces the tilde state obtained from the recursion method. The difference between eq. (2.41) and (2.49) is 0 if N is an integer (which is satisfied) and k is in the upper half of the complex plane. Thus, the tilde state we have for $E > 0$ is the correct analytic continuation from the bound state expression.

For the toy model, the solution to the full Schrödinger equation is known analytically, and the Green's function $1/(E - H + i\epsilon)$ can be evaluated in coordinate space. This allows a direct test of the master equation and the evaluation of matrix elements, as follows.

Consider

$$\frac{1}{E - HQ + i\epsilon} - \frac{1}{E - TQ + i\epsilon}$$
$$= \frac{1}{E - TQ + i\epsilon}(E - TQ)\frac{1}{E - HQ + i\epsilon} - \frac{1}{E - TQ + i\epsilon}(E - HQ)\frac{1}{E - HQ + i\epsilon}$$
$$= \frac{1}{E - TQ + i\epsilon}VQ\frac{1}{E - HQ + i\epsilon}. \tag{2.50}$$

Using this relation, the operator M_{pp} can be rewritten as

$$M_{pp} = PT\frac{1}{E - QT + i\epsilon}P$$
$$+ P\frac{E}{E - TQ + i\epsilon}\left(V + V\frac{1}{E - QH + i\epsilon}QV\right)\frac{1}{E - QT + i\epsilon}P$$
$$= PT\frac{1}{E - QT + i\epsilon}P$$
$$+ P\frac{E}{E - TQ + i\epsilon}\left(V + VQ\frac{1}{E - HQ + i\epsilon}V\right)\frac{1}{E - QT + i\epsilon}P$$
$$= PT\frac{1}{E - QT + i\epsilon}P + P\frac{E}{E - HQ + i\epsilon}V\frac{1}{E - QT + i\epsilon}P. \tag{2.51}$$

Writing this in terms of $1/(E-T+i\epsilon)$ and $1/(E-H+i\epsilon)$,

$$
\begin{aligned}
M_{pp} =& PT\frac{1}{E-T+i\epsilon}\left[P\frac{1}{E-T+i\epsilon}P\right]^{-1} \\
&+ \left[P\frac{1}{E-H+i\epsilon}P\right]^{-1}\frac{E}{E-H+i\epsilon}V\frac{1}{E-T+i\epsilon}\left[P\frac{1}{E-T+i\epsilon}P\right]^{-1}.
\end{aligned}
\tag{2.52}
$$

Similarly, the master equation is rewritten as

$$
\begin{aligned}
P|\psi> =& P|\phi> +PMP\frac{1}{E-HQ+i\epsilon}V|\phi> \\
=& P|\phi> +PM\left[P\frac{1}{E-H+i\epsilon}P\right]^{-1}\frac{1}{E-H+i\epsilon}V|\phi>.
\end{aligned}
\tag{2.53}
$$

The expression for the free Green's function $1/(E-T+i\epsilon)$ has been given earlier. To evaluate the Green's function for the full Hamiltonian, consider two radial wavefunctions that solve the square well problem; one solution, $R_<(r)$, is regular at the origin, and the other, $R_>(r)$, is asymptotically an outgoing wave as $r\to\infty$. Then the Green's function is given by the kernel

$$
g_l(r_1,r_2) = \frac{2}{\hbar\omega W[R_<,R_>](r)}R_<(r_<)R_>(r_>),
\tag{2.54}
$$

where $W[R_<,R_>](r)$ is the Wronskian of the two wavefunctions. Numerical calculation using Mathematica showed that $P|\psi>$ obtained from the master equation matches the P-space projection of the analytical solution to the Lippmann-Schwinger equation. This is a useful check on the derivation of eq. (2.32) and its numerical implementation.

2.6 Contact-Gradient Expansion

The starting point for contact-gradient expansion for HOBET is the contact-gradient expansion used in the plane wave basis. All contact-gradient operators up to N³LO are given in Table 2.1. In the plane wave basis, the contact-gradient expansion amounts to a power series in k^2 around momentum 0, since

$$
\overrightarrow{\nabla}^2 e^{i\vec{k}\cdot\vec{r}} \equiv -k^2 e^{i\vec{k}\cdot\vec{r}} \to 0
\tag{2.55}
$$

as $k\to 0$. In HOBET, the series expansion should be around momentum $k\sim 1/b$. In [17], it was demonstrated that replacing contact-gradient operators by

$$
O \to \bar{O} = e^{r^2/2}Oe^{r^2/2}
\tag{2.56}
$$

results in a series expansion around this intermediate scale. The matrix elements of $\bar{O}$ between HO eigenstates and tilde states are given in appendix B.3.

Transitions	LO	NLO	NNLO	N³LO
$^3S_1 \leftrightarrow\, ^3S_1$ or $^1S_0 \leftrightarrow\, ^1S_0$	$\delta(\mathbf{r})$	$(\overleftarrow{\nabla}^2\delta(\mathbf{r}) + \delta(\mathbf{r})\overrightarrow{\nabla}^2)$	$\overleftarrow{\nabla}^2\delta(\mathbf{r})\overrightarrow{\nabla}^2$ $(\overleftarrow{\nabla}^4\delta(\mathbf{r}) + \delta(\mathbf{r})\overrightarrow{\nabla}^4)$	$(\overleftarrow{\nabla}^4\delta(\mathbf{r})\overrightarrow{\nabla}^2 + \overleftarrow{\nabla}^2\delta(\mathbf{r})\overrightarrow{\nabla}^4)$ $(\overleftarrow{\nabla}^6\delta(\mathbf{r}) + \delta(\mathbf{r})\overrightarrow{\nabla}^6)$
$^3S_1 \leftrightarrow\, ^3D_1$		$(\overleftarrow{D}^0\delta(\mathbf{r}) + \delta(\mathbf{r})\overrightarrow{D}^0)$	$(\overleftarrow{\nabla}^2\delta(\mathbf{r})\overrightarrow{D}^0 + \overleftarrow{D}^0\delta(\mathbf{r})\overrightarrow{\nabla}^2)$ $(\delta(\mathbf{r})\overrightarrow{\nabla}^2\overrightarrow{D}^0 + \overleftarrow{D}^0\overleftarrow{\nabla}^2\delta(\mathbf{r}))$	$(\overleftarrow{\nabla}^4\delta(\mathbf{r})\overrightarrow{D}^0 + \overleftarrow{D}^0\delta(\mathbf{r})\overrightarrow{\nabla}^4)$ $(\overleftarrow{\nabla}^2\delta(\mathbf{r})\overrightarrow{\nabla}^2\overrightarrow{D}^0 + \overleftarrow{D}^0\overleftarrow{\nabla}^2\delta(\mathbf{r})\overrightarrow{\nabla}^2)$ $(\delta(\mathbf{r})\overrightarrow{\nabla}^4\overrightarrow{D}^0 + \overleftarrow{D}^0\overleftarrow{\nabla}^4\delta(\mathbf{r}))$
$^1D_2 \leftrightarrow\, ^1D_2$ or $^3D_J \leftrightarrow\, ^3D_J$			$\overleftarrow{D}^2\delta(\mathbf{r})\overrightarrow{D}^2$	$(\overleftarrow{D}^2\overleftarrow{\nabla}^2\delta(\mathbf{r})\overrightarrow{D}^2 + \overleftarrow{D}^2\delta(\mathbf{r})\overrightarrow{\nabla}^2\overrightarrow{D}^2$
$^3D_3 \leftrightarrow\, ^3G_3$				$(\overleftarrow{D}^2\delta(\mathbf{r})\overrightarrow{G}^2 + \overleftarrow{G}^2\delta(\mathbf{r})\overrightarrow{D}^2)$
$^1P_1 \leftrightarrow\, ^1P_1$ or $^3P_J \leftrightarrow\, ^3P_J$		$\overleftarrow{\nabla}\delta(\mathbf{r})\overrightarrow{\nabla}$	$(\overleftarrow{\nabla}\overleftarrow{\nabla}^2\delta(\mathbf{r})\overrightarrow{\nabla} + \overleftarrow{\nabla}\delta(\mathbf{r})\overrightarrow{\nabla}^2\overrightarrow{\nabla})$	$\overleftarrow{\nabla}\overleftarrow{\nabla}^2\delta(\mathbf{r})\overrightarrow{\nabla}^2\overrightarrow{\nabla}$ $(\overleftarrow{\nabla}\overleftarrow{\nabla}^4\delta(\mathbf{r})\overrightarrow{\nabla} + \overleftarrow{\nabla}\delta(\mathbf{r})\overrightarrow{\nabla}^4\overrightarrow{\nabla})$
$^3P_2 \leftrightarrow\, ^3F_2$			$(\overleftarrow{\nabla}\delta(\mathbf{r})\overrightarrow{F}^1 + \overleftarrow{F}^1\delta(\mathbf{r})\overrightarrow{\nabla})$	$(\overleftarrow{\nabla}\overleftarrow{\nabla}^2\delta(\mathbf{r})\overrightarrow{F}^1 + \overleftarrow{F}^1\delta(\mathbf{r})\overrightarrow{\nabla}^2\overrightarrow{\nabla})$ $(\overleftarrow{\nabla}\delta(\mathbf{r})\overrightarrow{\nabla}^2\overrightarrow{F}^1 + \overleftarrow{F}^1\overleftarrow{\nabla}^2\delta(\mathbf{r})\overrightarrow{\nabla})$
$^1F_3 \leftrightarrow\, ^1F_3$ or $^3F_J \leftrightarrow\, ^3F_J$				$\overleftarrow{F}^3\delta(\mathbf{r})\overrightarrow{F}^3$

Table 2.1: The bare contact-gradient operators. Operator definitions are $\vec{D}^2_M = (\nabla \otimes \nabla)_{2M}$, $\vec{D}^0_0 = [(\sigma(1) \otimes \sigma(2))_2 \otimes D^2]_{00}$, $\vec{F}^3_M = (\nabla \otimes D^2)_{3M}$, $\vec{F}^1_M = [(\sigma(1) \otimes \sigma(2))_2 \otimes F^3]_{1M}$, $\vec{G}^4_M = (D^2 \otimes D^2)_{4M}$, and $\vec{G}^2_M = [(\sigma(1) \otimes \sigma(2))_2 \otimes G^4]_{2M}$. (From [17])

This replacement has the virtue of eliminating operator mixing. The LO coefficient, a^{3S1}_{LO} is determined solely from the $(n', n) = (1, 1)$ matrix element, as higher-order contact-gradient operators do not contribute to this matrix element. Similarly, the NLO coefficient is determined from the $(1, 2)$ matrix element, and NNLO and N³LO results do not affect this value, and so on for each operator.

As in [17], the contact-gradient expansion replaces the expression

$$V \frac{1}{E - QH + i\epsilon} QV. \tag{2.57}$$

There are 2 replacements. The first is in the operator M_{pp},

$$
\begin{aligned}
M_{pp} =& PT \frac{1}{E - QT + i\epsilon} P \\
&+ P \frac{E}{E - TQ + i\epsilon} \left(V + V \frac{1}{E - QH + i\epsilon} QV \right) \frac{1}{E - QT + i\epsilon} P \\
\to & PT \frac{1}{E - QT + i\epsilon} P \\
&+ P \frac{E}{E - TQ + i\epsilon} \left(V + \sum_{i=LO,NLO,...} \bar{O}^i \right) \frac{1}{E - QT + i\epsilon} P. \tag{2.58}
\end{aligned}
$$

The second replacement acts directly on the free state $|\phi>$,

$$P|\psi> = P|\phi> + PMP\frac{1}{E-TQ+i\epsilon}\left(V + V\frac{1}{E-QH+i\epsilon}QV\right)|\phi>$$
$$\rightarrow P|\phi> + PMP\frac{1}{E-TQ+i\epsilon}\left(V + \sum_{i=LO,NLO,...}\bar{O}^i\right)|\phi>. \tag{2.59}$$

2.7   Square-Well Results

Since the square well allows all the matrix elements in the master equation to be written down as a power series in k, we can find the contact-gradient coefficients by matching to the matrix elements of

$$<\tilde{n}'|V\frac{1}{E-QH+i\epsilon}QV|\tilde{n}>. \tag{2.60}$$

The $(n',n)=(1,1)$ matrix element is compared with the $(1,1)$ matrix element of the LO operator (delta function) to determine a_{LO}, then the $(1,2)$ matrix element is compared to that of the NLO operator, etc. One way to see what kind of physics is being captured by the contact-gradient expansion is to examine how the coefficients respond to changes in the square well parameters.

Such a test was performed for the LO coefficient, a_{LO}, by adjusting the radius and strength of the core. For our original potential that reproduces deuteron binding energy ($a_1 = 0.25$fm, $a_2 = 1.7$fm, $V_1 = 40 \times 54.531$MeV, $V_2 = 54.531$MeV), the LO coefficient has the value

$$a_{LO} = -7.58182\text{MeV}. \tag{2.61}$$

Repeating an argument in [17], it is reasonable to assume that rescattering in Q-space effectively results in a potential of the form

$$V_0 e^{-r^2/a^2}, \tag{2.62}$$

having some strength V_0 and range a. Comparing the matrix elements of this potential and of the contact-gradient operators, this ansatz gives the behavior of contact-gradient coefficients as

$$a_{N^n LO} \sim V_0\left(\frac{a^2}{a^2+b^2}\right)^{n+3/2}. \tag{2.63}$$

For the square well with a highly repulsive core, V_0 should be roughly proportional to the height of the core V_1, and a should approximately be the core radius a_1. From this argument, the prediction for a_{LO} is that it is determined by $V_1 a_1^3$ when a_1 is small compared to b.

The results of redefining the radius of the core a_1 to $a_1' = a_1/2$, $a_1/4$, and $a_1/8$ are shown in Figure 2.1. One sees that the LO coefficient a_{LO} depends roughly linearly on the new potential height V_1'. Also, it is interesting to look at the height of the core, V_1, that is required to keep a_{LO} constant. The Figure 2.2 shows the result of this test. As a_1' is taken

to values that are less than $1/100$ of the scattering length b, the LO coefficient is almost entirely determined by the combination $a_1^{'3}V_1^{'}$. This exercise shows that, as predicted, the LO term in the HOBET contact-gradient expansion captures the overall strength of the core, as measured by a^3V_0.

