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MICROSCOPIC THEORY OF THE KNIGHT SHIFT

by

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This dissertation is the beginning of the development of a microscopic theory of the Knight shift. The Knight shift experiment has been used in superconductivity research throughout history, however, a complete understanding of the Knight shift in conventional as well as unconventional superconductors does not yet exist. Motivated by the results of a literature review, which discusses Knight shift anomalies in multiple superconducting materials, this research studies a new model of the Knight shift, which involves the processes involved in nuclear magnetic resonance measurements in metals.

The result of this study is a microscopic model of nuclear magnetic resonance in metals. The spins of the spin-1/2 local nucleus and its surrounding orbital electrons interact with the arbitrary constant $B_0$ and perpendicular time-oscillatory magnetic inductions $B_1(t)$ and with each other via an anisotropic hyperfine interaction. An Anderson-like Hamiltonian describes the excitations of the relevant occupied local orbital electrons into the conduction bands, each described by an anisotropic effective mass with corresponding Landau orbits and an anisotropic spin $g$ tensor. Local orbital electron correlation effects are included using the mean-field decoupling procedure of Lacroix. The metallic contributions to the Knight shift resonance frequency and linewidth shifts are evaluated to leading orders in the hyperfine and Anderson excitation interactions. While respectively proportional to $(B_1/B_0)^2$ and a constant for weak $B_0 >> B_1$, both shifts are shown to depend strongly upon $B_0$ when a Landau level is near the Fermi energy.
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CHAPTER 1: INTRODUCTION

This study is the beginning of a new microscopic model of the Knight shift, which is used for detecting parallel spin states in superconducting materials. The Knight shift is a nuclear magnetic resonance (NMR) measurement which measures a change in NMR frequency for a nucleus in a metal from that in a vacuum or an insulator.

The Knight shift has been used for detecting possible parallel spin states, such as Sr$_2$RuO$_4$, however an improved theory of the Knight shift is needed to explain anomalies that have been found in a variety of superconductors below the superconducting transition temperature. The standard model includes the Zeeman and hyperfine interactions of the magnetic field and the electron-electron pairing interaction necessary for superconductivity. The new model now includes a term based on the Anderson model of local moments in metals. The local electron states in the original Anderson model now correspond to local atomic electron orbitals. When these orbital energies are singly occupied and close to the Fermi energy, the spins of the electrons can interact with the nuclear spins and the spins of the electrons in the conducting bands. Additionally, the paths of the electrons in the conduction bands depend on the direction and magnitude of the magnetic induction. With these new terms included, this model has the potential to describe the anomalous behavior seen in Knight shift measurements performed in unconventional superconductors.

The original motivation behind this project was to clarify the nature of the superconductivity seen in Sr$_2$RuO$_4$, a compound that was discovered to be superconducting in 1994. Sr$_2$RuO$_4$ has a layered perovskite structure, similar to that of the high temperature superconducting cuprates. This characteristic made it an attractive compound for researchers early on. Early Knight shift measurements on Sr$_2$RuO$_4$ gave results suggestive of a parallel-spin state, how-
ever recent experiments give contradicting results suggestive of a conventional, \(s\)-wave, BCS superconductor. Upper critical field calculations of the chiral \(p\)-wave states with parallel- and antiparallel-spin pairing and of conventional \(s\)-wave pairing were performed and fitted to recent experimental data, the results of which strongly rule out the parallel-spin state possibility in \(\text{Sr}_2\text{RuO}_4\).

The Knight shift anomaly seen in \(\text{Sr}_2\text{RuO}_4\) initiated a literature review to find other superconductors with similar anomalies. Among the materials that were found to also have contradicting Knight shift and upper critical field measurements were the well known superconductors \(\text{UPt}_3\) and \(\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}\). The results of this search led to the realization that a more comprehensive model the Knight was necessary to explain these anomalies.

Analysis of this new model in metals has resulted in an analytic expression for the shift in the NMR frequency as well as shift in the linewidth, which has not been studied before. Future work on the model will involve its analysis in superconducting materials. Eventually, we hope to use this model to finally clarify the nature of the superconductivity of \(\text{Sr}_2\text{RuO}_4\). Knowledge of the superconductivity of \(\text{Sr}_2\text{RuO}_4\) is important for the research and later development of topological superconductors, an area of great research interest. This new state of matter is believed to be possible through the proximity of a spin triplet superconductor. Although the community advertises \(\text{Sr}_2\text{RuO}_4\) as a candidate, our calculations that \(\text{Sr}_2\text{RuO}_4\) is actually a spin singlet superconductor would rule it out as such. However, the use of more likely spin triplet superconductors, such as \(\text{URhGe}\) or \(\text{UCoGe}\), would be better suited for the proximity injection of parallel-spin pairs into a topological insulator.
CHAPTER 2: KNIGHT SHIFT ANOMALIES

The Knight shift experiment has been used in superconductivity research throughout history; however, a complete understanding of the Knight shift in conventional as well as unconventional superconductors does not exist. The current theory of the Knight shift dates back to the early days of the Bardeen Cooper Schrieffer theory of conventional superconductivity and when applied to experimental data, it can contradict results obtained through thermodynamic measurements such as upper critical field measurements or specific heat measurements. The following are specific examples of such cases.

2.1 Yttrium Barium Copper Oxide

The material YBa$_2$Cu$_3$O$_{7-\delta}$ is known to be a singlet pair-spin superconductor, as is seen in many experiments including the $^{63}$Cu Knight shifts performed by Barrett et al. [1]; however, one inconsistency in these Knight shift measurements has been overlooked. Figure 2.1 gives the Knight shift results where NMR experiments were performed on the copper atoms in the chains (the Cu(1) atoms) as well as on the copper atoms in the planes (the Cu(2)), where superconductivity is believed to occur. Of the four measurements, all except one show a temperature dependence that follows the Yosida function, with the exception being in the Cu(2) atom, where the applied magnetic field is parallel to the c-axis. In this measurement, the Knight shift has no temperature dependence. This problematic result was also seen by Slichter, who called it “fortuitous” [2].
2.2 Sodium Cobaltate

Another material with an observable discrepancy is Na\textsubscript{x}CoO\textsubscript{2}·yH\textsubscript{2}O. Figure 2.2 shows the sodium and cobalt NMR data which was performed with the magnetic field parallel to the planes of the material [3]. The sodium data does not show a shift, in fact the data is at zero percent, while the cobalt data only shows a shift at low fields. On the contrary, the upper critical field data in Figure 2.3 show that the upper critical field is Pauli limited for the field parallel to the plane [4]. Using the existing theory of the Knight shift, these two experiments give results that are in direct contradiction: the Knight shift suggesting a triplet spin state and the upper critical field suggesting a singlet spin state.

2.3 Cerium Cobalt Indium 5

NMR and Upper Critical Field data for the superconductor CeCoIn\textsubscript{5} also give inconsistent results. Figure 2.4 shows Indium and Cobalt NMR data by Kohori [5], which give different results. The Indium data show a temperature dependent Knight shift both perpendicu-
Figure 2.2: Na and Co NMR data for Na$_x$CoO$_2$·yH$_2$O from Michioka et al. [3].

Figure 2.3: Upper critical field data for Na$_x$CoO$_2$·yH$_2$O from Chou et al. [4].

lar and parallel to the layers, while the Cobalt data only show a temperature dependence perpendicular to the planes. This is inconsistent with the Upper Critical Field data [6] in Figure 2.5, which shows that the material is Pauli limited perpendicular to the field but violates the Pauli limit in the planes.

2.4 Strontium Ruthenate

The well known superconductor Sr$_2$RuO$_4$ does not appear to have a temperature dependent Knight shift, suggestive of spin triplet superconductivity; however, its Upper Critical Field
Sr$_2$RuO$_4$ has a layered perovskite structure as shown in Figure 2.10. This complex structure turns out to be very important in the discovery of the superconductivity in Sr$_2$RuO$_4$. Despite this complex lattice structure, the material is mainly two dimensional and has three fairly simple Fermi surfaces. These Fermi surfaces are shown in Figure 2.11. They consist of three nearly cylindrical shells, which are the $\alpha$, $\beta$, and $\gamma$ bands. There is only a little bit of is Pauli limited. See Figures 2.6-2.9.
2.4.1 Discovery of Superconductivity in Strontium Ruthenate

The superconductivity of Sr$_2$RuO$_4$ was first discovered by Yoshiteru Maenu in 1994 [10]. Although phenomenologically of type II, the microscopic nature of the superconductivity it exhibits is yet to be completely understood. The peculiarly high $T_c$ superconductivity of the cuprates, discovered in the late 1980’s, spurred the search for more materials of similar structure, specifically the layered perovskite structure mentioned earlier. Since Sr$_2$RuO$_4$ dispersion in each of them, creating the somewhat barrel shaped cylinders.
has this same perovskite structure, one would expect that it would have been found to be superconducting soon after the cuprates were discovered. However, it took eight years until superconductivity was finally discovered in ruthenium oxides. This was largely due to the much lower superconducting transition temperature $T_c$, which is around 1.5 K in the cleanest samples, as compared to that of the cuprates, which are around 100 K.
Following the discovery of its superconductivity, measurements of Fermi liquid parameters gave yet more surprising results. The parameters turned out to be similar to those of $^3$He, a spin triplet superfluid, directing researchers to believe that they had indeed found a rare spin triplet superconductor. The first to suggest spin triplet superconductivity in Sr$_2$RuO$_4$ were Rice, Sigrist, and Baskaran [12]. Their reasoning was that since Sr$_2$RuO$_4$ did not have magnetic impurities, which were the only explanation for the reduction in $T_c$ from 1.5 K to 1.13 K in an s-wave superconductor, it could not be an s-wave superconductor. Further
measurements suggestive of the same result include NMR measurements at high fields and NQR measurements at lower fields, both in the normal and superconducting states.