Returning to the original square well potential, contact-gradient coefficients for all terms up to N^{3}LO can be determined by matching the numerically evaluated matrix elements

$$< \tilde{n}'|V\frac{1}{E-QH+i\epsilon}QV|\tilde{n} > . \tag{2.64}$$

Truncating the contact-gradient expansion at each order, the master equation can be solved for $P|\psi>$, and this P-space wavefunction can in turn be used to calculate $k\cot\delta_0$. The ET is working if there is a systematic convergence to the full solution as more terms in the contact-gradient expansion are added.

One problem that we encounter when $k\cot\delta_0$ is extracted from the approximate solution for $P|\psi>$ is that $k\cot\delta_0$ is seen to contain odd powers of k with small imaginary coefficients. For example, the effective range expansion calculated using the contact-gradient series truncated at LO is

$$k\cot\delta_0 \approx -0.479119 + 3.43056 \times 10^{-3}ik + 0.315273k^2 + 6.91152 \times 10^{-4}ik^3$$
$$+ 0.0105398k^4 + 7.57313 \times 10^{-5}ik^5 + 0.000338711k^6. \tag{2.65}$$

Since local, real potentials lead to $k\cot\delta_0$ that is real and contains only even powers of k, it would be an indication of a serious mistake if replacing $V(r)$ by a delta function introduced such imaginary parts. But that is not what was done in our calculation. What was replaced by the contact-gradient expansion was not $V(r)$, but the non-local interaction

$$< \tilde{n}'|V\frac{1}{E-QH+i\epsilon}QV|\tilde{n} > . \tag{2.66}$$

The imaginary parts appear to be a consequence of approximating this in a way that does not ensure that the resulting wavefunction is real. In tests with smaller P-spaces, it was shown that approximating (2.66) by a contact-gradient expansion continues to introduce odd powers of k in the final answer, until all matrix elements have been matched and the contact-gradient expansion is identical to (2.66).

What may be a more serious problem is that there are 2 places in the master equation where the contact gradient expansion is inserted (see eqs. (2.58) and (2.59)), and matrix elements that are being replaced by the contact-gradient operators are not identical in the 2 cases. Matching to the matrix elements of (2.66) was motivated by the replacement in eq. (2.58), and the contact-gradient expansion would reproduce these matrix elements exactly if we used enough operators to account for all independent matrix elements. However, the matrix elements that are being replaced in eq. (2.59) have the form

$$< \tilde{n}'|V\frac{1}{E-QH+i\epsilon}QV|\phi >, \tag{2.67}$$

Figure 2.1: a_{LO} plotted against V_1'/V_2, for 3 different values of the core radius a_1'. The dashed line is the value of a_{LO} for the original potential.

(a) $a_1' = a_1/2$

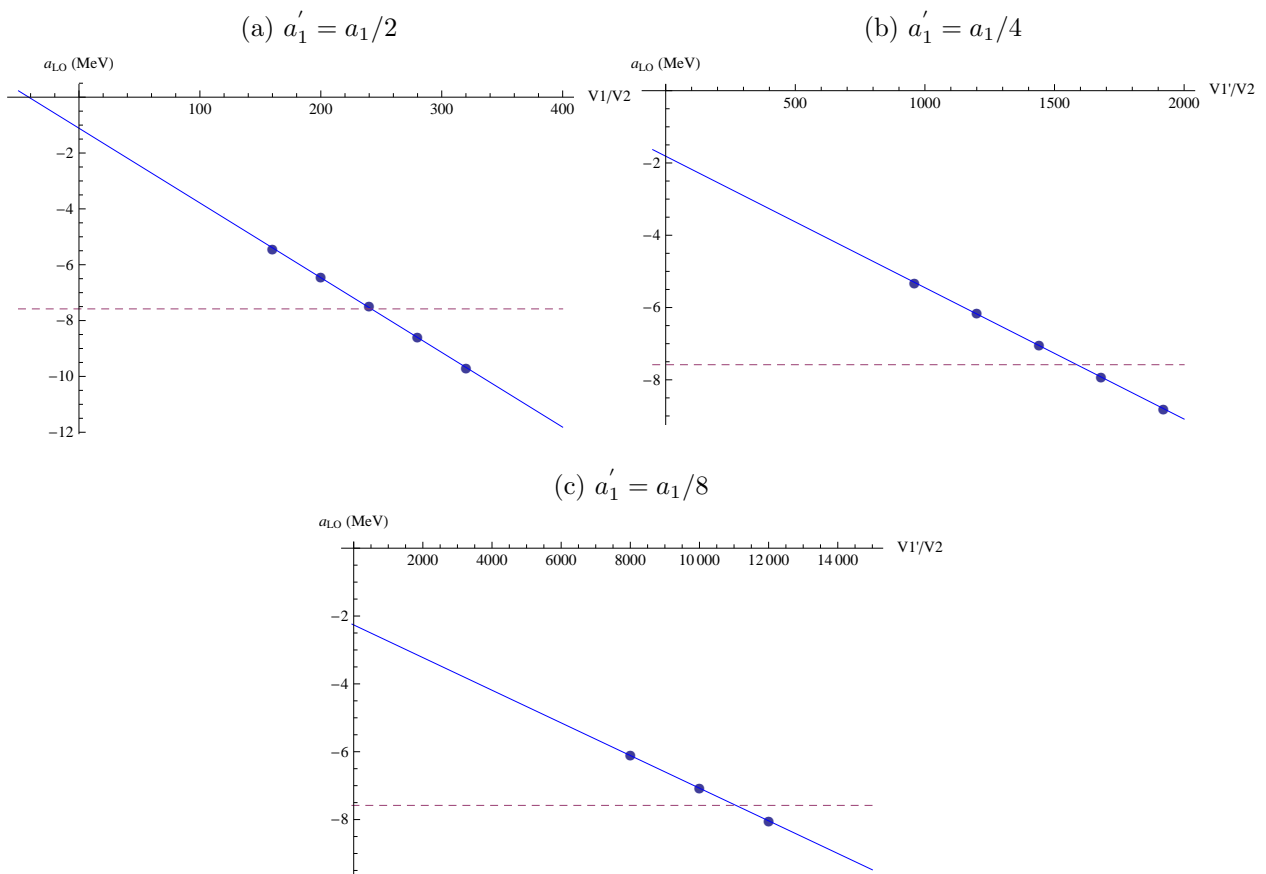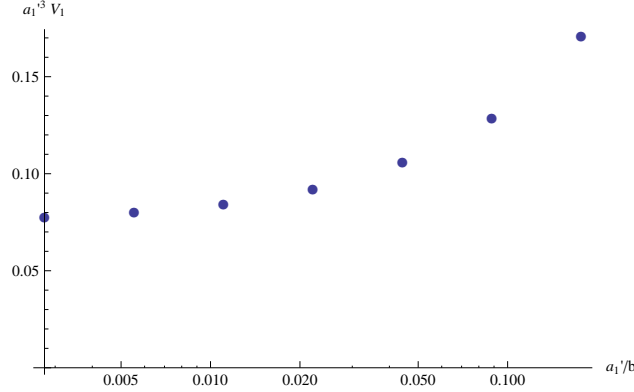(b) $a_1' = a_1/4$

(c) $a_1' = a_1/8$

Figure 2.2: Potential height of the core, V_1', that is required to maintain the same LO contact-gradient coefficient as the core radius a_1' was halved repeatedly from its original value of 0.25 fm. The potential height is plotted as $a_1'^3 V_1'$ in units of $b^3 \hbar \omega$, which is expected to be invariant for small a_1'.

which is not the same as (2.66). Since our procedure preferentially treats (2.66) as the matrix elements to be reproduced, we encounter errors in approximating (2.67). In the future, this error must either be eliminated by adjusting the procedure, or its value must be quantified so that reliable error bars to the results of our calculations can be given.

2.8   Conclusions

This work was motivated by the conventional method of generating effective NN interactions. First, a phenomenological potential is created from experimental data. Then the high-momentum part of the potential is integrated out to produce a soft potential for use in the HO basis. HOBET is an attempt to bypass the intermediate step, arriving at the effective interaction immediately from experimental data.

In [17], it was shown that analytical treatment of the repeated summation of T and representation of the short-range interaction by contact-gradient operators can produce an effective interaction with high accuracy, with errors of order 1keV. This showed that an effective interaction in HO basis can be constructed without referencing high-momentum components to the potential, and suggested the possibility of going directly from data to ET. In this dissertation, it was demonstrated using a toy model that HOBET can reproduce the phase shift with increasing accuracy as higher order contact-gradient operators are added.

The next step in this project is to solve the reverse problem of finding the low-energy coefficients, given phase shift information. Despite the promising result of order-by-order improvement, the approach taken here has two problems: there are two sets of contact-gradient series in the master equation, and the contact gradient series from ET contains odd powers of k. Possibilities for evading these problems will be explored in the future. This may

Figure 2.3: $k \cot \delta_0$ at different orders in the HOBET contact-gradient expansion. The plots show only the real parts of the effective range expansion. HOBET results contain small imaginary parts (see text for discussion). In (a), solid line is the exact solution, dashed line comes from eliminating the contact-gradient expansion, dotted line comes from truncating at LO, dot-dashed line from truncating at NLO. In (b), the solid line is NLO, dashed line NNLO, and dotted line N^3LO.

(a) $k \cot \delta_0$ obtained using contact-gradient series truncated at no terms, LO, and NLO, plotted with the exact answer.
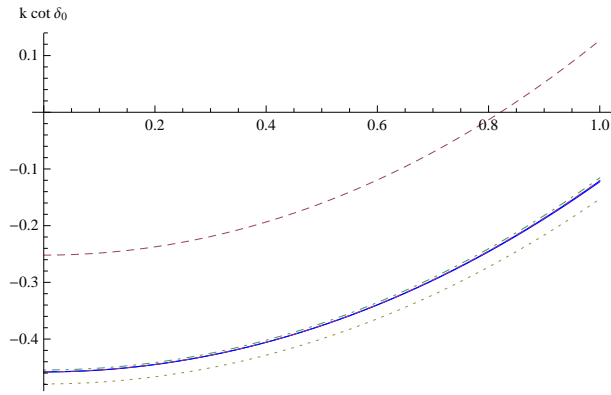
(b) The difference between the NLO, NNLO, and N^3LO solutions to $k \cot \delta_0$ from the exact solution.
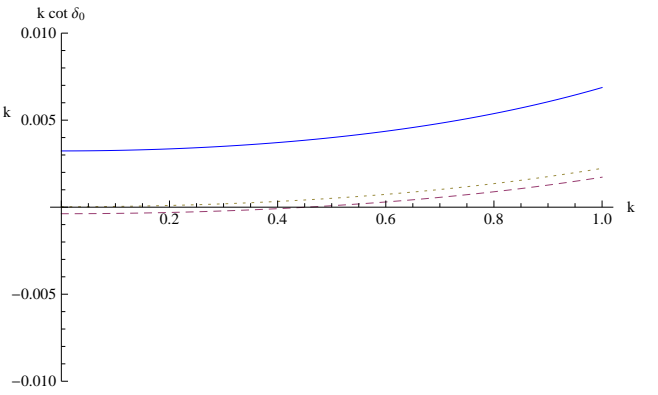
be in the framework of the current approach, or it may involve solving the BH equation, as in [17], or another similar equation.

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Appendix A

Atomic Electric Dipole Moment

A.1   Multipole Analysis

We expand the Breit interaction

$$
V_{eN} + V_{e\tilde{N}} = -\alpha \iint d^3x d^3y \left[\frac{\rho_e(\vec{x})\rho_N(\vec{y})}{|\vec{x}-\vec{y}|} \right.
$$
$$
\left. -\frac{1}{2} \left(\frac{\vec{j}_e(\vec{x}) \odot \vec{j}_N(\vec{y})}{|\vec{x}-\vec{y}|} + \frac{\vec{j}_e(\vec{x}) \odot (\vec{x}-\vec{y}) \, \vec{j}_N(\vec{y}) \odot (\vec{x}-\vec{y})}{|\vec{x}-\vec{y}|^3} \right) \right], \quad \text{(A.1)}
$$

in terms of electronic and nuclear multipoles.