2.4.2 Knight Shift Measurements

The most important experiments, that many believe to be proof of spin triplet superconductivity in Sr$_2$RuO$_4$, are the Knight shift measurements. In one form of this experiment, the spin susceptibility is measured at different temperatures using NMR. The basic theory is that a change in spin susceptibility when passing through the superconducting transition temperature means that the material has singlet pairing. This shift is known as the Knight shift in a superconductor, and its temperature dependence follows a Yosida function. The measurements performed on Sr$_2$RuO$_4$ by Ishida et al. in 1998 showed no such shift [13]. Figure 2.6 shows the results obtained from these experiments. Therefore, Sr$_2$RuO$_4$ was identified as a $p$-wave parallel-spin triplet superconductor. Ever since this measurement, most literature on this material presents Sr$_2$RuO$_4$ as being a triplet superconductor without question. Thus, many in the condensed matter community have moved on to researching uses of Sr$_2$RuO$_4$ as a spin triplet superconductor. But is that really the end of the story?

Besides the polarized neutron results for Sr$_2$RuO$_4$, the results for two other compounds that mostly do show a temperature-dependent Knight shift below $T_c$ are shown in Figure 2.1 and Figure 2.12. With one exception, these data are very different from those obtained in the experiments with Sr$_2$RuO$_4$. Except for the upper panel of Figure 2.1(a), which also shows no temperature dependence to the spin susceptibility of the planar $^{63}$Cu spins in YBa$_2$Cu$_3$O$_{7-\delta}$ with $\mathbf{H}||\hat{c}$, the other graphs clearly show an increasing susceptibility with increasing temperature $T$ that seems to reach a maximum at the superconducting transition temperature $T_c$. As discussed previously, Sr$_2$RuO$_4$ was expected to behave similarly to the
cuprates in superconductivity; however, this is evidently not the case. From the upper part of Figure 2.1(a), which does not show the expected behavior for YBa$_2$Cu$_3$O$_{7-\delta}$ below $T_c$ with $H||\hat{c}$, the Knight shift measurements alone are not reliable enough to determine the superconductivity of a material.

Figure 2.12: Top: V$_3$Si Knight Shift. Bottom: Sr$_2$RuO$_4$ Knight Shift from polarized neutron with $B\perp c = 1$T [15].
2.4.3 Upper Critical Field Measurements

In 2009, upper critical field ($H_{c2}$) measurements were performed on Sr$_2$RuO$_4$ that gave results contradicting the theory that it is a parallel-spin chiral $p$-wave superconductor [9]. The plot in Figure 2.14 gives the results of this experiment, which was done by Kittaka et al. The graph clearly shows that $H_{c2}$ for Sr$_2$RuO$_4$ is Pauli limited for $H \perp \hat{c}$. Pauli limiting is a phenomenon also known as Pauli pair breaking, in which the superconducting Cooper pairs are broken up. This occurs when a magnetic field is applied to a BCS spin singlet superconductor until the Zeeman energy causes one of the paired electron’s spins to flip, thus breaking the pair. When this form of pair breaking is observed, the upper critical field
tends to be lower because the superconductivity is destroyed at lower fields than for orbital
diamagnetic pair breaking alone. Thus, $H_{c2}$ data tend to curve downward rather than follow a
straighter line with decreasing temperature. This is exactly as shown in Figure 2.15, showing
that Sr$_2$RuO$_4$ is Pauli limited, which strongly contradicts the indications of the Knight shift
experiments. Theoretical calculations of the upper critical field of the polar state (1), the
generalized Anderson-Brinkman-Morel (ABM) or SK state (2), the $s$-wave state (3), and
the ABM state (4) all without Pauli-limiting effects for clean superconductors are shown in
Figure 2.15 for reference. If Sr$_2$RuO$_4$ were a chiral $p$-wave parallel-spin superconductor, its
$H_{c2}$ for $H \perp c$ ($0^\circ$) should fit the polar state curve (1). Figures 2.16 and 2.17 are earlier
specific heat measurements of $H_{c2}$ in Sr$_2$RuO$_4$. The dotted line in the inset of Figure 2.16 is
the expected data without Pauli limiting, which Sr$_2$RuO$_4$ clearly does not follow.

![Figure 2.14: Sr$_2$RuO$_4$ $H_{c2}(\theta, T)$ by Kittaka et al. [9].](image)

Figure 2.14: Sr$_2$RuO$_4$ $H_{c2}(\theta, T)$ by Kittaka et al. [9].
Figure 2.15: Theoretical calculations of the reduced upper critical field $h_{c2}$ versus the reduced temperature $t = T/T_c$. (1) Polar state $h_{c2}(t)$. (2) SK state $h_{c2}(t)$. (3) s-wave $h_{c2}(t)$ without Pauli limiting. (4) ABM state $h_{c2}(t)$. [16].

Figure 2.16: Sr$_2$RuO$_4$ $H_{c2}$ by Deguchi et al. [16].
In comparison with Sr$_2$RuO$_4$, there are two materials that are very likely to be $p$-wave superconductors, namely URhGe and UCoGe. Figure 2.19 shows the results of upper critical field experiments performed on URhGe by Hardy and Huxley. These curves clearly show that this superconductor does not follow the BCS prediction, which is represented by the dashed lines. Instead, it seems to be a completely broken symmetry polar state.

**Figure 2.17:** Sr$_2$RuO$_4$ $H_{c2}$ by Deguchi et al. [16].

**Figure 2.18:** Fit to the Deguchi Sr$_2$RuO$_4$ $H_{c2}(\theta)$ data by Machida et al. [43][12].
Figure 2.19: $H_{c2}(T)$ for $H||\hat{a}, \hat{b}, \hat{c}$ for URhGe [13]. Theoretical slopes at $T_c$ are adjusted to fit the three curves. Solid curves: Polar state $H_{c2}(T)$ (top) and completely broken symmetry state $H_{c2}(T)$ (middle and bottom). Dashed curves: $s$-wave state without Pauli limiting. Dashed curve: $s$-wave state with Pauli limiting. Theoretical curves are from [17].

2.4.4 Point Tunneling Spectroscopy Measurements

One very recent experiment showed critical evidence for conventional $s$-wave pairing superconductivity in Sr$_2$RuO$_4$. Point tunneling spectroscopy measurements were performed by Suderow et al. in 2009 and the results are shown in Figure 2.20 [18]. These results match up almost perfectly with the standard BCS theory of superconductivity, strongly suggesting that Sr$_2$RuO$_4$ exhibits conventional $s$-wave pairing. One can easily see that the line predicted by conventional BCS theory matches the experimental data points extremely well.
2.4.5 Corner Junction Experiments

Another attempt at proving $p$-wave superconductivity in Sr$_2$RuO$_4$ was using corner junction experiments. The setup of this experiment is shown in Figure 2.21 and the results are plotted in the graph of Figure 2.22. A truly $p$-wave superconducting material would have a critical current that behaves as $I_c = I_0 \cos[(\phi/\phi_0 + 1/2)\pi]$, where $\phi_0 = \hbar/2e$ is the flux quantum. This would exhibit a current at the magnetic flux $\phi = BA = 0$. Note that as $T$ decreases from 0.42K to 0.21K, $I_c$ approaches the midpoint between a minimum and a maximum, rendering the experiment completely unreliable. The results shown in Figure 2.22 are difficult to interpret. Hence, this experiment does not prove $p$-wave superconductivity in Sr$_2$RuO$_4$. 
2.4.6 Crystal Growth

In addition to contradicting experimental results, Sr$_2$RuO$_4$ is a messy material, which complicates the analysis of all experimental results. Although theoretically Sr$_2$RuO$_4$ is a rather simple material, with three cylindrical Fermi surfaces, in practice it is difficult to grow pure
Sr$_2$RuO$_4$ crystals. The material is grown by Maeno, who uses a floating-zone method [20]. Unfortunately, because of the difficulty in growing this material, Maeno is currently the only one growing it, which adds to the uncertainties of experimental results. Hopefully, in the near future, other experimentalists will begin growing Sr$_2$RuO$_4$, which will allow for comparisons of the same experiments performed on different samples of Sr$_2$RuO$_4$. One of the concerns with the currently available samples is that it is grown rather quickly and that Ru oxides tend to deoxygenate during the process. Figure 2.23 shows a crystal of Sr$_2$RuO$_4$.