A.1.1   Charge-Charge Interaction

First, consider the charge-charge term. The spherical expansion of $1/|\vec{x}-\vec{y}|$ is

$$
\begin{aligned}
\frac{1}{|\vec{x}-\vec{y}|} =& 4\pi \sum_{lm} \frac{1}{2l+1} \frac{r_<^l}{r_>^{l+1}} Y_{lm}^*(\Omega_x) Y_{lm}(\Omega_y) \\
=& 4\pi \sum_l \frac{1}{2l+1} \frac{y^l}{x^{l+1}} Y_l(\Omega_x) \odot Y_l(\Omega_y) \\
& + \theta(y-x) 4\pi \sum_l \frac{1}{2l+1} \left(\frac{x^l}{y^{l+1}} - \frac{y^l}{x^{l+1}} \right) Y_l(\Omega_x) \odot Y_l(\Omega_y). \quad \text{(A.2)}
\end{aligned}
$$

Simply substituting,

$$
\begin{aligned}
&-\alpha \iint d^3x\, d^3y\, \frac{\rho_e(\vec{x})\rho_N(\vec{y})}{|\vec{x}-\vec{y}|} \\
&= -4\pi\alpha \sum_l \frac{1}{2l+1} \iint d^3x\, d^3y \left(\left[\frac{1}{x^{l+1}} \rho_e(\vec{x}) Y_l(\Omega_x) \right] \odot \left[y^l \rho_N(\vec{y}) Y_l(\Omega_y) \right] \right. \\
&\quad + \theta(y-x) \left\{ \left[x^l \rho_e(\vec{x}) Y_l(\Omega_x) \right] \odot \left[\frac{1}{y^{l+1}} \rho_N(\vec{y}) Y_l(\Omega_y) \right] \right. \\
&\quad \left.\left. - \left[\frac{1}{x^{l+1}} \rho_e(\vec{x}) Y_l(\Omega_x) \right] \odot \left[y^l \rho_N(\vec{y}) Y_l(\Omega_y) \right] \right\} \right).
\end{aligned}
\tag{A.3}
$$

Here, it is convenient to define charge multipoles

$$
C_{lm}^N \equiv \int d^3y \left(\frac{y}{R_N} \right)^l Y_{lm}(\Omega_y) \rho_N(\vec{y})
$$

$$
C_{lm}^A \equiv \int d^3x \left(\frac{R_A}{x} \right)^{l+1} Y_{lm}(\Omega_x) \rho_e(\vec{y}) = \sum_{i=1}^{Z} \left(\frac{R_A}{x_i} \right)^{l+1} Y_{lm}(\Omega_i)
\tag{A.4}
$$

for the first term in eq. (A.3), and

$$
C_l^{A+}(i) \odot C_l^{N-}(i) = \left(\frac{x_i}{R_N} \right)^l Y_l(\Omega_i) \odot \int d^3y\, \theta(y-x_i) \left(\frac{R_N}{y} \right)^{l+1} Y_l(\Omega_y) \rho_N(\vec{y})
$$

$$
C_l^{A-}(i) \odot C_l^{N+}(i) = \left(\frac{R_N}{x_i} \right)^{l+1} Y_l(\Omega_i) \odot \int d^3y\, \theta(y-x_i) \left(\frac{y}{R_N} \right)^l Y_l(\Omega_y) \rho_N(\vec{y})
\tag{A.5}
$$

for the penetration terms. Using these, eq. (A.3) can be written as

$$
\begin{aligned}
V_{\text{charge}} &= -\alpha \iint d^3x\, d^3y\, \frac{\rho_e(\vec{x})\rho_N(\vec{y})}{|\vec{x}-\vec{y}|} \\
&= -\frac{4\pi\alpha}{R_A} \sum_l \frac{1}{2l+1} \left(\frac{R_N}{R_A} \right)^l C_l^A \odot C_l^N \\
&\quad - \frac{4\pi\alpha}{R_N} \sum_l \sum_i^Z \left[C_l^{A+}(i) \odot C_l^{N-}(i) - C_l^{A-}(i) \odot C_l^{N+}(i) \right]
\end{aligned}
\tag{A.6}
$$

A.1.2 Current-Current Interaction

Next, consider the current-current part of the interaction. First, expand as in the charge-charge case:

$$
\frac{\vec{j}_e(\vec{x}) \odot \vec{j}_N(\vec{y})}{|\vec{x}-\vec{y}|} = 4\pi \sum_l \left[\frac{1}{2l+1}\frac{y^l}{x^{l+1}} + \theta(y-x)\frac{1}{2l+1}\left(\frac{x^l}{y^{l+1}} - \frac{y^l}{x^{l+1}}\right) \right]
$$
$$
\times \left(\vec{j}_e(\vec{x}) \odot \vec{j}_N(\vec{y})\right)\left(Y_l(\Omega_x) \odot Y_l(\Omega_y)\right). \tag{A.7}
$$

We want to separate the electronic and nuclear coordinates so that we can define multipoles for each. This can be achieved using 9-j symbols (coordinates are suppressed).

$$
\left(\vec{j}_e \odot \vec{j}_N\right)\left(Y_l(\Omega_x) \odot Y_l(\Omega_y)\right)
$$
$$
= (-1)^{l+1}[1][l]\left[\left[\vec{j}_e \otimes \vec{j}_N\right]_0 \otimes \left[Y_l(\Omega_x) \otimes Y_l(\Omega_y)\right]_0\right]_{00}
$$
$$
= (-1)^{l+1}[1][l]\sum_{l'}\left[\left[\vec{j}_e \otimes Y_l(\Omega_x)\right]_{l'} \otimes \left[\vec{j}_N \otimes Y_l(\Omega_y)\right]_{l'}\right]_{00}[0]^2[l']^2 \left\{\begin{array}{ccc} 1 & 1 & 0 \\ l & l & 0 \\ l' & l' & 0 \end{array}\right\}
$$
$$
= (-1)^{l+1}[1][l]\sum_{l'}[l']^2\left[\left[\vec{j}_e \otimes Y_l(\Omega_x)\right]_{l'} \otimes \left[\vec{j}_N \otimes Y_l(\Omega_y)\right]_{l'}\right]_{00} \times \left(\frac{1}{[1][l][l']}\right)
$$
$$
= (-1)^{l+1}\sum_{l'}[l']\left[\left[\vec{j}_e \otimes Y_l(\Omega_x)\right]_{l'} \otimes \left[\vec{j}_N \otimes Y_l(\Omega_y)\right]_{l'}\right]_{00}
$$
$$
= (-1)^{l+1}\sum_{l'}(-1)^{l'}\left[\vec{j}_e \otimes Y_l(\Omega_x)\right]_{l'} \odot \left[\vec{j}_N \otimes Y_l(\Omega_y)\right]_{l'}
$$
$$
= \left[\vec{j}_e \otimes Y_l(\Omega_x)\right]_{l-1} \odot \left[\vec{j}_N \otimes Y_l(\Omega_y)\right]_{l-1}
$$
$$
- \left[\vec{j}_e \otimes Y_l(\Omega_x)\right]_l \odot \left[\vec{j}_N \otimes Y_l(\Omega_y)\right]_l
$$
$$
+ \left[\vec{j}_e \otimes Y_l(\Omega_x)\right]_{l+1} \odot \left[\vec{j}_N \otimes Y_l(\Omega_y)\right]_{l+1}. \tag{A.8}
$$

Finally, consider the last term in eq. (A.1). Note that

$$
\vec{\nabla}\frac{1}{r} = -\frac{\vec{r}}{r^3}. \tag{A.9}
$$

This gives us the relation

$$
\frac{\vec{j}_e(\vec{x}) \odot (\vec{x}-\vec{y})\, \vec{j}_N(\vec{y}) \odot (\vec{x}-\vec{y})}{|\vec{x}-\vec{y}|^3} = \left(\vec{j}_e(\vec{x}) \odot \vec{x}\right)\left(\vec{j}_N(\vec{y}) \odot \vec{\nabla}_y\right)\frac{1}{|\vec{x}-\vec{y}|}
$$
$$
+ \left(\vec{j}_e(\vec{x}) \odot \vec{y}\right)\left(\vec{j}_N(\vec{y}) \odot \vec{\nabla}_x\right)\frac{1}{|\vec{x}-\vec{y}|}. \tag{A.10}
$$

To separate the x- and y-dependent parts, use the following relations:

$$
\left[\vec{j_e} \otimes \vec{\nabla}_x\right]_0 \odot \left[\vec{j_N} \otimes \vec{y}\right]_0 = \frac{1}{3}\left(\vec{j_e} \odot \vec{\nabla}_x\right)\left(\vec{j_N} \odot \vec{y}\right)
$$

$$
\left[\vec{j_e} \otimes \vec{\nabla}_x\right]_1 \odot \left[\vec{j_N} \otimes \vec{y}\right]_1 = -\sqrt{3}\left[\left[\vec{j_e} \otimes \vec{\nabla}_x\right]_1 \otimes \left[\vec{j_N} \otimes \vec{y}\right]_1\right]_{00}
$$

$$
= \frac{1}{2}\left(\vec{j_e} \odot \vec{y}\right)\left(\vec{\nabla}_x \odot \vec{j_N}\right) - \frac{1}{2}\left(\vec{j_e} \odot \vec{j_N}\right)\left(\vec{\nabla}_x \odot \vec{y}\right)
$$

$$
\left[\vec{j_e} \otimes \vec{\nabla}_x\right]_2 \odot \left[\vec{j_N} \otimes \vec{y}\right]_2 = \sqrt{5}\left[\left[\vec{j_e} \otimes \vec{\nabla}_x\right]_2 \otimes \left[\vec{j_N} \otimes \vec{y}\right]_2\right]_{00}
$$

$$
= \frac{1}{2}\left(\vec{j_e} \odot \vec{y}\right)\left(\vec{\nabla}_x \odot \vec{j_N}\right) + \frac{1}{2}\left(\vec{j_e} \odot \vec{j_N}\right)\left(\vec{\nabla}_x \odot \vec{y}\right)
$$

$$
- \frac{1}{3}\left(\vec{j_e} \odot \vec{\nabla}_x\right)\left(\vec{j_N} \odot \vec{y}\right). \tag{A.11}
$$

Eq. (A.10) can now be written

$$
\frac{\vec{j_e} \odot (\vec{x} - \vec{y})\, \vec{j_N} \odot (\vec{x} - \vec{y})}{|\vec{x} - \vec{y}|^3}
$$

$$
= \left(3\left[\vec{j_e} \otimes \vec{x}\right]_0 \odot \left[\vec{j_N} \otimes \vec{\nabla}_y\right]_0 + \sum_L \left[\vec{j_e} \otimes \vec{\nabla}_x\right]_L \odot \left[\vec{j_N} \otimes \vec{y}\right]_L\right)\frac{1}{|\vec{x} - \vec{y}|}, \tag{A.12}
$$

where L runs from 0 to 2.

Consider the first term in eq. (A.12), and expand the denominator as before.

$$
3\left[\vec{j_e} \otimes \vec{x}\right]_0 \odot \left[\vec{j_N} \otimes \vec{\nabla}_y\right]_0 \frac{1}{|\vec{x} - \vec{y}|} = \left(\vec{j_e} \odot \vec{x}\right)\left(\vec{j_N} \odot \vec{\nabla}_y\right)\sum_l \frac{4\pi}{2l+1}\frac{r_<^l}{r_>^{l+1}}Y_l(\Omega_x) \odot Y_l(\Omega_y) \tag{A.13}
$$

First, assume $x > y$, and the expression for the $x < y$ case follows from symmetry.

$$
\left[\left(\vec{j_e} \odot \vec{x}\right)\left(\vec{j_N} \odot \vec{\nabla}_y\right)\sum_l \frac{4\pi}{2l+1}\frac{y^l}{x^{l+1}}Y_l(\Omega_x) \odot Y_l(\Omega_y)\right]_{x>y}
$$

$$
= \sum_{lm}(-1)^m \frac{4\pi}{2l+1}\left[\vec{j_e} \cdot x^{-l}\left(-\sqrt{\frac{l+1}{2l+1}}\vec{\mathcal{Y}}_{l,l+1,1}^m(\Omega_x) + \sqrt{\frac{l}{2l+1}}\vec{\mathcal{Y}}_{l,l-1,1}^m(\Omega_x)\right)\right]
$$

$$
\times \vec{j_N} \cdot \left(\sqrt{l(2l+1)}y^{l-1}\vec{\mathcal{Y}}_{l,l-1,1}^m(\Omega_y)\right)
$$

$$
= \sum_l \frac{4\pi}{2l+1}\frac{y^{l-1}}{x^l}\left(-\sqrt{l(l+1)}\left[Y_{l+1}(\Omega_x) \otimes \vec{j_e}\right]_l + l\left[Y_{l-1}(\Omega_x) \otimes \vec{j_e}\right]_l\right)
$$

$$
\odot \left[Y_{l-1}(\Omega_y) \otimes \vec{j_N}\right]_l. \tag{A.14}
$$

Consider the second term in eq. (A.12). For this, rewrite the tensor part first as follows:

$$\sum_{L,l}\left(\left[\vec{j}_e\otimes\overrightarrow{\nabla}_x\right]_L\odot\left[\vec{j}_N\otimes\vec{y}\right]_L\right)(Y_l(\Omega_x)Y_l(\Omega_y))$$

$$=\sum_{L,l}(-1)^{L+l}[L][l]\left[\left[\left[\vec{j}_e\otimes\overrightarrow{\nabla}_x\right]_L\otimes\left[\vec{j}_N\otimes\vec{y}\right]_L\right]_0\otimes[Y_l(\Omega_x)\otimes Y_l(\Omega_y)]_0\right]_{00}$$

$$=\sum_{L,l}(-1)^{L+l}[L][l]\sum_{l'}\left[\left[\left[\vec{j}_e\otimes\overrightarrow{\nabla}_x\right]_L\otimes Y_l(\Omega_x)\right]_{l'}\otimes\left[\left[\vec{j}_N\otimes\vec{y}\right]_L\otimes Y_l(\Omega_y)\right]_{l'}\right]_{00}$$

$$\times[0]^2[l']^2\left\{\begin{array}{ccc}L & L & 0\\ l & l & 0\\ l' & l' & 0\end{array}\right\}$$

$$=\sum_{L,l,l'}(-1)^{L+l+l'}\left[\left[\vec{j}_e\otimes\overrightarrow{\nabla}_x\right]_L\otimes Y_l(\Omega_x)\right]_{l'}\odot\left[\left[\vec{j}_N\otimes\vec{y}\right]_L\otimes Y_l(\Omega_y)\right]_{l'}. \qquad\text{(A.15)}$$

Now, including the factors of x and y,

$$\left[\sum_L\left[\vec{j}_e\otimes\overrightarrow{\nabla}_x\right]_L\odot\left[\vec{j}_N\otimes\vec{y}\right]_L\frac{1}{|\vec{x}-\vec{y}|}\right]_{x>y}$$

$$=\sum_{L,l,l'}(-1)^{L+l+l'}\frac{4\pi}{2l+1}\left[\left[\vec{j}_e\otimes\overrightarrow{\nabla}_x\right]_L\otimes Y_l(\Omega_x)\right]_{l'}\odot\left[\left[\vec{j}_N\otimes\vec{y}\right]_L\otimes Y_l(\Omega_y)\right]_{l'}\frac{y^l}{x^{l+1}}. \qquad\text{(A.16)}$$

We will look at the x- and y-dependent parts separately, then combine the results. The

x-dependent part is

$$
\left[\left[\vec{j}_e \otimes \vec{\nabla}_x\right]_L \otimes Y_l(\Omega_x)\right]_{l'm'} x^{-l-1}
$$

$$
= \sum_{M,m,m_1,m_2} <LMlm|l'm'><1m_1 1m_2|LM> j_{e,m_1} \nabla_{x,m_2}\left(x^{-l-1}Y_{lm}(\Omega_x)\right)
$$

$$
= \sum_{M,m,m_1,m_2} <LMlm|l'm'><1m_1 1m_2|LM>
$$

$$
\times j_{e,m_1}\hat{e}_{m_2} \cdot \left(\sqrt{(l+1)(2l+1)}x^{-l-2}\vec{\mathcal{Y}}^m_{l,l+1,1}(\Omega_x)\right)
$$

$$
= \sum_{M,m,m_1,m_2,q} (-1)^{m_2} <LMlm|l'm'><1m_1 1m_2|LM><l+1,q,1,-m_2|lm>
$$

$$
\times j_{e,m_1}\left(\sqrt{(l+1)(2l+1)}x^{-l-2}Y_{l+1,q}(\Omega_x)\right)
$$

$$
= -\sum_{m_1,q}[L][l]<l+1,q,1,m_1|l'm'> \left\{ \begin{array}{ccc} l' & l+1 & 1 \\ 1 & L & l \end{array} \right\}
$$

$$
\times j_{e,m_1}\left(\sqrt{(l+1)(2l+1)}x^{-l-2}Y_{l+1,q}(\Omega_x)\right)
$$

$$
= -[L][l]\sqrt{(l+1)(2l+1)} \left\{ \begin{array}{ccc} l' & l+1 & 1 \\ 1 & L & l \end{array} \right\} x^{-l-2}\left[Y_{l+1}(\Omega_x) \otimes \vec{j}_e\right]_{l'm'}. \qquad (A.17)
$$

Evaluating the y-dependent part,

$$
\left[\left[\vec{j}_N \otimes \vec{y}\right]_L \otimes Y_l(\Omega_y)\right]_{l'm'}
$$

$$
= \sum_{M,m,m_1,m_2} <LMlm|l'm'><1m_1 1m_2|LM>
$$

$$
\times j_{N,m_1}y\hat{e}_{m_2} \cdot \left[-\sqrt{\frac{l+1}{2l+1}}\vec{\mathcal{Y}}^m_{l,l+1,1}(\Omega_y) + \sqrt{\frac{l}{2l+1}}\vec{\mathcal{Y}}^m_{l,l-1,1}(\Omega_y)\right]
$$

$$
= -[L]\left(-\sqrt{l+1}\left\{ \begin{array}{ccc} l' & l+1 & 1 \\ 1 & L & l \end{array} \right\} y\left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N\right]_{l'm'}\right.
$$

$$
\left. +\sqrt{l}\left\{ \begin{array}{ccc} l' & l-1 & 1 \\ 1 & L & l \end{array} \right\} y\left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N\right]_{l'm'}\right). \qquad (A.18)
$$

Combining eqs. (A.17) and (A.18),

$$\sum_{L,l,l'} (-1)^{L+l+l'} \frac{4\pi}{2l+1} \left[\left[\vec{j_e} \otimes \vec{\nabla}_x \right]_L \otimes Y_l(\Omega_x) \right]_{l'} \odot \left[\left[\vec{j_N} \otimes \vec{y} \right]_L \otimes Y_l(\Omega_y) \right]_{l'} \frac{y^l}{x^{l+1}}$$

$$= \sum_{L,l,l'} (-1)^{L+l+l'+1} \frac{4\pi}{2l+1} \frac{y^{l+1}}{x^{l+2}}$$

$$\times (-1)[L][l]\sqrt{(l+1)(2l+1)} \left\{ \begin{matrix} l' & l+1 & 1 \\ 1 & L & l \end{matrix} \right\} \left[Y_{l+1}(\Omega_x) \otimes \vec{j_e} \right]_{l'}$$

$$\odot (-1)[L] \left(\sqrt{l+1} \left\{ \begin{matrix} l' & l+1 & 1 \\ 1 & L & l \end{matrix} \right\} \left[Y_{l+1}(\Omega_y) \otimes \vec{j_N} \right]_{l'} \right.$$

$$\left. -\sqrt{l} \left\{ \begin{matrix} l' & l-1 & 1 \\ 1 & L & l \end{matrix} \right\} \left[Y_{l-1}(\Omega_y) \otimes \vec{j_N} \right]_{l'} \right)$$

$$= \sum_{L,l,l'} (-1)^{L+l+l'+1} (4\pi)[L]^2 \frac{y^{l+1}}{x^{l+2}} \left\{ \begin{matrix} l' & l+1 & 1 \\ 1 & L & l \end{matrix} \right\} \left[Y_{l+1}(\Omega_x) \otimes \vec{j_e} \right]_{l'}$$

$$\odot \left((l+1) \left\{ \begin{matrix} l' & l+1 & 1 \\ 1 & L & l \end{matrix} \right\} \left[Y_{l+1}(\Omega_y) \otimes \vec{j_N} \right]_{l'} \right.$$

$$\left. -\sqrt{l(l+1)} \left\{ \begin{matrix} l' & l-1 & 1 \\ 1 & L & l \end{matrix} \right\} \left[Y_{l-1}(\Omega_y) \otimes \vec{j_N} \right]_{l'} \right)$$

$$= \sum_{l,l'} (-1)^{l+l'+1} (4\pi) \frac{y^{l+1}}{x^{l+2}} \left[Y_{l+1}(\Omega_x) \otimes \vec{j_e} \right]_{l'}$$

$$\odot \left((l+1) \left\{ \begin{matrix} 1 & l & l+1 \\ 1 & l' & l+1 \end{matrix} \right\} \left[Y_{l+1}(\Omega_y) \otimes \vec{j_N} \right]_{l'} \right.$$

$$\left. -\sqrt{l(l+1)} \left\{ \begin{matrix} 1 & l & l-1 \\ 1 & l' & l+1 \end{matrix} \right\} \left[Y_{l-1}(\Omega_y) \otimes \vec{j_N} \right]_{l'} \right). \tag{A.19}$$

$l' = l, l+1, l+2$ are possible for the first term, and only $l' = l$ is allowed for the second

term.

$$
\sum_{L,l,l'}(-1)^{L+l+l'}\frac{4\pi}{2l+1}\left[\left[\vec{j}_e\otimes\vec{\nabla}_x\right]_L\otimes Y_l(\Omega_x)\right]_{l'}\odot\left[\left[\vec{j}_N\otimes\vec{y}\right]_L\otimes Y_l(\Omega_y)\right]_{l'}\frac{y^l}{x^{l+1}}
$$

$$
=\sum_l(4\pi)\frac{y^{l+1}}{x^{l+2}}
$$

$$
\times\left\{-(l+1)\frac{1}{(l+1)(2l+1)(2l+3)}\left[Y_{l+1}(\Omega_x)\otimes\vec{j}_e\right]_l\odot\left[Y_{l+1}(\Omega_y)\otimes\vec{j}_N\right]_l\right.
$$

$$
+(l+1)\left[-\frac{1}{(l+1)(2l+3)}\right]\left[Y_{l+1}(\Omega_x)\otimes\vec{j}_e\right]_{l+1}\odot\left[Y_{l+1}(\Omega_y)\otimes\vec{j}_N\right]_{l+1}
$$

$$
-(l+1)\frac{1}{2l+3}\left[Y_{l+1}(\Omega_x)\otimes\vec{j}_e\right]_{l+2}\odot\left[Y_{l+1}(\Omega_y)\otimes\vec{j}_N\right]_{l+2}
$$

$$
\left.-\sqrt{l(l+1)}\frac{1}{2l+1}\left[Y_{l+1}(\Omega_x)\otimes\vec{j}_e\right]_l\odot\left[Y_{l-1}(\Omega_y)\otimes\vec{j}_N\right]_l\right\}
$$

$$
=\sum_l\frac{4\pi}{2l+1}\frac{y^{l+1}}{x^{l+2}}\left\{-\frac{1}{2l+3}\left[Y_{l+1}(\Omega_x)\otimes\vec{j}_e\right]_l\odot\left[Y_{l+1}(\Omega_y)\otimes\vec{j}_N\right]_l\right.
$$

$$
-\frac{2l+1}{2l+3}\left[Y_{l+1}(\Omega_x)\otimes\vec{j}_e\right]_{l+1}\odot\left[Y_{l+1}(\Omega_y)\otimes\vec{j}_N\right]_{l+1}
$$

$$
-\frac{(l+1)(2l+1)}{2l+3}\left[Y_{l+1}(\Omega_x)\otimes\vec{j}_e\right]_{l+2}\odot\left[Y_{l+1}(\Omega_y)\otimes\vec{j}_N\right]_{l+2}
$$

$$
\left.+\sqrt{l(l+1)}\left[Y_{l+1}(\Omega_x)\otimes\vec{j}_e\right]_l\odot\left[Y_{l-1}(\Omega_y)\otimes\vec{j}_N\right]_l\right\}\tag{A.20}
$$

To get the full expression for the current-current interaction in the point-nucleus limit,

use eqs. (A.8), (A.14), and (A.20).

$$\left[\frac{\vec{j}_e(\vec{x}) \odot \vec{j}_N(\vec{y})}{|\vec{x}-\vec{y}|} + \frac{\vec{j}_e(\vec{x}) \odot (\vec{x}-\vec{y})\,\vec{j}_N(\vec{y}) \odot (\vec{x}-\vec{y})}{|\vec{x}-\vec{y}|^3}\right]_{x>y}$$

$$= \left[\sum_{l=1}^{\infty} \frac{4\pi}{2l+1} \frac{y^l}{x^{l+1}} \left[\vec{j}_e \otimes Y_l(\Omega_x)\right]_{l-1} \odot \left[\vec{j}_N \otimes Y_l(\Omega_y)\right]_{l-1}\right.$$

$$-\sum_{l=0}^{\infty} \frac{4\pi}{2l+1} \frac{y^l}{x^{l+1}} \left[\vec{j}_e \otimes Y_l(\Omega_x)\right]_{l} \odot \left[\vec{j}_N \otimes Y_l(\Omega_y)\right]_{l}$$

$$\left.+\sum_{l=0}^{\infty} \frac{4\pi}{2l+1} \frac{y^l}{x^{l+1}} \left[\vec{j}_e \otimes Y_l(\Omega_x)\right]_{l+1} \odot \left[\vec{j}_N \otimes Y_l(\Omega_y)\right]_{l+1}\right]$$

$$+\left[-\sum_{l=1}^{\infty} \frac{4\pi}{2l+1} \frac{y^{l-1}}{x^l} \sqrt{l(l+1)} \left[Y_{l+1}(\Omega_x) \otimes \vec{j}_e\right]_{l} \odot \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N\right]_{l}\right.$$

$$\left.+\sum_{l=1}^{\infty} \frac{4\pi}{2l+1} \frac{y^{l-1}}{x^l} l \left[Y_{l-1}(\Omega_x) \otimes \vec{j}_e\right]_{l} \odot \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N\right]_{l}\right]$$

$$+\left[-\sum_{l=0}^{\infty} \frac{4\pi}{2l+1} \frac{y^{l+1}}{x^{l+2}} \frac{1}{2l+3} \left[Y_{l+1}(\Omega_x) \otimes \vec{j}_e\right]_{l} \odot \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N\right]_{l}\right.$$

$$-\sum_{l=0}^{\infty} \frac{4\pi}{2l+1} \frac{y^{l+1}}{x^{l+2}} \frac{2l+1}{2l+3} \left[Y_{l+1}(\Omega_x) \otimes \vec{j}_e\right]_{l+1} \odot \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N\right]_{l+1}$$

$$-\sum_{l=0}^{\infty} \frac{4\pi}{2l+1} \frac{y^{l+1}}{x^{l+2}} \frac{(l+1)(2l+1)}{2l+3} \left[Y_{l+1}(\Omega_x) \otimes \vec{j}_e\right]_{l+2} \odot \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N\right]_{l+2}$$

$$\left.+\sum_{l=0}^{\infty} \frac{4\pi}{2l+1} \frac{y^{l+1}}{x^{l+2}} \sqrt{l(l+1)} \left[Y_{l+1}(\Omega_x) \otimes \vec{j}_e\right]_{l} \odot \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N\right]_{l}\right\}$$

$$= \sum_{l=1}^{\infty} \frac{4\pi}{2l+1} \left\{\frac{2l}{2l+3} \frac{y^{l+1}}{x^{l+2}} \left[Y_{l+1}(\Omega_x) \otimes \vec{j}_e\right]_{l} \odot \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N\right]_{l}\right.$$

$$-\frac{4\pi}{2l+1} 2\frac{y^l}{x^{l+1}} \left[Y_l(\Omega_x) \otimes \vec{j}_e\right]_{l} \odot \left[Y_l(\Omega_y) \otimes \vec{j}_N\right]_{l}$$

$$+\frac{2(l+1)}{2l-1} \frac{y^{l-1}}{x^l} \left[Y_{l-1}(\Omega_x) \otimes \vec{j}_e\right]_{l} \odot \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N\right]_{l}$$

$$\left.-\sqrt{l(l+1)}\frac{y^{l-1}}{x^l} \left(1 - \frac{y^2}{x^2}\right) \left[Y_{l+1}(\Omega_x) \otimes \vec{j}_e\right]_{l} \odot \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N\right]_{l}\right\}. \tag{A.21}$$