![Figure 2.23: Image on a Sr$_2$RuO$_4$ crystal [20].](image)
CHAPTER 3: THE MODEL

3.1 The Original Knight Shift Model

Following the original discovery and confirmations that the nuclear magnetic resonance (NMR) frequency was different for the nuclear species in a metal from that in vacuum or an insulator[21], Yosida proposed a simple model of this Knight shift in a superconductor[22]. In that model, the nuclear moment is assumed to somehow couple to the conduction electron spins, and the spin susceptibility of those conduction electrons was shown to have a strong temperature \( T \) dependence below the superconducting transition temperature \( T_c \) for superconductors with the paired electrons in a singlet spin state[22]. Shortly thereafter, it was proposed that the Knight shift in singlet superconductors could be explained by surface spin-orbit scattering effects[23]. However, there have been many examples of Knight shift measurements in anisotropic and unconventional superconductors in which the Knight shift was found to be independent of the temperature \( T \) for \( T < T_c \), and this behavior was interpreted in many cases as providing evidence for a parallel-spin state of the paired superconducting electrons[24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34]. In the case of UCoGe, recent \(^{59}\)Co Knight shift results were independent of \( T \) well below \( T_c \) [35], which is consistent with the huge upper critical field \( H_{c2} \) measurements on that ferromagnetic superconductor, providing additional support to the notion that it is a parallel-spin superconductor[36]. Similar results on UPt\(_3\) have provided strong evidence for a triplet spin state in that material with three superconducting phases[37], which appears to resolve contradictory Knight shift and \( H_{c2} \) measurements. However, \(^{63}\)Cu Knight shift measurements on the high-temperature superconductor YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) showed the standard Yosida \( T \) dependence for the Cu atoms in the CuO chains for all three applied magnetic field \( \mathbf{H} \) directions, but for the Cu atoms
in the CuO$_2$ layers, such behavior was only seen for $H$ parallel to the layers[38, 39, 40]. In contrast, for $H$ normal to the layers, the Knight shift corresponding to the planar $^{63}$Cu atoms was independent of $T$ for all $T < T_c$, where $T_c \approx 90$K. Slichter was surprised at this result[39], and described it as “fortuitous”, concluding that the probed planar $^{63}$Cu spins must be coupling to the planar near-neighbor $^{63}$Cu spins in such a way as to cancel the effect. However, such behavior has since been seen in a variety of layered and quasi-one-dimensional superconductors [24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34].

One case of widespread interest is Sr$_2$RuO$_4$ [13, 41, 15, 42, 43, 44, 45, 46, 47, 48, 18]. All Knight shift and polarized neutron diffraction experiments showed no $T$ dependences below $T_c$[13, 41, 15, 42], suggesting a parallel-spin chiral $p$-wave state. However, a parallel-spin state is inconsistent with $H_{c2}$ measurements for $H$ near to parallel to the layers[43, 44, 45]. It was suggested that this conflict might be explained by the triplet pair-spin $d$-vector rotating with the magnetic field[46]. However, quantitative fits to the more recent $H_{c2}(T)$ data at angles varying from perpendicular to parallel to the layers for the non-chiral helical $p$-wave states were obtained, but the predictions for the chiral states did not come close to fitting the parallel field data with or without a rotated $d$-vector, evidently due to strong Pauli limiting effects, consistent with a singlet pair state[47]. However, even these helical state possibilities are suspect, as there appears to be strong spin-orbit coupling in the electronic states of Sr$_2$RuO$_4$[48], strongly hindering such a $d$-vector rotation. Moreover, careful scanning tunneling microscopy studies of Sr$_2$RuO$_4$ are consistent with a nodeless gap function [18]. Since there are three Fermi surfaces in that material, it is remarkable that the density of states nearly fits the BCS predictions for a single isotropic gap[18]. Thus, that experiment is most likely inconsistent with a triplet spin state. We note that an isotropic gap for a $p$-wave superconductor could only arise for an isotropic pairing interaction[49], which is unlikely to occur in a highly layered compound[50].
Hence, it appears that not enough is understood about Knight shift measurements. Since the nature of the superconductivity in Sr$_2$RuO$_4$ and in other compounds is still highly controversial, it is important to develop a microscopic model of the Knight shift in anisotropic and correlated metals, in order to better understand the problems in interpreting such experimental data. Here we present and study a microscopic model that may be helpful in that regard. Our model not only leads to an analytic formula for the Knight shift, but also to an analytic formula for the corresponding change in the resonant linewidth due to the probed nucleus being in the metal.

In a solid-state NMR experiment, a strong, time-independent magnetic induction $B_0$ is produced in some arbitrary direction $B_0 = B_0(\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$ with respect to the crystalline $x, y, z$ axes, and an oscillatory magnetic induction $B_1(t)$ with magnitude $B_1$ and angular frequency $\omega_0$ that is rotated in the plane perpendicular to $B_0$, $B_1(t) = B_1[\cos(\omega_0 t)\hat{\theta} - \sin(\omega_0 t)\hat{\phi}]$, where $\hat{\theta} = (\cos \theta \cos \phi, \cos \theta \sin \phi, -\sin \theta)$, $\hat{\phi} = (-\sin \phi, \cos \phi, 0)$, and these inductions interact with the magnetic moment $\mu_n$ of the relevant nuclear isotope via the Zeeman interaction. In addition, the probed nucleus is surrounded by orbital electrons, which also interact with $B_0$ and $B_1(t)$ with magnetic moment $\mu_e$, and the orbital electron spins are coupled to their nuclei by the hyperfine interaction. We then assume that the nuclear and local electron orbital spin quantization directions are both along $B_0$.

In an insulator, these interactions may be written for $I = 1/2$ in first quantization as $H_0 = H_{n,0} + H_{e,0} + H_{hf}$,

\begin{align}
H_{n,0} &= -\omega_n \sum_i I_i^r - \frac{\Omega_n}{2} \sum_i \left( I_i^+ e^{i\omega_0 t} + I_i^- e^{-i\omega_0 t} \right), \\
H_{e,0} &= -\omega_e \sum_{i,q} S_{i,q}^r - \frac{\Omega_e}{2} \sum_{i,q} \left( S_{i,q}^+ e^{i\omega_0 t} + S_{i,q}^- e^{-i\omega_0 t} \right), \\
H_{hf} &= -D_z \sum_{i,q} S_{i,q}^r I_i^r - \frac{D_x}{2} \sum_{i,q} \left( S_{i,q}^+ I_i^- + S_{i,q}^- I_i^+ \right)
\end{align}
where \( \omega_p = \mu_p \cdot B_0, \Omega_p = |\mu_p| B_1 \) for \( p = n, e \), where \( \mu_n \) and \( \mu_e = \mu_B \) are the particular nuclear and Bohr magnetons, \( I_i \) and \( S_{q,i} \) are the nuclear and \( q^{th} \) orbital electron spin operators \([q \in (n, \ell, m)]\) at the site \( i \) of the \( N \) probed nuclei. Although it has been often suggested that the only hyperfine interaction is the Fermi contact interaction between the \( s \) orbital electron spins and the nuclear spin[42], and that the conduction electrons would have to exchange positions with the localized atomic \( s \)-orbitals, this is not correct[51]. The hyperfine interaction between electrons in \( p, d, \) and \( f \) orbitals can be comparable to that of the Fermi contact interaction of the \( s \)-orbital electrons at the position of the nucleus due to magnetic dipole-dipole and higher-order induced interactions[51, 52]. Surely in the case of the \( ^{195}\text{Pt} \) Knight shift in \( \text{UPt}_3[? \ldots] \), the \( s \) orbitals would have little exchange interactions with the electrons in the conduction bands. Hence, the hyperfine interaction is a tensor, but for simplicity we study only a diagonal hyperfine tensor with tetragonal symmetry, \( D_x = D_y \) (or \( D_\theta = D_\phi \)). Note that \( I_i^r \) and \( S_{i,q}^r \) are the operators for the nuclear moment and local electron spin in the orbital \( q \) along the quantization axis \( B_0 = B_0 \hat{r} \) on the site \( i \). The \( I_i^\pm \) are defined from \( I_i^\theta = (I_i^+ + I_i^-)/2 \) and \( I_i^\phi = (I_i^+ - I_i^-)/(2i) \), and the \( S_{i,q}^\pm \) are defined analogously.

Without the hyperfine interaction, the solutions to the Schrödinger equations for the nuclear or electron spins are given in textbooks, and the probability spectrum of flipping a spin of particle \( p = n, e \) at the applied frequency \( \omega_0 \) is

\[
P_p(\omega_0) = \frac{2\Omega_p^2}{2\Omega_p^2 + (\omega_0 - \omega_p)^2}
\]

which is unity at the resonance frequency \( \omega_0 = \omega_p \). These results are unaltered to first order in the two hyperfine interactions \( D_z \) and \( D_x \) in the above model.

The Knight shift is the relative difference \( K = (\omega_{n,m} - \omega_n)/\omega_n \) between the nuclear resonance frequency \( \omega_{n,m} \) of the same nuclear isotope \( n \) in a metal from that \( (\omega_n) \) in an insulator[21, 22],
which is usually measured in a few percent of fractions of one percent for a given nuclear isotope \( n \). Although not generally mentioned, one would expect the same physics to give rise to a change in the linewidth \( 2\Omega_n \), which we propose to quantify analogously, \( L = (\Omega_{n,m} - \Omega_n)/\Omega_n \).