By symmetry (exchanging $\vec{x} \leftrightarrow \vec{y}$ and $\vec{j}_e \leftrightarrow \vec{j}_N$),

$$
\left[\frac{\vec{j}_e(\vec{x}) \odot \vec{j}_N(\vec{y})}{|\vec{x}-\vec{y}|} + \frac{\vec{j}_e(\vec{x}) \odot (\vec{x}-\vec{y}) \, \vec{j}_N(\vec{y}) \odot (\vec{x}-\vec{y})}{|\vec{x}-\vec{y}|^3}\right]_{x<y}
$$

$$
= \sum_{l=1}^{\infty} \frac{4\pi}{2l+1} \left\{ \frac{2l}{2l+3} \frac{x^{l+1}}{y^{l+2}} \left[Y_{l+1}(\Omega_x) \otimes \vec{j}_e\right]_l \odot \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N\right]_l \right.
$$

$$
- \frac{4\pi}{2l+1} 2 \frac{x^l}{y^{l+1}} \left[Y_l(\Omega_x) \otimes \vec{j}_e\right]_l \odot \left[Y_l(\Omega_y) \otimes \vec{j}_N\right]_l
$$

$$
+ \frac{2(l+1)}{2l-1} \frac{x^{l-1}}{y^l} \left[Y_{l-1}(\Omega_x) \otimes \vec{j}_e\right]_l \odot \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N\right]_l
$$

$$
\left. - \sqrt{l(l+1)} \frac{x^{l-1}}{y^l} \left(1 - \frac{x^2}{y^2}\right) \left[Y_{l-1}(\Omega_x) \otimes \vec{j}_e\right]_l \odot \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N\right]_l \right\}. \tag{A.22}
$$

In order to organize the terms, define transverse electric multipoles

$$
T^N_{\mathrm{el},l} \equiv \frac{1}{R_N^{l-1}} \int d^3y \left[\vec{\nabla} \times \left(y^l \vec{\mathcal{Y}}^m_{ll1}(\Omega_y)\right)\right] \cdot \vec{j}_N(\vec{y})
$$

$$
= i\sqrt{(l+1)(2l+1)} \int d^3y \left(\frac{y}{R_N}\right)^{l-1} \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l
$$

$$
T'^N_{\mathrm{el},l} \equiv \frac{1}{R_N^{l+1}} \int d^3y \left[\vec{\nabla} \times \left(-\frac{y^{l+2}}{2(2l+3)} \vec{\mathcal{Y}}^m_{ll1}(\Omega_y)\right)\right] \cdot \vec{j}_N(\vec{y})
$$

$$
= -i\left\{\frac{1}{2l+3}\sqrt{\frac{l}{2l+1}} \int d^3y \left(\frac{y}{R_N}\right)^{l+1} \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l \right.
$$

$$
\left. + \frac{1}{2}\sqrt{\frac{l+1}{2l+1}} \int d^3y \left(\frac{y}{R_N}\right)^{l+1} \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l \right\}
$$

$$
T^A_{\mathrm{el},l} \equiv R_A^{l+2} \int d^3x \left[\vec{\nabla} \times \left(\frac{1}{x^{l+1}} \vec{\mathcal{Y}}^m_{ll1}(\Omega_x)\right)\right] \cdot \vec{j}_e(\vec{x})
$$

$$
= -i\sqrt{l(2l+1)} \sum_{i=1}^{Z} \left(\frac{R_A}{x_i}\right)^{l+2} [Y_{l+1}(\Omega_i) \otimes \vec{\alpha}_i(\vec{x}_i)]_l
$$

$$
T'^A_{\mathrm{el},l} \equiv R_A^l \int d^3x \left[\vec{\nabla} \times \left(\frac{1}{2(2l-1)} \vec{\mathcal{Y}}^m_{ll1}(\Omega_x)\right)\right] \cdot \vec{j}_e(\vec{x})
$$

$$
= i\left\{-\frac{1}{2}\sqrt{\frac{l}{2l+1}} \sum_{i=1}^{Z} \left(\frac{R_A}{x_i}\right)^l [Y_{l+1}(\Omega_i) \otimes \vec{\alpha}_i(\vec{x}_i)]_l \right.
$$

$$
\left. + \frac{1}{2l-1}\sqrt{\frac{l+1}{2l+1}} \sum_{i=1}^{Z} \left(\frac{R_A}{x_i}\right)^l [Y_{l-1}(\Omega_i) \otimes \vec{\alpha}_i(\vec{x}_i)]_l \right\} \tag{A.23}
$$

and transverse magnetic multipoles

$$T_{\text{mag},l}^N \equiv \frac{1}{R_N^l} \int d^3y \, y^l \vec{\mathcal{Y}}_{ll1}^m(\Omega_y) \cdot \vec{j}_N(\vec{y}) = \int d^3y \left(\frac{y}{R_N}\right)^l \left[Y_l(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l$$

$$T_{\text{mag},l}^A \equiv R_A^{l+1} \int d^3x \, \frac{1}{x^{l+1}} \vec{\mathcal{Y}}_{ll1}^m(\Omega_x) \cdot \vec{j}_e(\vec{x}) \sum_{i=1}^Z \left(\frac{R_A}{x_i}\right)^{l+1} \left[Y_l(\Omega_i) \otimes \vec{\alpha}_i(\vec{x}_i)\right]_l. \qquad \text{(A.24)}$$

With these, the interaction in the point-nucleus limit is

$$\begin{aligned}
V_{eN}^{\text{point}} = \frac{4\pi\alpha}{R_A} &\left[\sum_{l=0}^\infty \frac{1}{2l+1} \left(\frac{R_N}{R_A}\right)^l C_l^A \odot C_l^N \right. \\
&+ \sum_{l=1}^\infty \left\{ \frac{1}{2l+1} \left(\frac{R_N}{R_A}\right)^l T_{\text{mag},l}^A \odot T_{\text{mag},l}^N \right. \\
&+ \left. \left. \frac{1}{2l+1} \left(\frac{R_N}{R_A}\right)^{l+1} T_{\text{el},l}^A \odot T_{\text{el},l}'^N + \frac{1}{2l+1} \left(\frac{R_N}{R_A}\right)^{l-1} T_{\text{el},l}'^A \odot T_{\text{el},l}^N \right\} \right]. \qquad \text{(A.25)}
\end{aligned}$$

For the penetration terms, make the following definitions:

$$\begin{aligned}
T_{\text{mag},l}^{A+}(i) \odot T_{\text{mag},l}^{N-}(i) &= \left(\frac{x_i}{R_N}\right)^l \left[Y_l(\Omega_i) \otimes \vec{\alpha}_i\right]_l \\
&\odot \int d^3y \, \theta(y - x_i) \left(\frac{R_N}{y}\right)^{l+1} \left[Y_l(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l \\
T_{\text{mag},l}^{A-}(i) \odot T_{\text{mag},l}^{N+}(i) &= \left(\frac{R_N}{x_i}\right)^{l+1} \left[Y_l(\Omega_i) \otimes \vec{\alpha}_i\right]_l \\
&\odot \int d^3y \, \theta(y - x_i) \left(\frac{y}{R_N}\right)^l \left[Y_l(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l
\end{aligned}$$

$$T_{\text{el},l}^{A+}(i) \odot T_{\text{el},l}'^{N-}(i) = i\sqrt{(l+1)(2l+1)} \left(\frac{x_i}{R_N}\right)^{l-1} [Y_{l-1}(\Omega_i) \otimes \vec{\alpha}_i]_l$$

$$\odot\, i \left\{ -\frac{1}{2}\sqrt{\frac{l}{2l+1}} \int d^3y\, \theta(y-x_i) \left(\frac{R_N}{y}\right)^l \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l \right.$$

$$\left. +\frac{1}{2l-1}\sqrt{\frac{l+1}{2l+1}} \int d^3y\, \theta(y-x_i) \left(\frac{R_N}{y}\right)^l \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l \right\}$$

$$T_{\text{el},l}^{A-}(i) \odot T_{\text{el},l}'^{N+}(i) = -\,i\sqrt{l(2l+1)} \left(\frac{R_N}{x_i}\right)^{l+2} [Y_{l+1}(\Omega_i) \otimes \vec{\alpha}_i]_l$$

$$\odot\, (-i) \left\{ \frac{1}{2l+3}\sqrt{\frac{l}{2l+1}} \int d^3y\, \theta(y-x_i) \left(\frac{y}{R_N}\right)^{l+1} \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l \right.$$

$$\left. +\frac{1}{2}\sqrt{\frac{l+1}{2l+1}} \int d^3y\, \theta(y-x_i) \left(\frac{y}{R_N}\right)^{l+1} \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l \right\}$$

$$T_{\text{el},l}'^{A+}(i) \odot T_{\text{el},l}^{N-}(i) = -\,i \left\{ \frac{1}{2l+3}\sqrt{\frac{l}{2l+1}} \left(\frac{x_i}{R_N}\right)^{l+1} [Y_{l+1}(\Omega_i) \otimes \vec{\alpha}_i]_l \right.$$

$$\left. +\frac{1}{2}\sqrt{\frac{l+1}{2l+1}} \left(\frac{x_i}{R_N}\right)^{l+1} [Y_{l-1}(\Omega_i) \otimes \vec{\alpha}_i]_l \right\}$$

$$\odot\, (-i)\sqrt{l(2l+1)} \int d^3y\, \theta(y-x_i) \left(\frac{R_N}{y}\right)^{l+2} \left[Y_{l+1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l$$

$$T_{\text{el},l}'^{A-}(i) \odot T_{\text{el},l}^{N+}(i) = i \left\{ \frac{1}{2}\sqrt{\frac{l}{2l+1}} \left(\frac{R_N}{x_i}\right)^l [Y_{l+1}(\Omega_i) \otimes \vec{\alpha}_i]_l \right.$$

$$\left. +\frac{1}{2l-1}\sqrt{\frac{l+1}{2l+1}} \left(\frac{R_N}{x_i}\right)^l [Y_{l-1}(\Omega_i) \otimes \vec{\alpha}_i]_l \right\}$$

$$\odot\, i\sqrt{(l+1)(2l+1)} \int d^3y\, \theta(y-x_i) \left(\frac{y}{R_N}\right)^{l-1} \left[Y_{l-1}(\Omega_y) \otimes \vec{j}_N(\vec{y})\right]_l.$$

$$(A.26)$$

The penetration correction is then

$$
\begin{aligned}
V_{eN}^{\text{pen}} = -\frac{4\pi\alpha}{R_A} \Bigg(& \sum_{l=0}^{\infty} \frac{1}{2l+1} \sum_{i=1}^{Z} \left[C_l^{A+}(i) \odot C_l^{N-}(i) - C_l^{A-}(i) \odot C_l^{N+}(i) \right] \\
& + \sum_{l=1}^{\infty} \Bigg\{ \frac{1}{2l+1} \sum_{i=1}^{Z} \left[T_{\text{mag},l}^{A+}(i) \odot T_{\text{mag},l}^{N-}(i) - T_{\text{mag},l}^{A-}(i) \odot T_{\text{mag},l}^{N+}(i) \right] \\
& + \frac{1}{2l+1} \sum_{i=1}^{Z} \left[T_{\text{el},l}^{A+}(i) \odot T_{\text{el},l}'^{N-}(i) - T_{\text{el},l}^{A-}(i) \odot T_{\text{el},l}'^{N+}(i) \right] \\
& + \frac{1}{2l+1} \sum_{i=1}^{Z} \left[T_{\text{el},l}'^{A+}(i) \odot T_{\text{el},l}^{N-}(i) - T_{\text{el},l}'^{A-}(i) \odot T_{\text{el},l}^{N+}(i) \right] \Bigg\} \Bigg).
\end{aligned}
\tag{A.27}
$$

$V_{eN} + V_{e\tilde{N}}$ can be written

$$
V_{eN} + V_{e\tilde{N}} = V_{eN}^{\text{point}} + V_{eN}^{\text{pen}}
\tag{A.28}
$$

A.2 Penetration Correction

We can put the penetration terms in a more convenient form, when it is directly sandwiched between atomic states. Expanding the electronic wavefunctions in Taylor series

$$
\psi(\vec{x}) \approx \psi(0) + \vec{x} \cdot \vec{\nabla} \psi(\vec{x}),
\tag{A.29}
$$

Evaluating the charge multipoles first, C_0^{A+} and C_0^{A-} become

$$
\begin{aligned}
\int d^3 x_i \left(\frac{x_i}{R_N} \right)^l Y_{lm}(\Omega_i) \rho_e(\vec{x}_i) \theta(y - x_i) \xrightarrow{l=0} & \delta_{l0} \sqrt{4\pi} \int_0^y x_i^2 dx_i \psi_f^*(0) \psi_i(0) \\
= & \delta_{l0} \frac{\sqrt{4\pi} y^3}{3} \psi_f^*(0) \psi_i(0)
\end{aligned}
\tag{A.30}
$$

and

$$
\begin{aligned}
\int d^3 x_i \left(\frac{R_N}{x_i} \right)^{l+1} Y_{lm}(\Omega_i) \rho_e(\vec{x}_i) \theta(y - x_i) \xrightarrow{l=0} & \delta_{l0} \sqrt{4\pi} \int_0^y x_i^2 dx_i \frac{R_N}{x_i} \psi_f^*(0) \psi_i(0) \\
= & \delta_{l0} \frac{\sqrt{4\pi} R_N y^2}{2} \psi_f^*(0) \psi_i(0).
\end{aligned}
\tag{A.31}
$$

Similarly for C_1^{A+} and C_1^{A-},

$$\int d^3x_i \left(\frac{x_i}{R_N}\right)^l Y_{lm}(\Omega_i)\rho_e(\vec{x}_i)\theta(y-x_i)$$

$$\xrightarrow{l=1} \delta_{l1}\int d\Omega_i \int_0^y x_i^2 dx_i \frac{x_i}{R_N} Y_{1m}(\Omega_i)\vec{x}_i \cdot \left[\psi_f^*(0)\overleftarrow{\nabla}_i\psi_i(0) + \psi_f^*(0)\overrightarrow{\nabla}_i\psi_i(0)\right]$$

$$=\delta_{l1}\sqrt{\frac{4\pi}{3}}\frac{y^5}{5R_N}\left[\psi_f^*(0)\overleftarrow{\nabla}_{i,m}\psi_i(0) + \psi_f^*(0)\overrightarrow{\nabla}_{i,m}\psi_i(0)\right] \tag{A.32}$$

and

$$\int d^3x_i \left(\frac{R_N}{x_i}\right)^{l+1} Y_{lm}(\Omega_i)\rho_e(\vec{x}_i)\theta(y-x_i)$$

$$\xrightarrow{l=1} \delta_{l1}\int d\Omega_i \int_0^y x_i^2 dx_i \left(\frac{R_N}{x_i}\right)^2 Y_{1m}(\Omega_i)\vec{x}_i \cdot \left[\psi_f^*(0)\overleftarrow{\nabla}_i\psi_i(0) + \psi_f^*(0)\overrightarrow{\nabla}_i\psi_i(0)\right]$$

$$=\delta_{l1}\sqrt{\frac{4\pi}{3}}\frac{R_N^2 y^2}{2}\left[\psi_f^*(0)\overleftarrow{\nabla}_{i,m}\psi_i(0) + \psi_f^*(0)\overrightarrow{\nabla}_{i,m}\psi_i(0)\right] \tag{A.33}$$

Next, consider the magnetic multipoles:

$$\int d^3x_i \left(\frac{x_i}{R_N}\right)^l \left[Y_l(\Omega_i) \otimes \vec{j}_e(\vec{x}_i)\right]_{lm}\theta(y-x_i)$$

$$\xrightarrow{l=1} \delta_{l1}\sum_{m_1,m_2} <1m_11m_2|1m> \int d\Omega_i \int_0^y x_i^2 dx_i \frac{x_i}{R_N} Y_{1m_1}(\Omega_i)\alpha_{i,m_2}$$

$$\times \vec{x}_i \cdot \left[\psi_f^*(0)\overleftarrow{\nabla}_i\psi_i(0) + \psi_f^*(0)\overrightarrow{\nabla}_i\psi_i(0)\right]$$

$$=\delta_{l1}\sqrt{\frac{4\pi}{3}}\frac{y^5}{5R_N}\left[\psi_f^*(0)\left[\overleftarrow{\nabla}_i \otimes \vec{\alpha}_i\right]_1 \psi_i(0) + \psi_f^*(0)\left[\overrightarrow{\nabla}_i \otimes \vec{\alpha}_i\right]_1 \psi_i(0)\right] \tag{A.34}$$

$$\int d^3x_i \left(\frac{R_N}{x_i}\right)^{l+1} \left[Y_l(\Omega_i) \otimes \vec{j}_e(\vec{x}_i)\right]_{lm}\theta(y-x_i)$$

$$\xrightarrow{l=1} \delta_{l1}\sum_{m_1,m_2} <1m_11m_2|1m> \int d\Omega_i \int_0^y x_i^2 dx_i \left(\frac{R_N}{x_i}\right)^2 Y_{1m_1}(\Omega_i)\alpha_{i,m_2}$$

$$\times \vec{x}_i \cdot \left[\psi_f^*(0)\overleftarrow{\nabla}_i\psi_i(0) + \psi_f^*(0)\overrightarrow{\nabla}_i\psi_i(0)\right]$$

$$=\delta_{l1}\sqrt{\frac{4\pi}{3}}\frac{R_N^2 y^2}{2}\left[\psi_f^*(0)\left[\overleftarrow{\nabla}_i \otimes \vec{\alpha}_i\right]_1 \psi_i(0) + \psi_f^*(0)\left[\overrightarrow{\nabla}_i \otimes \vec{\alpha}_i\right]_1 \psi_i(0)\right] \tag{A.35}$$

Finally, consider the transverse electric terms. For $l = 1$, the contribution from multipoles of the form $[Y_0 \otimes \vec{\alpha}_i]_1$ can be calculated easily using the C_0 results in eqs. (A.30) and

(A.31). The $l = 2$ case requires a new calculation:

$$\int d^3x_i \left(\frac{x_i}{R_N}\right)^{l-1} \left[Y_{l-1}(\Omega_i) \otimes \vec{j}_e(\vec{x}_i)\right]_{lm} \theta(y - x_i)$$

$$\xrightarrow{l=2} \delta_{l2} \sum_{m_1,m_2} <1m_1 1m_2|2m> \int d\Omega_i \int_0^y x_i^2 dx_i \frac{x_i}{R_N} Y_{1m_1}(\Omega_i)\alpha_{i,m_2}$$

$$\times \vec{x}_i \cdot \left[\psi_f^*(0)\overleftarrow{\nabla}_i\psi_i(0) + \psi_f^*(0)\overrightarrow{\nabla}_i\psi_i(0)\right]$$

$$= \delta_{l2}\sqrt{\frac{4\pi}{3}}\frac{y^5}{5R_N}\left[\psi_f^*(0)\left[\overleftarrow{\nabla}_i \otimes \vec{\alpha}_i\right]_2 \psi_i(0) + \psi_f^*(0)\left[\overrightarrow{\nabla}_i \otimes \vec{\alpha}_i\right]_2 \psi_i(0)\right], \qquad (A.36)$$

$$\int d^3x_i \left(\frac{x_i}{R_N}\right)^{l+1} \left[Y_{l-1}(\Omega_i) \otimes \vec{j}_e(\vec{x}_i)\right]_{lm} \theta(y - x_i)$$

$$\xrightarrow{l=2} \delta_{l2} \sum_{m_1,m_2} <1m_1 1m_2|2m> \int d\Omega_i \int_0^y x_i^2 dx_i \left(\frac{x_i}{R_N}\right)^3 Y_{1m_1}(\Omega_i)\alpha_{i,m_2}$$

$$\times \vec{x}_i \cdot \left[\psi_f^*(0)\overleftarrow{\nabla}_i\psi_i(0) + \psi_f^*(0)\overrightarrow{\nabla}_i\psi_i(0)\right]$$

$$= \delta_{l2}\sqrt{\frac{4\pi}{3}}\frac{y^7}{7R_N^3}\left[\psi_f^*(0)\left[\overleftarrow{\nabla}_i \otimes \vec{\alpha}_i\right]_2 \psi_i(0) + \psi_f^*(0)\left[\overrightarrow{\nabla}_i \otimes \vec{\alpha}_i\right]_2 \psi_i(0)\right], \qquad (A.37)$$

and

$$\int d^3x_i \left(\frac{R_N}{x_i}\right)^{l} \left[Y_{l-1}(\Omega_i) \otimes \vec{j}_e(\vec{x}_i)\right]_{lm} \theta(y - x_i)$$

$$\xrightarrow{l=2} \delta_{l2} \sum_{m_1,m_2} <1m_1 1m_2|2m> \int d\Omega_i \int_0^y x_i^2 dx_i \left(\frac{R_N}{x_i}\right)^2 Y_{1m_1}(\Omega_i)\alpha_{i,m_2}$$

$$\times \vec{x}_i \cdot \left[\psi_f^*(0)\overleftarrow{\nabla}_i\psi_i(0) + \psi_f^*(0)\overrightarrow{\nabla}_i\psi_i(0)\right]$$

$$= \delta_{l2}\sqrt{\frac{4\pi}{3}}\frac{R_N^2 y^2}{2}\left[\psi_f^*(0)\left[\overleftarrow{\nabla}_i \otimes \vec{\alpha}_i\right]_2 \psi_i(0) + \psi_f^*(0)\left[\overrightarrow{\nabla}_i \otimes \vec{\alpha}_i\right]_2 \psi_i(0)\right]. \qquad (A.38)$$

Noting that the final results in these calculations can be reproduced by matrix elements

of delta functions between atomic states, we can rewrite the penetration terms as follows:

$$-\frac{4\pi\alpha}{R_A}\sum_{i=1}^{Z}\left[C_0^{A+}(i)\odot C_0^{N-}(i)-C_0^{A-}(i)\odot C_0^{N+}(i)\right]$$

$$\rightarrow\frac{4\pi\alpha}{R_A}\frac{\sqrt{4\pi}}{6}\left(\frac{R_N}{R_A}\right)^2\left[R_A^3\sum_{i=1}^{Z}\delta^3(\vec{x}_i)\right]\odot\int d^3y\left(\frac{y}{R_N}\right)^2\rho_N(\vec{y})Y_0(\Omega_y),$$

$$-\frac{4\pi\alpha}{3R_A}\sum_{i=1}^{Z}\left[C_1^{A+}(i)\odot C_1^{N-}(i)-C_1^{A-}(i)\odot C_1^{N+}(i)\right]$$

$$\rightarrow\frac{4\pi\alpha}{R_A}\frac{1}{10}\sqrt{\frac{4\pi}{3}}\left(\frac{R_N}{R_A}\right)^3\left[R_A^4\sum_{i=1}^{Z}\left(\overleftarrow{\vec{\nabla}}_{i,m}\delta^3(\vec{x}_i)+\delta^3(\vec{x}_i)\overrightarrow{\vec{\nabla}}_{i,m}\right)\right]$$

$$\odot\int d^3y\left(\frac{y}{R_N}\right)^3\rho_N(\vec{y})Y_1(\Omega_y),$$

$$-\frac{4\pi\alpha}{3R_A}\sum_{i=1}^{Z}\left[T_{\mathrm{mag},1}^{A+}(i)\odot T_{\mathrm{mag},1}^{N-}(i)-T_{\mathrm{mag},1}^{A-}(i)\odot T_{\mathrm{mag},1}^{N+}(i)\right]$$

$$\rightarrow\frac{4\pi\alpha}{R_A}\frac{1}{10}\sqrt{\frac{4\pi}{3}}\left(\frac{R_N}{R_A}\right)^3\left[R_A^4\sum_{i=1}^{Z}\left(\left[\overleftarrow{\vec{\nabla}}_{i,m}\otimes\vec{\alpha}_i\right]_1\delta^3(\vec{x}_i)+\delta^3(\vec{x}_i)\left[\overrightarrow{\vec{\nabla}}_{i,m}\otimes\vec{\alpha}_i\right]_1\right)\right]$$

$$\odot\int d^3y\left(\frac{y}{R_N}\right)^3\left[Y_1(\Omega_y)\otimes\vec{j}_N(\vec{y})\right]_1,$$

$$
-\frac{4\pi\alpha}{3R_A}\sum_{i=1}^{Z}\left[T_{\text{el},1}^{A+}(i)\odot T_{\text{el},1}^{'N-}(i)-T_{\text{el},1}^{A-}(i)\odot T_{\text{el},1}^{'N+}(i)\right]
$$

$$
\rightarrow\frac{4\pi\alpha}{R_A}\frac{2}{9}\sqrt{4\pi}\left(\frac{R_N}{R_A}\right)^2\left[R_A^3\sum_{i=1}^{Z}\delta^3(\vec{x_i})\vec{\alpha}_i\right]
$$

$$
\odot\left[\int d^3y\left(\frac{y}{R_N}\right)^2\left[Y_0(\Omega_y)\otimes\vec{j}_N(\vec{y})\right]_1-\frac{1}{2\sqrt{2}}\int d^3y\left(\frac{y}{R_N}\right)^2\left[Y_2(\Omega_y)\otimes\vec{j}_N(\vec{y})\right]_1\right],
$$

$$
-\frac{4\pi\alpha}{3R_A}\sum_{i=1}^{Z}\left[T_{\text{el},2}^{A+}(i)\odot T_{\text{el},2}^{'N-}(i)-T_{\text{el},2}^{A-}(i)\odot T_{\text{el},2}^{'N+}(i)\right]
$$

$$
\rightarrow\frac{4\pi\alpha}{R_A}\frac{1}{25}\sqrt{\frac{4\pi}{3}}\left(\frac{R_N}{R_A}\right)^3\left[R_A^4\sum_{i=1}^{Z}\left(\left[\overleftarrow{\nabla}_{i,m}\otimes\vec{\alpha}_i\right]_2\delta^3(\vec{x_i})+\delta^3(\vec{x_i})\left[\overrightarrow{\nabla}_{i,m}\otimes\vec{\alpha}_i\right]_2\right)\right]
$$

$$
\odot\left[\int d^3y\left(\frac{y}{R_N}\right)^3\left[Y_1(\Omega_y)\otimes\vec{j}_N(\vec{y})\right]_2-\sqrt{\frac{3}{2}}\int d^3y\left(\frac{y}{R_N}\right)^3\left[Y_3(\Omega_y)\otimes\vec{j}_N(\vec{y})\right]_2\right],
$$

$$
-\frac{4\pi\alpha}{3R_A}\sum_{i=1}^{Z}\left[T_{\text{el},1}^{'A+}(i)\odot T_{\text{el},1}^{N-}(i)-T_{\text{el},1}^{'A-}(i)\odot T_{\text{el},1}^{N+}(i)\right]
$$

$$
\rightarrow-\frac{4\pi\alpha}{R_A}\frac{\sqrt{4\pi}}{3}\left(\frac{R_N}{R_A}\right)^2\left[R_A^3\sum_{i=1}^{Z}\delta^3(\vec{x_i})\vec{\alpha}_i\right]
$$

$$
\odot\left[\int d^3y\left(\frac{y}{R_N}\right)^2\left[Y_0(\Omega_y)\otimes\vec{j}_N(\vec{y})\right]_1-\frac{\sqrt{2}}{10}\int d^3y\left(\frac{y}{R_N}\right)^2\left[Y_2(\Omega_y)\otimes\vec{j}_N(\vec{y})\right]_1\right],
$$

$$
-\frac{4\pi\alpha}{3R_A}\sum_{i=1}^{Z}\left[T_{\text{el},2}^{'A+}(i)\odot T_{\text{el},2}^{N-}(i)-T_{\text{el},2}^{'A-}(i)\odot T_{\text{el},2}^{N+}(i)\right]
$$

$$
\rightarrow-\frac{4\pi\alpha}{R_A}\frac{1}{10}\sqrt{\frac{4\pi}{3}}\left(\frac{R_N}{R_A}\right)^3\left[R_A^4\sum_{i=1}^{Z}\left(\left[\overleftarrow{\nabla}_{i,m}\otimes\vec{\alpha}_i\right]_2\delta^3(\vec{x_i})+\delta^3(\vec{x_i})\left[\overrightarrow{\nabla}_{i,m}\otimes\vec{\alpha}_i\right]_2\right)\right]
$$

$$
\odot\left[\int d^3y\left(\frac{y}{R_N}\right)^3\left[Y_1(\Omega_y)\otimes\vec{j}_N(\vec{y})\right]_2-\frac{\sqrt{6}}{7}\int d^3y\left(\frac{y}{R_N}\right)^3\left[Y_3(\Omega_y)\otimes\vec{j}_N(\vec{y})\right]_2\right].
$$

$$\tag{A.39}$$

These effective operators apply only to cases where the bare V_{eN}^{pen} is evaluated between atomic states. For $[A, V_{eN}^{\text{pen}}]$, a different set of effective operators need to be derived.