In the first treatments, it was assumed that the conduction electrons interacted with the nuclear spin via the Fermi contact hyperfine interaction\[22\], and that one could set \( \omega_0 \to 0 \), and replace the calculation of the actual shift in the nuclear resonance frequency by the conduction electron spin susceptibility, which could be approximated by its \( B_0, B_1 \to 0 \) limits. In simple metals, such as the single element Hg, this was often found to be the case\[21, 22\]. However, in correlated and anisotropic metals, there are many examples in which this approximation fails. The measured Knight shift often depends strongly upon the direction of \( B_0 \), and can depend strongly upon the temperature \( T \), even in nonmagnetic metals. Moreover, there are many examples of Knight shift measurements in superconductors for which there is little, if any, detected temperature dependence for a variety of nuclear isotopes and \( B_0 \) directions, supposedly indicative of a parallel-spin pairing state, but that interpretation is contradicted by thermodynamic measurements on the same material. Hence, it is imperative to develop a microscopic theory of the Knight shift in metals, and then to use that procedure to extend the calculation to a variety of superconducting materials. In this Fast Track Communication, we propose a model that contains all of the microscopic processes that are important in determining the Knight and linewidth shifts in anisotropic and correlated metals.

### 3.2 The New Model

The starting Hamiltonian for this problems is

\[
\mathcal{H} = \mathcal{H}_Z + \mathcal{H}_hf + \mathcal{H}_{Anderson} + \mathcal{H}_{pairing}
\]  

(3.5)
We define the Zeeman term to be

\[ \mathcal{H}_Z = -\gamma_e \mathbf{B}_e \cdot \vec{g}_e \cdot \mathbf{S}_i - \gamma_n \mathbf{B}_n \cdot \vec{g}_n \cdot \mathbf{I}_i \]  

(3.6)

\( \mathcal{H}_Z \) is the Zeeman interaction between the magnetic induction and the spin operator on the \( i^{th} \) ion, where \( \mathbf{B}_e \) and \( \mathbf{B}_n \) are the electron and nuclear magnetic inductions, \( \mathbf{S}_i \) and \( \mathbf{I}_i \) are electron and nuclear spin operators, \( g_{ei} \) and \( g_{ni} \) are generally dimensionless tensors and \( \gamma_e \) and \( \gamma_n \) are the gyromagnetic ratios of the particular nuclear species of the ion. (see ref 217, 219)

\[ \mathcal{H}_{hf} = \mathbf{I}_i \cdot \vec{D}_{i,q} \cdot \mathbf{S}_{i,q} \]  

(3.7)

The hyperfine interaction occurs between the nuclear spin operator \( \mathbf{I}_i \) on the \( i^{th} \) ion and the electron spin operator \( \mathbf{S}_{i,q} \), where \( \mathbf{S}_{i,q}^- = c_{i,q,\uparrow}^\dagger c_{i,q,\downarrow}, \quad \mathbf{S}_{i,q}^+ = c_{i,q,\downarrow}^\dagger c_{i,q,\uparrow}, \quad \text{and} \quad \mathbf{S}_{i,q}^z = \frac{1}{2}(n_{i,q,\uparrow} - n_{i,q,\downarrow}) \), where \( c_{i,q,\uparrow}^\dagger \) creates an electron with up-spin in the highest singly occupied \( q^{th} \) atomic orbital \( (q = nlm) \) around the \( i^{th} \) ion with an interaction with the nuclear ion spin. This model ensures that only singly occupied atomic orbital levels can contribute to the hyperfine interaction and hence to the Knight shift. Each of the spin operators \( \mathbf{S}_{i,q}^\pm \) and \( \mathbf{S}_{i,q}^z \) annihilates an empty or doubly occupied atomic orbital, so that the only atomic orbitals that can make a contribution to the hyperfine energies are the singly occupied levels. The next term in our model is a variation of the Anderson hamiltonian,

\[ \mathcal{H}_{\text{Anderson}}^{(0)} = \sum_{i,q,\sigma} \epsilon_q n_{i,q,\sigma} + \sum_{i,q} U_q n_{i,q,\uparrow} n_{i,q,\downarrow} \]  

(3.8)

\[ \mathcal{H}_{\text{Anderson}}^{(1)} = \sum_{i,q,j,k,\sigma} (v_{i,q,j,k} c_{j,k,\sigma}^\dagger c_{i,q,\sigma} + H.c.) \]  

(3.9)

\[ \mathcal{H}_{\text{Anderson}}^{(2)} = \sum_{j,k,\sigma} \left( \varepsilon_{j,\sigma}(k + e\mathbf{A}) - \mu |c_{j,k,\sigma}^\dagger c_{j,k,\sigma} + H.c. \right) \]  

(3.10)
In this term, $\epsilon_q$ is the energy of the highest singly occupied atomic orbital on the $i^{th}$ ion and $n_{i,q,\sigma} = c_{i,q,\sigma}^\dagger c_{i,q,\sigma}$ is a generalization of the Anderson model of local impurity moments in interacting with the conduction electrons of charge $-e$ to include the highest energy $q^{th}$ local atomic orbital on the $i^{th}$ ion in the regular crystalline array, the magnetic vector potential $A$, multiple conduction bands indexed by $j$ that cross the Fermi energy $E_F$, and the appropriate spin-orbit coupling in those conduction bands with zero-field dispersions $\epsilon_{j,\sigma}$.

3.3 Initial Calculations

We take the external magnetic field to be

$$H(t) = \hat{z}H_0 + \hat{x}H_1 \cos \omega_0 t - \hat{y}H_1 \sin \omega_0 t$$

(3.11)

The magnetic inductions are then $B_e = \mu_e H(t)$ and $B_n = \mu_n H(t)$. For the following calculations, it is necessary to use the Hamiltonian in spin operator forms. For this specific calculation we use $I = \frac{1}{2}$ and $S = \frac{1}{2}$, and let $\omega_e = \gamma_e g_e \mu_e H_0$, $\omega_n = \gamma_n g_n \mu_n H_0$, $\Omega_e = \frac{1}{2} \gamma_e g_e \mu_e H_1$, and $\Omega_n = \frac{1}{2} \gamma_n g_n \mu_n H_1$. We then obtain the following Hamiltonian.

$$\mathcal{H} = -\omega_e S_z - \omega_n I_z - \Omega_e \left( e^{i\omega_0 t} S_+ + e^{-i\omega_0 t} S_- \right) - \Omega_n \left( e^{i\omega_0 t} I_+ + e^{-i\omega_0 t} I_- \right) + \tilde{D} I_z S_z + \frac{\tilde{D}}{2} I_- S_+ + \frac{\tilde{D}}{2} I_+ S_-$$

(3.12)
CHAPTER 4: FIRST QUANTIZATION

4.1 The Hamiltonian

4.1.1 The Model

The starting Hamiltonian for this problems is

\[ H = -\gamma_i B_e \cdot \vec{g}_{Bi} \cdot S_i - \gamma_i B_N \cdot \vec{g}_{Ni} \cdot I_i + I \cdot \vec{D} \cdot S \]  \hspace{1cm} (4.1)

We define the Zeeman term to be

\[ H_Z = -\gamma_i B_e \cdot \vec{g}_{Bi} \cdot S_i - \gamma_i B_N \cdot \vec{g}_{Ni} \cdot I_i \]  \hspace{1cm} (4.2)

and the Hyperfine Interaction term to be

\[ H_{hf} = I \cdot \vec{D} \cdot S \]  \hspace{1cm} (4.3)

\( H_Z \) is the Zeeman interaction between the magnetic induction and the spin operator on the \( i^{th} \) ion, where \( B_e \) and \( B_n \) are the electron and nuclear magnetic inductions, \( S_i \) and \( I_i \) are electron and nuclear spin operators, \( \vec{g}_{Bi} \) and \( \vec{g}_{Ni} \) are generally dimensionless tensors and \( \gamma_i \) is the gyromagnetic ratio of the particular nuclear species of the ion. (see ref 217, 219)

We take the external magnetic field to be

\[ H(t) = \hat{z}H_0 + \hat{x}H_1 \cos \omega t - \hat{y}H_1 \sin \omega t \]  \hspace{1cm} (4.4)
The magnetic inductions are then $B_e = \mu_B H(t)$ and $B_N = \mu_N H(t)$. For the following calculations, it is necessary to use the Hamiltonian in spin operator forms. For this specific calculation we use $I = \frac{1}{2}$ and $S = \frac{1}{2}$, and let

$$
\omega_B = \gamma_i g_B \mu_B H_0 \\
\Omega_B = \frac{1}{2} \gamma_i g_B \mu_B H_1 \\
\omega_N = \gamma_i g_N \mu_N H_0 \\
\Omega_N = \frac{1}{2} \gamma_i g_N \mu_N H_1
$$

We then obtain the following Hamiltonian. (See Appendix A for details of the calculation.)

$$
\mathcal{H} = -\omega_B S_z - \omega_N I_z - \Omega_B \left(e^{i\omega t} S_+ + e^{-i\omega t} S_-\right) \\
- \Omega_N \left(e^{i\omega t} I_+ + e^{-i\omega t} I_-\right) + \tilde{D} I_z S_z + \frac{\tilde{D}}{2} I_+ S_z + \frac{\tilde{D}}{2} I_+ S_-
$$

### 4.1.2 Spin Notation

There are four types of spin configurations: $|\uparrow\uparrow\rangle$, $|\uparrow\downarrow\rangle$, $|\downarrow\downarrow\rangle$, and $|\downarrow\uparrow\rangle$. In the $|F, F_z\rangle$ notation, $F^2 = (I + S)^2$ and here $I = S = \frac{1}{2}$. The spin configurations are defined as

$$
a = |0, 0\rangle, \quad b = |1, 0\rangle, \quad c = |1, -1\rangle, \quad d = |1, 1\rangle
$$