A.3   Perturbation theory

Here, we give the standard results for perturbation theory in non-relativistic quantum mechanics. First, write the Hamiltonian, H, as

$$H \equiv H_0 + V'. \tag{A.40}$$

We have two eigenvalue problems, one for the total Hamiltonian, H, and one for the unperturbed Hamiltonian, H_0.

$$\begin{aligned} H|a> &= E_a|a> \\ H_0|\alpha> &= E_\alpha|\alpha> \end{aligned} \tag{A.41}$$

Typically, H_0 is a Hamiltonian that has been solved, and H is to be solved in successive approximations using the eigenvalues and eigenstates of H_0.

First, expand the energy eigenvalue, E_a, and the eigenfunction, $|a>$, as

$$\begin{aligned} E_a &= E_a^{(0)} + E_a^{(1)} + E_a^{(2)} + \ldots \\ |a> &= |a^{(0)}> + |a^{(1)}> + |a^{(2)}> + \ldots, \end{aligned} \tag{A.42}$$

where the superscripts indicate the number of insertions of the perturbation, V', in the term. Substituting these into eq. (A.41) and then matching the terms that are of the same order in the perturbation, one can show the leading order results

$$\begin{aligned} E_a^{(0)} &= E_\alpha \\ |a^{(0)}> &= |\alpha>, \end{aligned} \tag{A.43}$$

and the recursion relations

$$\begin{aligned} E_a^{(n)} &= <a|V'|a^{(n-1)}> \\ |a^{(n)}> &= \frac{Q_a}{E_a - H_0} \\ &\quad \times \left[\left(E_a^{(1)} - V' \right)|a^{(n-1)}> + E_a^{(2)}|a^{(n-2)}> + E_a^{(3)}|a^{(n-3)}> + \cdots + E_a^{(n)}|a^{(0)}> \right], \end{aligned} \tag{A.44}$$

where Q_a is the projection operator out of $|a>$.

Carrying out the recursion, the results up to 3rd order in the perturbation are

$$E_a^{(1)} = <a|V'|a>$$

$$|a^{(1)}> = \frac{Q_a}{E_a - H_0} V'|a>$$

$$E_a^{(2)} = <a|V'\frac{Q_a}{E_a - H_0} V'|a>$$

$$|a^{(2)}> = \frac{Q_a}{E_a - H_0} V'\frac{Q_a}{E_a - H_0} V'|a> - \frac{Q_a}{(E_a - H_0)^2} V'|a><a|V'|a>$$

$$E_a^{(3)} = <a|V'\frac{Q_a}{E_a - H_0} V'\frac{Q_a}{E_a - H_0} V'|a> - <a|V'\frac{Q_a}{(E_a - H_0)^2} V'|a><a|V'|a>$$

$$\begin{aligned}
|a^{(3)}> =& \frac{Q_a}{E_a - H_0} V'\frac{Q_a}{E_a - H_0} V'\frac{Q_a}{E_a - H_0} V'|a> \\
& - \frac{Q_a}{E_a - H_0} V'\frac{Q_a}{(E_a - H_0)^2} V'|a><a|V'|a> \\
& - \frac{Q_a}{(E_a - H_0)^2} V'\frac{Q_a}{E_a - H_0} V'|a><a|V'|a> \\
& + \frac{Q_a}{(E_a - H_0)^3} V'|a> (<a|V'|a>)^2 \\
& - \frac{Q_a}{(E_a - H_0)^2} V'|a><a|V'\frac{Q_a}{E_a - H_0} V'|a> .
\end{aligned} \tag{A.45}$$

A.4   Projection Theorem

For an atomic state with total angular momentum F, the energy shift due to external field $\vec{E}^{\text{ext}}$ is

$$\Delta E = \vec{E}^{\text{ext}} \cdot <FM_f|\vec{d}|FM_i> \tag{A.46}$$

$$= \sum_m (-1)^m E_{1,-m}^{\text{ext}} <FM_f|\vec{d}_{1m}|FM_i> \tag{A.47}$$

$$= \sum_m (-1)^{m+F-M_f} E_{1,-m}^{\text{ext}} \begin{pmatrix} F & 1 & F \\ -M_f & m & M_i \end{pmatrix} <F||\vec{d}||F>, \tag{A.48}$$

where $\vec{d} = \vec{d}_e + \vec{d}_N$.

Compare this with the expression involving the angular momentum operator $\vec{F}$,

$$< FM_f|F_{1m}|FM_i > \tag{A.49}$$

$$= (-1)^{F-M_f} \begin{pmatrix} F & 1 & F \\ -M_f & m & M_i \end{pmatrix} < F||F||F > \tag{A.50}$$

$$= (-1)^{F-M_f} \begin{pmatrix} F & 1 & F \\ -M_f & m & M_i \end{pmatrix} \begin{pmatrix} F & 1 & F \\ -F & 0 & F \end{pmatrix}^{-1} < FF|F_{10}|FF > \tag{A.51}$$

$$= (-1)^{F-M_f} \begin{pmatrix} F & 1 & F \\ -M_f & m & M_i \end{pmatrix} \left(\frac{F}{\sqrt{F(F+1)(2F+1)}} \right)^{-1} F \tag{A.52}$$

$$= (-1)^{F-M_f} \begin{pmatrix} F & 1 & F \\ -M_f & m & M_i \end{pmatrix} \sqrt{F(F+1)(2F+1)}, \tag{A.53}$$

we get the following alternative expression for the energy shift

$$\Delta E = \sum_m (-1)^m E^{\text{ext}}_{1,-m} \left(\frac{< FM_f|F_{1m}|FM_i >}{\sqrt{F(F+1)(2F+1)}} \right) < F||\vec{d}||F > \tag{A.54}$$

$$= \frac{< FM_f|\vec{E}^{\text{ext}} \cdot \vec{F}|FM_i >}{\sqrt{F(F+1)(2F+1)}} < F||\vec{d}||F > . \tag{A.55}$$

This shows that the reduced matrix elements of $\vec{d}$ are what we need to calculate the atomic EDM response.

Appendix B

Harmonic-Oscillator-Based Effective Theory

B.1 Harmonic Oscillator Eigenstates

Harmonic oscillator (HO) Hamiltonian is

$$H_{HO} = T + V_{HO} = \frac{\hbar\omega}{2}\left(-\overrightarrow{\nabla}^2 + r^2\right), \tag{B.1}$$

where $\omega = \hbar/mb^2$. The coordinates are Jacobi relative coordinates for the 2 nucleons,

$$\vec{r} = \frac{\vec{r}_1 - \vec{r}_2}{\sqrt{2}}, \tag{B.2}$$

and lengths are measured in units of b; $\vec{r}$ and $\overrightarrow{\nabla}$ are dimensionless.

A HO eigenstate is identified with nodal quantum number n and angular momentum l, and its coordinate wavefunction is

$$< r|nlm >= R_{nl}(r)Y_{lm}(\Omega), \tag{B.3}$$

with the definition

$$R_{nl}(r) \equiv \sqrt{\frac{2(n-1)!}{\Gamma(n+l+1/2)}} r^l e^{-r^2/2} L_{n-1}^{l+1/2}(r^2)$$

$$= \sqrt{2(n-1)!\Gamma(n+l+1/2)} \sum_{m=0}^{n-1} \frac{(-1)^m r^{2m+l} e^{-r^2/2}}{m!(n-m-1)!\Gamma(l+m+3/2)}. \tag{B.4}$$

$L_n^\alpha(r)$ is the associated Laguerre polynomial.

Taking the Fourier transform, the HO wavefunction in momentum space is

$$
\begin{aligned}
< p|nlm > &= \int d^3r \ < p|r >< r|nlm > \\
&= \int d^3r \ e^{-i\vec{p}\cdot\vec{r}} \sqrt{\frac{2(n-1)!}{\Gamma(n+l+1/2)}} L_{n-1}^{l+1/2}(r^2) r^l e^{-r^2/2} Y_{lm}(\Omega) \\
&= \sqrt{\frac{2(n-1)!}{\Gamma(n+l+1/2)}} \int d\Omega \int_0^\infty r^2 dr \ 4\pi(-i)^l j_l(pr) Y_{lm}(\Omega_p) Y_{lm}^*(\Omega) \times \\
&\quad \times L_{n-1}^{l+1/2}(r^2) r^l e^{-r^2/2} Y_{lm}(\Omega) \\
&= 4\pi(-i)^l \sqrt{\frac{2(n-1)!}{\Gamma(n+l+1/2)}} Y_{lm}(\Gamma_p) \\
&\quad \times \int dr \ r^{l+2} e^{-r^2/2} L_{n-1}^{l+1/2}(p^2) \sqrt{\frac{\pi}{2pr}} J_{l+1/2}(pr) \\
&= (2\pi)^{3/2}(-i)^l(-1)^{n-1} \sqrt{\frac{2(n-1)!}{\Gamma(n+l+1/2)}} p^l e^{-p^2/2} L_{n-1}^{l+1/2}(p^2) Y_{lm}(\Gamma_p). \quad\text{(B.5)}
\end{aligned}
$$

The last integral was done using 7.421.4 of [20]. Up to a constant, the functional forms of (B.4) and (B.5) are identical.

B.2 Evaluation of $\frac{1}{E-T+i\epsilon}$ in HO Basis

Using the coordinate space representation of the kinetic energy

$$
T = -\frac{\hbar\omega}{2}\overrightarrow{\nabla}^2, \quad\text{(B.6)}
$$

the momentum space representation of $1/(E - T + i\epsilon)$ is

$$
\begin{aligned}
< p'|\frac{1}{E-T+i\epsilon}|p > &= \int d^3r \ < p'|r >< r|\frac{1}{E-T+i\epsilon}|r >< r|p > \\
&= \int d^3r \ e^{-i\vec{p'}\cdot\vec{r}} \frac{1}{E + \frac{\hbar\omega}{2}\overrightarrow{\nabla}^2 + i\epsilon} e^{i\vec{p}\cdot\vec{r}} \\
&= \frac{2}{\hbar\omega} \frac{1}{k^2 - p^2 + i\epsilon} \int d^3r \ e^{i(\vec{p}-\vec{p'})\cdot\vec{r}} \\
&= \frac{2}{\hbar\omega} \frac{1}{k^2 - p^2 + i\epsilon} (2\pi)^3 \delta^3(\vec{p} - \vec{p'}). \quad\text{(B.7)}
\end{aligned}
$$

Using the momentum space HO wavefunction from the last section,

$$
\begin{aligned}
&< n'lm|\frac{1}{E-T+i\epsilon}|nlm> \\
&=\frac{2}{\hbar\omega}\int\frac{d^3p}{(2\pi)^3}< n'lm|p>\frac{1}{k^2-p^2+i\epsilon}< p|nlm> \\
&=\frac{2}{\hbar\omega}\int_0^\infty\frac{dp\,p}{(2\pi)^3}(2\pi)^{3/2}i^l(-1)^{n'-1}\sqrt{\frac{2(n'-1)!}{\Gamma(n'+l+1/2)}}p^l e^{-p^2/2}L_{n'-1}^{l+1/2}(p^2)Y_{lm}^*(\Gamma_p)\times \\
&\quad\times(2\pi)^{3/2}(-i)^l(-1)^{n-1}\sqrt{\frac{2(n-1)!}{\Gamma(n+l+1/2)}}p^l e^{-p^2/2}L_{n-1}^{l+1/2}(p^2)Y_{lm}(\Gamma_p) \\
&=\frac{2}{\hbar\omega}(-1)^{n+n'}\sqrt{\frac{4(n-1)!(n'-1)!}{\Gamma(n+l+1/2)\Gamma(n'+l+1/2)}}\int_0^\infty\frac{dp\,p^{2(l+1)}e^{-p^2}L_{n-1}^{l+1/2}(p^2)L_{n'-1}^{l+1/2}(p^2)}{k^2-p^2+i\epsilon}.
\end{aligned}
\tag{B.8}
$$

Expanding the Laguerre polynomials,

$$
\begin{aligned}
&< n'lm|\frac{1}{E-T+i\epsilon}|nlm> \\
&=\frac{2}{\hbar\omega}(-1)^{n+n'}\sqrt{\frac{4(n-1)!(n'-1)!}{\Gamma(n+l+1/2)\Gamma(n'+l1/2)}}\times \\
&\quad\times\sum_{m=0}^{n-1}\sum_{m'=0}^{n'-1}\frac{(-1)^{m+m'}\Gamma(n+l+1/2)\Gamma(n'+l+1/2)}{m!m'!(n-1-m)!(n'-1-m')!\Gamma(l+m+3/2)\Gamma(l+m'+3/2)}\times \\
&\quad\times\int_0^\infty\frac{dp\,p^{2l+2m+2m'+2}e^{-p^2}}{k^2-p^2+i\epsilon} \\
&=\frac{2}{\hbar\omega}\sum_{m=0}^{n-1}\sum_{m'=0}^{n'-1}\frac{(-1)^{m+m'+n+n'}\sqrt{4(n-1)!(n'-1)!\Gamma(n+l+1/2)\Gamma(n'+l+1/2)}}{m!m'!(n-1-m)!(n'-1-m')!\Gamma(l+m+3/2)\Gamma(l+m'+3/2)}\times \\
&\quad\times\int_0^\infty\frac{dp\,p^{2l+2m+2m'+2}e^{-p^2}}{k^2-p^2+i\epsilon}.
\end{aligned}
\tag{B.9}
$$

Now, evaluate this last integral.