And in the $|m_s, m_I\rangle$ notation,

$$
e = |\frac{1}{2}, -\frac{1}{2}\rangle, \quad f = |\frac{1}{2}, \frac{1}{2}\rangle, \quad c = |\frac{1}{2}, -\frac{1}{2}\rangle, \quad d = |\frac{1}{2}, \frac{1}{2}\rangle
$$
Thus,

\[
a = \frac{e - f}{\sqrt{2}} \quad (4.10)
\]

\[
b = \frac{e + f}{\sqrt{2}} \quad (4.11)
\]

### 4.1.3 Quartic Equation for \(\gamma_i\)

Now operating on each of these four states, we obtain the following Hamiltonian equation matrix. (See Appendix A for details.)

\[
\mathbf{H} = \begin{pmatrix}
    a & b \\
    b & c \\
    c & d \\
    d & a
\end{pmatrix}
\]

\[
(4.12)
\]

This system of equations is now solved using the Schrödinger equation, \(\dot{\psi} = \mathcal{H}\psi\) and
with the substitutions $a = Ae^{-\frac{i\Omega}{4}}$, $b = Be^{-\frac{i\Omega}{4}}$, $c = Ce^{-i\omega t-\frac{i\Omega}{4}}$, $d = De^{i\omega t-\frac{i\Omega}{4}}$, which gives

$$
\begin{pmatrix}
\dot{A} \\
\dot{B} \\
\dot{C} \\
\dot{D}
\end{pmatrix} =
\begin{pmatrix}
-\tilde{D} & \frac{1}{2}(\omega_N - \omega_B) & -\frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) & \frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) \\
\frac{1}{2}(\omega_N - \omega_B) & 0 & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) \\
\frac{1}{\sqrt{2}}(\Omega_N - \Omega_B) & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) & -\omega + \frac{1}{2}(\omega_N + \omega_B) & 0 \\
\frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) & 0 & \omega - \frac{1}{2}(\omega_N + \omega_B)
\end{pmatrix}
\begin{pmatrix}
A \\
B \\
C \\
D
\end{pmatrix}
$$

(4.13)

Now we make further substitutions to simplify the matrix: $\Gamma = \frac{D - C}{\sqrt{2}}$ and $\Delta = \frac{D + C}{\sqrt{2}}$. And we let $\alpha = \frac{1}{2}(\omega_N - \omega_B)$, $\beta = \omega - \frac{1}{2}(\omega_N + \omega_B)$, $\delta = \Omega_N + \Omega_B$, and $\epsilon = \Omega_B - \Omega_N$. Which gives

$$
\begin{pmatrix}
\dot{A} \\
\dot{B} \\
\dot{\Gamma} \\
\dot{\Delta}
\end{pmatrix} =
\begin{pmatrix}
-\tilde{D} & \alpha & \epsilon & 0 \\
\alpha & 0 & 0 & -\delta \\
\epsilon & 0 & 0 & \beta \\
0 & -\delta & \beta & 0
\end{pmatrix}
\begin{pmatrix}
A \\
B \\
\Gamma \\
\Delta
\end{pmatrix}
$$

(4.14)

Then, since the equations do not depend explicitly upon the time, I make the following substitutions: $A = A_0 e^{i\gamma t}$, $B = B_0 e^{i\gamma t}$, $\Gamma = \Gamma_0 e^{i\gamma t}$, and $\Delta = \Delta_0 e^{i\gamma t}$. Which gives

$$
\begin{pmatrix}
-\gamma_i A_0 \\
-\gamma_i B_0 \\
-\gamma_i \Gamma_0 \\
-\gamma_i \Delta_0
\end{pmatrix} =
\begin{pmatrix}
-\tilde{D} & \alpha & \epsilon & 0 \\
\alpha & 0 & 0 & -\delta \\
\epsilon & 0 & 0 & \beta \\
0 & -\delta & \beta & 0
\end{pmatrix}
\begin{pmatrix}
A_0 \\
B_0 \\
\Gamma_0 \\
\Delta_0
\end{pmatrix} e^{i\gamma t}
$$

(4.15)

This system of algebraic equations can now be solved to obtain the following quartic equation
for $\gamma_i$.

\[
0 = [\gamma_i^2 - (\Omega_N + \Omega_B)^2 - (\omega - \frac{1}{2}(\omega_N + \omega_B))^2][\gamma_i^2 - \gamma_i \tilde{D} - (\Omega_B - \Omega_N)^2 - \frac{1}{2}(\omega_N - \omega_B))^2] \\
- [(\frac{1}{2}(\omega_N - \omega_B))(\Omega_N + \Omega_B) - (\omega - \frac{1}{2}(\omega_N + \omega_B))(\Omega_B - \Omega_N)]^2
\]

(4.16)

### 4.2 Coefficient Equations

In order to find the probability of a spin to flip, I need to find the coefficient of each wavefunction. The four states have the following wavefunctions.

\[
a(t) = e^{-\frac{i\Omega t}{4}}(a_1 e^{i\gamma_1 t} + a_2 e^{i\gamma_2 t} + a_3 e^{i\gamma_3 t} + a_4 e^{i\gamma_4 t}) = e^{-i\tilde{t}} \sum_{j=1}^{4} a_j e^{i\tilde{a}_j t} = \sum_{j=1}^{4} a_j e^{i\tilde{a}_j t} \quad (4.17)
\]

\[
b(t) = e^{-\frac{i\Omega t}{4}}(b_1 e^{i\gamma_1 t} + b_2 e^{i\gamma_2 t} + b_3 e^{i\gamma_3 t} + b_4 e^{i\gamma_4 t}) = e^{-i\tilde{t}} \sum_{j=1}^{4} b_j e^{i\tilde{b}_j t} = \sum_{j=1}^{4} b_j e^{i\tilde{b}_j t} \quad (4.18)
\]

\[
c(t) = e^{i\omega t-\frac{i\Omega t}{4}}(c_1 e^{i\gamma_1 t} + c_2 e^{i\gamma_2 t} + c_3 e^{i\gamma_3 t} + c_4 e^{i\gamma_4 t}) = e^{i\omega t-i\tilde{t}} \sum_{j=1}^{4} c_j e^{i\tilde{c}_j t} = \sum_{j=1}^{4} c_j e^{i\tilde{c}_j t} \quad (4.19)
\]

\[
d(t) = e^{-i\omega t-\frac{i\Omega t}{4}}(d_1 e^{i\gamma_1 t} + d_2 e^{i\gamma_2 t} + d_3 e^{i\gamma_3 t} + d_4 e^{i\gamma_4 t}) = e^{-i\omega t-i\tilde{t}} \sum_{j=1}^{4} d_j e^{i\tilde{d}_j t} = \sum_{j=1}^{4} d_j e^{i\tilde{d}_j t} \quad (4.20)
\]

To simplify notation, let $\sum_{j=1}^{4} = \sum$

\[
a(t) = \sum a_j e^{i\tilde{a}_j t} \quad (4.21)
\]

\[
b(t) = \sum b_j e^{i\tilde{b}_j t} \quad (4.22)
\]

\[
c(t) = \sum c_j e^{i\tilde{c}_j t} \quad (4.23)
\]

\[
d(t) = \sum d_j e^{i\tilde{d}_j t} \quad (4.24)
\]
This means I need to determine $a_1$, $a_2$, $a_3$, $a_4$ and similarly for $b$, $c$, and $d$. However, I can be selective in which coefficients I need to calculate based on my initial state. I will do the calculation for only the nuclear spin to flip. There are two ways that this can happen.

For instance, let’s pick the initial condition that we are in the $|\uparrow\uparrow\rangle$ state. Now I want to only flip the nuclear spin, so I need to calculate the coefficients for the state $e$, which put the system in the $|\uparrow\downarrow\rangle$ state.

Similarly, the system could initially be in the $|\downarrow\downarrow\rangle$ state. These states will be denoted by $\bar{a}, \bar{b}, \bar{c}, \bar{d}, \bar{e}$ and $\bar{f}$. This means $\bar{a}(0) = \bar{b}(0) = \bar{d}(0) = 0$ and $\bar{c}(0) = 1$. Again, I want to only flip the nuclear spin, so I need to calculate the coefficients for the state $\bar{f}$, which put the system in the $|\downarrow\uparrow\rangle$ state.

For the first case, I begin with the initial conditions $a(0) = b(0) = c(0) = 0$ and $d(0) = 1$. After lengthy calculations (see Appendix B) I find the following set of equation for the coefficients of state $e$.

\begin{align*}
\sum e_j &= 0 \quad (4.25) \\
\sum (\gamma_j - \frac{\tilde{D}}{4}) e_j &= \frac{1}{2}(-\epsilon + \delta) \quad (4.26) \\
\sum (\gamma_j - \frac{\tilde{D}}{4})^2 e_j &= \frac{1}{2}(-\frac{\tilde{D}}{2} \epsilon - \alpha \delta + \beta \epsilon - \frac{\tilde{D}}{2} \delta + \alpha \epsilon - \beta \delta) \quad (4.27) \\
\sum (\gamma_j - \frac{\tilde{D}}{4})^3 e_j &= \frac{1}{2}(-\frac{7\tilde{D}^2}{16} \epsilon - \frac{\tilde{D}}{4} \alpha \delta + \frac{\tilde{D}}{4} \beta \epsilon - \alpha^2 \epsilon + \alpha \beta \delta - \beta^2 \epsilon - \epsilon^3 \\
&\quad + \frac{\tilde{D}}{4} \alpha \epsilon + \alpha^2 \delta - \alpha \beta \epsilon + \frac{3\tilde{D}}{4} \beta \delta + \delta^3 + \delta \beta^2 + \frac{3\tilde{D}^2}{16} \delta) \quad (4.28)
\end{align*}

In the second case, I begin with the initial conditions $\bar{a}(0) = \bar{b}(0) = \bar{d}(0) = 0$ and $\bar{c}(0) = 1$. And again, after lengthy calculations (see Appendix B) I find the following set of equation
for the coefficients of state $\bar{f}$.

$$\sum \bar{f}_j = 0 \quad (4.29)$$

$$\sum (\gamma_j - \frac{\tilde{D}}{4})\bar{f}_j = \frac{1}{2}(-\epsilon + \delta) \quad (4.30)$$

$$\sum (\gamma_j - \frac{\tilde{D}}{4})^2\bar{f}_j = \frac{1}{2}(-\tilde{D} - \frac{\tilde{D}}{4}\epsilon + \alpha\delta - \beta\epsilon - \frac{\tilde{D}}{2}\delta - \alpha\epsilon + \beta\delta) \quad (4.31)$$

$$\sum (\gamma_j - \frac{\tilde{D}}{4})^3\bar{f}_j = \frac{1}{2}(-\tilde{D} - \frac{7\tilde{D}^2}{16}\epsilon + \frac{\tilde{D}}{4}\alpha\delta - \frac{\tilde{D}}{4}\beta\epsilon - \alpha^2\epsilon + \alpha\beta\delta - \beta^2\epsilon - \epsilon^3$$

$$- \frac{\tilde{D}}{4}\alpha\epsilon + \alpha^2\delta - \alpha\beta\epsilon - \frac{3\tilde{D}^2}{4}\beta\delta + \delta^3 + \delta\beta^2 + \frac{3\tilde{D}^2}{16}\delta) \quad (4.32)$$

### 4.3 Checking Limits

Since the resulting equations of the last section are quite complicated, it is necessary to check the limiting cases. This will be done in multiple steps. First I will check the limit in which we only have the nuclear Zeeman interaction, then only the electron Zeeman interaction, and finally the nuclear and electron Zeeman interaction but without the coupling term. In this section, I will simply let $\gamma_i \to \gamma$ for ease of notation, however, this is still only the term corresponding to one ion site.

#### 4.3.1 Nuclear Magnetic Interaction

The original quartic equation for $\gamma$ is

$$0 = [\gamma^2 - (\Omega_N + \Omega_B)^2 - (\omega - \frac{1}{2}(\omega_N + \omega_B))^2][\gamma^2 - \gamma\tilde{D} - (\Omega_B - \Omega_N)^2 - (\frac{1}{2}(\omega_N - \omega_B))^2]$$

$$- [(\frac{1}{2}(\omega_N - \omega_B))(\Omega_N + \Omega_B) - (\omega - \frac{1}{2}(\omega_N + \omega_B))(\Omega_B - \Omega_N)]^2 \quad (4.33)$$
In the limit where only the nuclear magnetic interaction is considered, I let $\tilde{D} \to 0$, $\Omega_B \to 0$, and $\omega_B \to 0$. Then the equation for $\gamma$ becomes

$$0 = (\gamma^2 - \Omega_N^2 - \frac{(\omega_N)^2}{2})(\gamma^2 - \Omega_N^2 - (\omega - \frac{\omega_N}{2})^2) - (\Omega_N[\omega - \frac{\omega_N}{2}] + \Omega_N[\frac{\omega_N}{2}])^2$$ (4.34)

This is a simple quadratic equation that can be solved, giving the following equations for $\gamma$.

$$\gamma_{\pm}^2 = \frac{1}{2}[2\Omega_N^2 + \omega^2 - \omega\omega_N + \frac{1}{2}\omega_N^2 \pm \omega\sqrt{4\Omega_N^2 + (\omega - \omega_N)^2}]$$ (4.35)

Now I let

$$Q = 2\Omega_N^2 + \omega^2 - \omega\omega_N + \frac{1}{2}\omega_N^2$$ (4.36)

$$R = \sqrt{4\Omega_N^2 + (\omega - \omega_N)^2}$$ (4.37)

Finally, the four equations for $\gamma$ are

$$\gamma_1 = \sqrt{\frac{1}{2}(Q + \omega R)}$$ (4.38)

$$\gamma_2 = \sqrt{\frac{1}{2}(Q - \omega R)}$$ (4.39)

$$\gamma_3 = -\sqrt{\frac{1}{2}(Q + \omega R)} = -\gamma_1$$ (4.40)

$$\gamma_4 = -\sqrt{\frac{1}{2}(Q - \omega R)} = -\gamma_2$$ (4.41)

As briefly mentioned above, the probability for the nuclear spin to flip will be a combination of two possibilities: One that starts in state d ($|\frac{1}{2}, \frac{1}{2}\rangle$) and flips to state e ($|\frac{1}{2}, -\frac{1}{2}\rangle$) and one that starts in state $\bar{c}$ ($|-\frac{1}{2}, -\frac{1}{2}\rangle$) and flips to state $\bar{f}$ ($|-\frac{1}{2}, \frac{1}{2}\rangle$).

The equations for the coefficients $e_1, e_2, e_3$, and $e_4$ needed in the first case are the following
in this limit.

\[ e_1 + e_2 + e_3 + e_4 = 0 \]  
(4.42)

\[ \gamma_1 e_1 + \gamma_2 e_2 - \gamma_1 e_3 - \gamma_2 e_4 = \Omega_N \]  
(4.43)

\[ \gamma_1^2 e_1 + \gamma_2^2 e_2 + \gamma_1^2 e_3 + \gamma_2^2 e_4 = -\Omega_N \omega \]  
(4.44)

\[ \gamma_1^3 e_1 + \gamma_2^3 e_2 - \gamma_1^3 e_3 - \gamma_2^3 e_4 = \Omega_N [\Omega_N^2 + \omega^2 - \frac{1}{2} \omega \omega_N + \frac{1}{4} \omega_N^2] \]  
(4.45)

And the equations for the second case are

\[ \bar{f}_1 + \bar{f}_2 + \bar{f}_3 + \bar{f}_4 = 0 \]  
(4.46)

\[ \gamma_1 \bar{f}_1 + \gamma_2 \bar{f}_2 - \gamma_1 \bar{f}_3 - \gamma_2 \bar{f}_4 = \Omega_N \]  
(4.47)

\[ \gamma_1^2 \bar{f}_1 + \gamma_2^2 \bar{f}_2 + \gamma_1^2 \bar{f}_3 + \gamma_2^2 \bar{f}_4 = \Omega_N \omega \]  
(4.48)

\[ \gamma_1^3 \bar{f}_1 + \gamma_2^3 \bar{f}_2 - \gamma_1^3 \bar{f}_3 - \gamma_2^3 \bar{f}_4 = \Omega_N [\Omega_N^2 + \omega^2 - \frac{1}{2} \omega \omega_N + \frac{1}{4} \omega_N^2] \]  
(4.49)

The probability in question is determined by

\[ P_{\text{nuclear}} = e_1^2 + e_2^2 + e_3^2 + e_4^2 + \bar{f}_1^2 + \bar{f}_2^2 + \bar{f}_3^2 + \bar{f}_4^2 \]  
(4.50)

After lengthy calculations of the coefficients of \( E \) and \( F \) (see Appendix C), the probability finally becomes the following.

\[ P_{\text{nuclear}} = \frac{4\Omega_N^2}{4\Omega_N^2 + (\omega - \omega_N)^2} \]  
(4.51)

This result is exactly as we expect it to be, since it is simply the case of one nucleus interacting with an external magnetic field.
4.3.2 Electron Zeeman Interaction

Now the limit in which the only interaction is the electron Zeeman interaction is considered. Again, I begin with the original quartic equation for $\gamma$.

\[
0 = [\gamma^2 - (\Omega_N + \Omega_B)^2 - (\omega - \frac{1}{2}(\omega_N + \omega_B))^2][\gamma^2 - \gamma \tilde{D} - (\Omega_B - \Omega_N)^2 - (\frac{1}{2}(\omega_N - \omega_B))^2]
- [(\frac{1}{2}(\omega_N - \omega_B))(\Omega_N + \Omega_B) - (\omega - \frac{1}{2}(\omega_N + \omega_B))(\Omega_B - \Omega_N)]^2
\] (4.52)

In the limit where only the electron Zeeman interaction is considered, I let $\tilde{D} \to 0$, $\Omega_N \to 0$, and $\omega_N \to 0$. Then the equation for $\gamma$ becomes

\[
0 = (\gamma^2 - \Omega_B^2 - (\frac{\omega_B}{2})^2)(\gamma^2 - \Omega_B^2 - (\omega - \frac{\omega_B}{2})^2) - (\Omega_B[\omega - \frac{\omega_B}{2}] + \Omega_B[\frac{\omega_B}{2}])^2
\] (4.53)