$$
\begin{aligned}
\int_0^\infty\frac{dp\,p^{2N}e^{-p^2}}{k^2-p^2+i\epsilon}&=\int_0^\infty\frac{du}{2\sqrt{u}}\frac{u^N e^{-u}}{k^2-u+i\epsilon} \\
&=-\frac{1}{2}\int_0^\infty du\frac{u^{N-1/2}e^{-u}}{u-k^2-i\epsilon} \\
&=-\frac{1}{2}\left[\mathcal{P}\int_0^\infty du\frac{u^{N-1/2}e^{-u}}{u-k^2}+i\pi k^{2N-1}e^{-k^2}\right].
\end{aligned}
\tag{B.10}
$$

The principal value integral was done using Mathematica, then simplified

$$\mathcal{P}\int_0^\infty du \frac{u^{N-1/2}e^{-u}}{u-k^2} = -(N-1/2)e^{-k^2}(-k^2)^{N-1/2}\Gamma(N-1/2)\gamma(1/2-N,-k^2). \quad \text{(B.11)}$$

Substituting this back into eq. (B.10),

$$\int_0^\infty \frac{dp\, p^{2N}e^{-p^2}}{k^2-p^2+i\epsilon}$$
$$= \frac{e^{-k^2}}{2}\left[(N-1/2)(-k^2)^{N-1/2}\Gamma(N-1/2)\gamma(1/2-N,-k^2) - i\pi k^{2N-1}\right]. \quad \text{(B.12)}$$

Finally, the Green's function in HO basis is

$$< n'lm|\frac{1}{E-T+i\epsilon}|nlm >$$
$$= \frac{1}{\hbar\omega}\sum_{m=0}^{n-1}\sum_{m'=0}^{n'-1} \frac{(-1)^{m+m'+n+n'+1}\sqrt{4(n-1)!(n'-1)!\Gamma(n+l+1/2)\Gamma(n'+l+1/2)}}{m!m'!(n-1-m)!(n'-1-m')!\Gamma(l+m+3/2)\Gamma(l+m'+3/2)}$$
$$\times e^{-k^2}\left[(N-1/2)(-k^2)^{N-1/2}\Gamma(N-1/2)\gamma(1/2-N,-k^2) - i\pi k^{2N-1}\right], \quad \text{(B.13)}$$

where $N \equiv l + m + m' + 1$.

B.2.1   Comparison with $E < 0$

For k^2 real and less than 0, the integral also can be done on Mathematica, this time without using principal value:

$$\int_0^\infty \frac{dp\, p^{2N}e^{-p^2}}{k^2-p^2+i\epsilon} = -\frac{e^{-k^2}}{2}(-k^2)^{N-1/2}\Gamma(N+1/2)\Gamma(1/2-N,-k^2). \quad \text{(B.14)}$$

The difference between this result and eq. (B.12), after using FullSimplify on Mathematica, is

$$\delta I = \frac{\pi e^{-k^2}}{2}\left[-ik^{2N-1} + (-k^2)^{N-1/2}\sec N\pi - \left(\frac{1}{k^2}\right)^{1/2-N}\tan N\pi\right]. \quad \text{(B.15)}$$

For integer N, $\sec N\pi = (-1)^N$ and $\tan N\pi = 0$. So this simplifies to

$$\delta I = \frac{\pi e^{-k^2}}{2}\left[-ik^{2N-1} + (-1)^N(-k^2)^{N-1/2}\right], \quad \text{(B.16)}$$

which is 0 when k is in the upper complex plane. This shows that eq. (B.12) is the correct analytic continuation of eq. (B.14).

B.3 Matrix Elements of Contact-Gradient Operators

This section gives the matrix elements of the contact-gradient operators between HO eigenstates and tilde states. It follows the appendix of [17], with slight modifications to treat the $E > 0$ case.

The effect of the replacement

$$O \to \bar{O} \equiv e^{r^2/2} O e^{r^2/2} \tag{B.17}$$

is to make the contact-gradient operators (as listed on Table 2.1) act on the wavefunction $e^{r^2/2} R_{nl} Y_{lm}$ and $e^{r^2/2} \tilde{R}_{nl} Y_{lm}$ rather than the usual HO wavefunctions and tilde states. What we need are expressions that describe what happens when gradients and then a delta function act on these wavefunctions.

From

$$\overrightarrow{\nabla}^2 e^{r^2/2} R_{nl}(r) Y_{lm}(\Omega) = -4\sqrt{(n-1)(n+l-1/2)} e^{r^2/2} R_{n-1,l}(r) Y_{lm}(\Omega), \tag{B.18}$$

a more general expression

$$\left(\overrightarrow{\nabla}^2\right)^p e^{r^2/2} R_{nl}(r) Y_{lm}(\Omega)$$

$$= (-4)^p \left[\frac{(n-1)!\Gamma(n+l+1/2)}{(n-1-p)!\Gamma(n+l+1/2-p)} \right]^{1/2} e^{r^2/2} R_{n-p,l}(r) Y_{lm}(\Omega) \tag{B.19}$$

can be derived.

Defining $(\overrightarrow{\nabla}^q)_{q0}$ as maximally coupled q gradients,

$$\left(\overrightarrow{\nabla}^q\right)_{q0} e^{r^2/2} R_{nl}(r) Y_{l0}(\Omega) \Big|_{r=0} = \delta_{lq} 2^l \left[\frac{l!}{(2l+1)!!} \right]^{1/2} \frac{1}{\pi} \left[\frac{2\Gamma(n+l+1/2)}{(n-1)!} \right]^{1/2}. \tag{B.20}$$

Using eq. (B.19) then eq. (B.20),

$$\left(\overrightarrow{\nabla}^2\right)^p \left(\overrightarrow{\nabla}^q\right)_{q0} e^{r^2/2} R_{nl}(r) Y_{l0}(\Omega) \Big|_{r=0} = \delta_{lq} (-1)^p \frac{(n-1)!}{(n-1-p)!}$$

$$\times 2^l \left[\frac{l!}{(2l+1)!!} \right]^{1/2} \frac{1}{\pi} \left[\frac{2\Gamma(n+l+1/2)}{(n-1)!} \right]^{1/2}. \tag{B.21}$$

Eq. (B.21) is sufficient for determining the matrix elements for HO basis states. In principle, matrix elements of contact-gradient operators between tilde states can also be determined, after expanding the tilde state in HO basis. However, an easier method is to start

by recalling that

$$\tilde{R}_{nl}^{\alpha} Y_{lm}(\Omega) = \frac{1}{E - T + i\epsilon} |\alpha_{nlm}>$$

$$= -ikY_{lm}(\Omega) \left[\int_0^r r'^2 dr' j_l(kr') h_l^{(1)}(kr) R_{nl}^{\alpha}(r') \right.$$

$$\left. + \int_r^{\infty} r'^2 dr' j_l(kr) h_l^{(1)}(kr') R_{nl}^{\alpha}(r') \right], \tag{B.22}$$

and letting the gradients act on this expression.

Repeated application of Laplacian on the tilde state results in

$$\left(\vec{\nabla}^2\right)^p e^{r^2/2} \tilde{R}_{nl}^{\alpha}(r) Y_{lm}(\Omega) = e^{r^2/2} Y_{lm}(\Omega) \times$$

$$\left\{ -ik \left[f_h^p(k^2, r^2) h_l^{(1)}(kr) + f_{h'}^p(k^2, r^2) 2r \frac{d}{dr} h_l^{(1)}(kr) \right] \int_0^r r'^2 dr' j_l(kr') R_{nl}^{\alpha}(r') \right.$$

$$+ -ik \left[f_j^p(k^2, r^2) j_l(kr) + f_{j'}^p(k^2, r^2) 2r \frac{d}{dr} j_l(kr) \right] \int_r^{\infty} r'^2 dr' h_l^{(1)}(kr') R_{nl}^{\alpha}(r')$$

$$+ f_{\alpha}^p(k^2, r^2) e^{r^2/2} R_{nl}^{\alpha}(r) Y_{lm}(\Omega) + f_{\alpha'}^p(k^2, r^2) 2r \frac{d}{dr} \left(e^{r^2/2} R_{nl}^{\alpha}(r) Y_{lm}(\Omega) \right)$$

$$+ f_{\alpha''}^p(k^2, r^2) \vec{\nabla}^2 \left(e^{r^2/2} R_{nl}^{\alpha}(r) Y_{lm}(\Omega) \right) + f_{\alpha^{3'}}^p(k^2, r^2) 2r \frac{d}{dr} \vec{\nabla}^2 \left(e^{r^2/2} R_{nl}^{\alpha}(r) Y_{lm}(\Omega) \right)$$

$$+ f_{\alpha^{4'}}^p(k^2, r^2) \vec{\nabla}^4 \left(e^{r^2/2} R_{nl}^{\alpha}(r) Y_{lm}(\Omega) \right) + f_{\alpha^{5'}}^p(k^2, r^2) 2r \frac{d}{dr} \vec{\nabla}^4 \left(e^{r^2/2} R_{nl}^{\alpha}(r) Y_{lm}(\Omega) \right)$$

$$+ \dots \right\}. \tag{B.23}$$

Since a delta function acts on the expression at the end, only the behavior of this expression

at $r = 0$ is relevant. This is given by

$$
\left(\vec{\nabla}^2\right)^p e^{r^2/2}\tilde{R}_{nl}^\alpha(r)Y_{lm}(\Omega) \to \frac{e^{r^2/2}Y_{lm}(\Omega)}{(2l+1)!!} \times
$$

$$
\left(-ik\left[f_j^p(k^2,0) + 2lf_{j'}^p(k^2,0)\right]\int_0^\infty r'^2 dr' h_l^{(1)}(kr')R_{nl}^\alpha(r')\right.
$$

$$
+ 2^{l+1}\left[\frac{2\Gamma(n+l+1/2)}{\pi(n-1)!}\right]^{1/2} \times
$$

$$
\left\{\left[f_\alpha^p(k^2,0) + 2lf_{\alpha'}^p(k^2,0)\right]\left(-k^2 + 3n + l - 3/2 + \tilde{g}_1(k^2,n,l)\sqrt{n(n+l+1/2)}\right)\right.
$$

$$
- 4(n-1)\left[f_{\alpha^{2\prime}}^p(k^2,0) + 2lf_{\alpha^{3\prime}}^p(k^2,0)\right]
$$

$$
\times \left(-k^2 + 3n + l - 5/2 + \tilde{g}_1(k^2,n,l)\sqrt{n(n+l+1/2)}\right)
$$

$$
+ 16(n-1)(n-2)\left[f_{\alpha^{4\prime}}^p(k^2,0) + 2lf_{\alpha^{5\prime}}^p(k^2,0)\right]
$$

$$
\left.\left.\times \left(-k^2 + 3n + l - 7/2 + \tilde{g}_1(k^2,n,l)\sqrt{n(n+l+1/2)}\right)\right\}\right). \tag{B.24}
$$

Nonzero coefficients in eq. (B.24) are

$$
f_j^p(k^2,0) = \begin{cases} 1 & p=0 \\ 3 - k^2 & p=1 \\ 15 + 4l(l+1) - 10k^2 + k^4 & p=2 \\ 105 + 52l(l+1) - 105k^2 - 14l(l+1)k^2 + 21k^4 - k^6 & p=3 \end{cases}
$$

$$
f_{h'}^p(k^2,0) = f_{j'}^p(k^2,0) = \begin{cases} 1 & p=1 \\ 6 - k^2 & p=2 \\ 45 + 4l(l+1) - 30k^2 + 3k^4 & p=3 \end{cases}
$$

$$
f_\alpha^{p=1}(k^2,0) = f_{\alpha'}^{p=2}(k^2,0) = f_{\alpha^{2\prime}}^{p=2}(k^2,0) = f_{\alpha^{3\prime}}^{p=3}(k^2,0) = f_{\alpha^{4\prime}}^{p=3}(k^2,0) = 1
$$

$$
f_\alpha^{p=2}(k^2,0) = 7 - k^2
$$
$$
f_\alpha^{p=3}(k^2,0) = 57 + 4l(l+1) - 18k^2 + k^4
$$
$$
f_{\alpha^{2\prime}}^{p=3}(k^2,0) = 11 - k^2
$$

These are identical to the result shown in [17], with the replacement $\kappa^2 \to -k^2$, which is as expected, as they are simply different ways of showing the dependence on E.

The equivalent of eq. (B.21) is

$$
\left(\vec{\nabla}^2\right)^p \left(\vec{\nabla}^q\right)_{q0} e^{r^2/2} \tilde{R}^\alpha_{nl}(r) Y_{lm}(\Omega)\Big|_{r=0} = \delta_{lq} \sqrt{\frac{l!}{4\pi(2l+1)!!}} \times
$$

$$
\left\{ -ik \left[f^p_j(k^2,0) + 2l f^p_{j'}(k^2,0) \right] \int_0^\infty r'^2 dr' h^{(1)}_l(kr') R^\alpha_{nl}(r') \right.
$$

$$
+ 2^{l+1} \left[\frac{2\Gamma(n+l+1/2)}{\pi(n-1)!} \right]^{1/2} \times
$$

$$
\left\{ \left[f^p_\alpha(k^2,0) + 2l f^p_{\alpha'}(k^2,0) \right] \left(-k^2 + 3n + l - 3/2 + \tilde{g}_1(k^2,n,l)\sqrt{n(n+l+1/2)} \right) \right.
$$

$$
- 4(n-1) \left[f^p_{\alpha^{2'}}(k^2,0) + 2l f^p_{\alpha^{3'}}(k^2,0) \right]
$$

$$
\times \left(-k^2 + 3n + l - 5/2 + \tilde{g}_1(k^2,n,l)\sqrt{n(n+l+1/2)} \right)
$$

$$
+ 16(n-1)(n-2) \left[f^p_{\alpha^{4'}}(k^2,0) + 2l f^p_{\alpha^{5'}}(k^2,0) \right]
$$

$$
\left. \times \left(-k^2 + 3n + l - 7/2 + \tilde{g}_1(k^2,n,l)\sqrt{n(n+l+1/2)} \right) \right\}.
$$

$$
\tag{B.25}
$$