This equation is identical to the one in the nuclear Zeeman interaction limit, except that $\Omega_N$ is replaced with $\Omega_B$, and $\omega_N$ with $\omega_B$. Thus, my equations for $\gamma$ are

\[
\gamma_1 = \sqrt{\frac{1}{2}(Q + \omega R)} \quad (4.54)
\]

\[
\gamma_2 = \sqrt{\frac{1}{2}(Q - \omega R)} \quad (4.55)
\]

\[
\gamma_3 = -\sqrt{\frac{1}{2}(Q + \omega R)} = -\gamma_1 \quad (4.56)
\]

\[
\gamma_4 = -\sqrt{\frac{1}{2}(Q - \omega R)} = -\gamma_2 \quad (4.57)
\]
With $Q$ and $R$ defined as follows,

\begin{align*}
Q &= 2\Omega_B^2 + \omega^2 - \omega \omega_B + \frac{1}{2}\omega_B^2 \\
R &= \sqrt{4\Omega_B^2 + (\omega - \omega_B)^2}
\end{align*}

(4.58) (4.59)

Again, the probability for the electron spin to flip will be a combination of two possibilities:
One that starts in state $d$ ($|\frac{1}{2}, \frac{1}{2}\rangle$) and flips to state $f$ ($|-\frac{1}{2}, \frac{1}{2}\rangle$) and one that starts in state $\bar{c}$ ($|\frac{1}{2}, -\frac{1}{2}\rangle$) and flips to state $\bar{e}$ ($|\frac{1}{2}, -\frac{1}{2}\rangle$).

The equations for the coefficients $f_1, f_2, f_3,$ and $f_4$ needed in the first case are the following in this limit.

\begin{align*}
f_1 + f_2 + f_3 + f_4 &= 0 \\
\gamma_1 f_1 + \gamma_2 f_2 - \gamma_1 f_3 - \gamma_2 f_4 &= \Omega_B \\
\gamma_1^2 f_1 + \gamma_2^2 f_2 + \gamma_1^2 f_3 + \gamma_2^2 f_4 &= -\Omega_B \omega \\
\gamma_1^3 f_1 + \gamma_2^3 f_2 - \gamma_1^3 f_3 - \gamma_2^3 f_4 &= \Omega_B [\Omega_B^2 + \omega^2 - \frac{1}{2}\omega \omega_B + \frac{1}{4}\omega_B^2]
\end{align*}

(4.60) (4.61) (4.62) (4.63)

And the equations for the second case are

\begin{align*}
\bar{e}_1 + \bar{e}_2 + \bar{e}_3 + \bar{e}_4 &= 0 \\
\gamma_1 \bar{e}_1 + \gamma_2 \bar{e}_2 - \gamma_1 \bar{e}_3 - \gamma_2 \bar{e}_4 &= \Omega_B \\
\gamma_1^2 \bar{e}_1 + \gamma_2^2 \bar{e}_2 + \gamma_1^2 \bar{e}_3 + \gamma_2^2 \bar{e}_4 &= \Omega_B \omega \\
\gamma_1^3 \bar{e}_1 + \gamma_2^3 \bar{e}_2 - \gamma_1^3 \bar{e}_3 - \gamma_2^3 \bar{e}_4 &= \Omega_B [\Omega_B^2 + \omega^2 - \frac{1}{2}\omega \omega_B + \frac{1}{4}\omega_B^2]
\end{align*}

(4.64) (4.65) (4.66) (4.67)

The probability in question is determined by

\begin{equation}
P_{\text{electron}} = f_1^2 + f_2^2 + f_3^2 + f_4^2 + \bar{e}_1^2 + \bar{e}_2^2 + \bar{e}_3^2 + \bar{e}_4^2
\end{equation}

(4.68)
After lengthy calculations of the coefficients of $E$ and $F$ (see Appendix C), the probability finally becomes the following.

$$P_{electron} = \frac{4\Omega_B^2}{4\Omega_B^2 + (\omega - \omega_B)^2}$$

(4.69)

This result is exactly as we expect it to be, since it is simply the case of one electron interacting with an external magnetic field.

4.3.3 Uncoupled Electron Zeeman Interaction and Nuclear Magnetic Interaction

Next, I consider the limit in which only the coupling term of the electron and nucleus is neglected. I begin with the original quartic equation for $\gamma$.

$$0 = [\gamma^2 - (\Omega_N + \Omega_B)^2 - (\omega - \frac{1}{2}(\omega_N + \omega_B))^2][\gamma^2 - \gamma\tilde{D} - (\Omega_B - \Omega_N)^2 - \left(\frac{1}{2}(\omega_N - \omega_B)\right)^2]$$

$$- \left[\left(\frac{1}{2}(\omega_N - \omega_B)\right)(\Omega_N + \Omega_B) - (\omega - \frac{1}{2}(\omega_N + \omega_B))(\Omega_B - \Omega_N)\right]^2$$

(4.70)

Letting $\tilde{D} = 0$ gives,

$$0 = [\gamma^2 - (\Omega_N + \Omega_B)^2 - (\omega - \frac{1}{2}(\omega_N + \omega_B))^2][\gamma^2 - (\Omega_B - \Omega_N)^2 - \left(\frac{1}{2}(\omega_N - \omega_B)\right)^2]$$

$$- \left[\left(\frac{1}{2}(\omega_N - \omega_B)\right)(\Omega_N + \Omega_B) - (\omega - \frac{1}{2}(\omega_N + \omega_B))(\Omega_B - \Omega_N)\right]^2$$
This equation, although more complicated is still algebraically solvable. To simplify let,

\[ X = (\Omega_N + \Omega_B)^2 + (\omega - \frac{1}{2}(\omega_N + \omega_B))^2 \]  \hspace{1cm} (4.71)

\[ Y = (\Omega_B - \Omega_N)^2 + \left(\frac{1}{2}(\omega_N - \omega_B)\right)^2 \]  \hspace{1cm} (4.72)

\[ Z = \left[\frac{1}{2}(\omega_N - \omega_B)(\Omega_N + \Omega_B) - (\omega - \frac{1}{2}(\omega_N + \omega_B))(\Omega_B - \Omega_N)\right]^2 \]  \hspace{1cm} (4.73)

After some calculations, my four equations for \( \gamma \) are

\[ \gamma_1 = \sqrt{\frac{1}{2}(Q + R)} \]  \hspace{1cm} (4.74)

\[ \gamma_2 = \sqrt{\frac{1}{2}(Q - R)} \]  \hspace{1cm} (4.75)

\[ \gamma_3 = -\sqrt{\frac{1}{2}(Q + R)} = -\gamma_1 \]  \hspace{1cm} (4.76)

\[ \gamma_4 = -\sqrt{\frac{1}{2}(Q - R)} = -\gamma_2 \]  \hspace{1cm} (4.77)

Where

\[ Q = X + Y \]  \hspace{1cm} (4.78)

\[ R = \sqrt{(X - Y)^2 + 4Z} \]  \hspace{1cm} (4.79)

As before, we need to consider two cases in which the nuclear spin can flip to another state. The probability for this to happen is a combination of two possibilities: One that starts in state \( \text{d} \) (\(|\frac{1}{2}, \frac{1}{2}\rangle\)) and flips to state \( \text{e} \) (\(|\frac{1}{2}, -\frac{1}{2}\rangle\)) and one that starts in state \( \bar{\text{c}} \) (\(|-\frac{1}{2}, -\frac{1}{2}\rangle\)) and flips to state \( \bar{\text{f}} \) (\(|-\frac{1}{2}, \frac{1}{2}\rangle\)).

The equations for my coefficients \( e_1, e_2, e_3, \) and \( e_4 \) needed in the first case are the following
in this limit.

\[ e_1 + e_2 + e_3 + e_4 = 0 \] (4.80)

\[ \gamma_1 e_1 + \gamma_2 e_2 - \gamma_1 e_3 - \gamma_2 e_4 = \Omega_N \] (4.81)

\[ \gamma_1^2 e_1 + \gamma_2^2 e_2 + \gamma_1^2 e_3 + \gamma_2^2 e_4 = \Omega_N (\omega + \omega_B) \] (4.82)

\[ \gamma_1^3 e_1 + \gamma_2^3 e_2 - \gamma_1^3 e_3 - \gamma_2^3 e_4 = \Omega_N \left[ \Omega_N^2 + 3 \Omega_B^2 + \omega^2 - \frac{1}{2} \omega \omega_N - \frac{3}{2} \omega \omega_B + \frac{1}{4} \omega_N^2 + \frac{3}{4} \omega_B^2 \right] \] (4.83)

And the equations for the second case are

\[ \bar{f}_1 + \bar{f}_2 + \bar{f}_3 + \bar{f}_4 = 0 \] (4.84)

\[ \gamma_1 \bar{f}_1 + \gamma_2 \bar{f}_2 - \gamma_1 \bar{f}_3 - \gamma_2 \bar{f}_4 = \Omega_N \] (4.85)

\[ \gamma_1^2 \bar{f}_1 + \gamma_2^2 \bar{f}_2 + \gamma_1^2 \bar{f}_3 + \gamma_2^2 \bar{f}_4 = -\Omega_N (-\omega + \omega_B) \] (4.86)

\[ \gamma_1^3 \bar{f}_1 + \gamma_2^3 \bar{f}_2 - \gamma_1^3 \bar{f}_3 - \gamma_2^3 \bar{f}_4 = \Omega_N \left[ \Omega_N^2 + 3 \Omega_B^2 + \omega^2 - \frac{1}{2} \omega \omega_N - \frac{3}{2} \omega \omega_B + \frac{1}{4} \omega_N^2 + \frac{3}{4} \omega_B^2 \right] \] (4.87)

The probability for this limit will be called \( P_{B,N} \) and it is found using the following,

\[ P_{B,N} = e_1^2 + e_2^2 + e_3^2 + e_4^2 + \bar{f}_1^2 + \bar{f}_2^2 + \bar{f}_3^2 + \bar{f}_4^2 \] (4.88)

After lengthy calculations to be found in the Appendix, I finally obtain the following result.

\[ P_{B,N} = \frac{4 \Omega_N^2 [2 \Omega_B + (\omega - \omega_B)^2]}{(\omega - \omega_N)^2 (4 \Omega_B^2 + \frac{1}{2} (\omega - \omega_B)^2) + (\omega - \omega_B)^2 (4 \Omega_N^2 + \frac{1}{2} (\omega - \omega_N)^2) + 16 \Omega_B^2 \Omega_N^2} \] (4.89)
5.1 Second quantized Hamiltonian

We first rewrite $H_0$ in second quantization, using $I_i^r = \frac{1}{2} \sum_{\sigma=\pm} \sigma a_{i,\sigma}^\dagger a_{i,\sigma}$, $I_i^- = a_{i,-}^\dagger a_{i,-}$, $I_i^+ = a_{i,+}^\dagger a_{i,+}$, $S_{i,q}^r = \frac{1}{2} (\sum_{\sigma=\pm} \sigma b_{i,q,\sigma}^\dagger b_{i,q,\sigma}$, $S_{1,1}^+ = b_{1,q,+}^\dagger b_{1,q,+}$, and $S_{1,-}^+ = b_{1,q,-}^\dagger b_{1,q,-}$. These spin-1/2 fermion operators obey the anticommutation relations

\[
\{a_{i,\sigma}, a_{i',\sigma'}^\dagger\} = \delta_{i,i'} \delta_{\sigma,\sigma'}, \quad \{b_{i,q,\sigma}, b_{i',q',\sigma'}^\dagger\} = \delta_{i,i'} \delta_{q,q'} \delta_{\sigma,\sigma'}, \quad \{a_{i,\sigma}, b_{i',q',\sigma'}^\dagger\} = \{a_{i,\sigma}, b_{i',q',\sigma'}\} = \{a_{i,\sigma}, b_{i',q',\sigma'}\} = \{a_{i,\sigma}, b_{i',q',\sigma'}^\dagger\} = 0.
\]

We then incorporate the applied induction oscillatory time dependence into the operators themselves, setting $	ilde{a}_{i,\sigma} = a_{i,\sigma} e^{-i\sigma \omega_0 t/2}$, $	ilde{b}_{i,q,\sigma} = b_{i,q,\sigma} e^{-i\sigma \omega_0 t/2}$, and their Hermitian conjugates.

The bare Hamiltonian then becomes $\tilde{H}_0 = \tilde{H}_{n,0} + \tilde{H}_{e,0} + \tilde{H}_{hf}$, where

\[
\tilde{H}_{n,0} = -\frac{\omega_n}{2} \sum_{i,\sigma} \sigma \tilde{a}_{i,\sigma}^\dagger \tilde{a}_{i,\sigma} - \frac{\Omega_n}{2} \sum_{i,\sigma} \tilde{a}_{i,\sigma}^\dagger \tilde{a}_{i,-\sigma}, \quad (5.1)
\]

\[
\tilde{H}_{e,0} = -\frac{\omega_e}{2} \sum_{i,\sigma} \sigma \tilde{b}_{i,\sigma}^\dagger \tilde{b}_{i,\sigma} - \frac{\Omega_e}{2} \sum_{i,\sigma} \tilde{b}_{i,\sigma}^\dagger \tilde{b}_{i,-\sigma}, \quad (5.2)
\]

\[
\tilde{H}_{hf} = -\frac{D_z}{4} \sum_{i,q,\sigma,\sigma'} \sigma \sigma' \tilde{a}_{i,\sigma}^\dagger \tilde{a}_{i,-\sigma} \tilde{b}_{i,q,\sigma}^\dagger \tilde{b}_{i,q,-\sigma} - \frac{D_x}{2} \sum_{i,q,\sigma} \tilde{a}_{i,\sigma}^\dagger \tilde{a}_{i,-\sigma} \tilde{b}_{i,q,\sigma}^\dagger \tilde{b}_{i,q,-\sigma}, \quad (5.3)
\]

where $\sigma, \sigma' = \pm$. In order to calculate the Knight shift, it is essential to include the electrons in the conduction band(s). We therefore employ a variation on the Anderson model for magnetic impurities in a metal[53], which includes three terms, $\tilde{H}_A = \tilde{H}_{A,0} + \tilde{H}_{A,1} + \tilde{H}_{A,2}$.
\[\tilde{H}_{A,0} = \sum_{i,q,\sigma} \epsilon_q \hat{b}_{i,q,\sigma}^\dagger \hat{b}_{i,q,\sigma} + \frac{1}{2} \sum_{i,q,\sigma} U_q \hat{n}_{i,q,\sigma} \hat{n}_{i,q,-\sigma},\]  
(5.4)

\[\tilde{H}_{A,1} = \frac{1}{N_b} \sum_{i,q,j,\sigma} \int d^3r_j \left( v_{i,q,j} \hat{\psi}_{j,\sigma}^\dagger(r_j) \hat{b}_{i,q,\sigma} + H.c. \right) \delta(3)(r_j - r_i),\]  
(5.5)

\[\tilde{H}_{A,2} = \frac{1}{N_b} \sum_{j,\sigma,\sigma'} \int d^3r_j \hat{\psi}_{j,\sigma}^\dagger(r_j) \left( \sum_{\nu=1}^3 \frac{1}{2m_{j,\nu}} \left( \nabla_{j,\nu} / i - \sigma A_{j,\nu}(r_j) \right)^2 - \mu - \frac{\sigma \omega_j' e}{2} \right) \delta_{\sigma,\sigma'} \]  
\[-\frac{\Omega'_{j,e}}{2} \delta_{\sigma,-\sigma'} \hat{\psi}_{j,\sigma'}(r_j),\]  
(5.6)

where \(e\) is the electronic charge, \(A_j(r_j)\) is the magnetic vector potential at position \(r_j\) in band \(j\), \(\mu\) is the zero-field chemical potential, \(\hat{n}_{i,q,\sigma} = \hat{b}_{i,q,\sigma}^\dagger \hat{b}_{i,q,\sigma}\) is the local electron number operator, \(\hat{\psi}_{j,\sigma}(r_j) = \psi_{j,\sigma}(r_j)e^{-i\sigma \omega_0 t/2}\) as for the local electrons, where \(\psi_{j,\sigma}(r_j)\) annihilates an electron in the \(j\)th conduction band at position \(r_j\) and spin \(\sigma = \pm\). \(\omega_j' = |g_j \cdot B_0|\) is the Zeeman interaction and \((r_1, r_2, r_3) \equiv (x, y, z)\), \(N_b\) is the number of conduction bands, and we set \(\hbar = 1\). Note that \(\tilde{H}_{A,2}\) contains the term \(-\mu \hat{N}\) appropriate for the statistical grand canonical ensemble and the factor \(1/N_b\) appears in \(\tilde{H}_{A,1}\) and \(\tilde{H}_{A,2}\) to compensate for the overcounting of the conduction electrons in real space. For sufficiently weak spin-orbit coupling in the metal, \(g_j \cdot B_0\) defines the quantization axis direction of the conduction electron spin in an anisotropic but diagonal \(g_j\) tensor with elements \(g_{j,xx}, g_{j,yy},\) and \(g_{j,zz}\). Similarly, we define \(\Omega'_{j,e} = |g_j \cdot B_1|\), where \(g_j \cdot B_1\) is in the plane normal to \(g_j \cdot B_0\). This models the general normal (metallic) state conduction electron band structure without spin-orbit splitting as a sum of \(j\) ellipsoidal bands, each with effective masses \(m_{j,\nu}\). Although the time dependence of \(B\) is crucial for the nuclear magnetic resonance experiments, and plays a very important role in the flipping of the local and conduction electron spins, we neglect its influence on the orbital motion of the conduction electrons, setting \(A_j(r_j)\) to be independent of \(t\), and given
by the spatially uniform and time-independent $B_0 = \nabla_j \times A_j$. That is, the electron orbital motion is assumed to be in Landau orbits appropriate for the anisotropy of the conduction bands and the resulting Fermi surfaces.

It is useful to rewrite the conduction electron operators in position space, rather than the usual reciprocal space notation in the Anderson model[53]. That is, the diagrams relevant for the Knight shift involve the excitation of a local electron in orbital $q$ on the site $i \equiv r_i$ into an accessible conduction band, transport of that conduction electron away from that local site, the interaction of the magnetic induction upon the conduction electron’s motion and spin during its transport, the return of the conduction electron to the same site $i$ from which it was excited, the de-excitation of that conduction electron back into the same local orbital $q$ on that site, and then completing the hyperfine interaction with the nuclear spin probed in the resonance experiment.

In this model, we assume that the small parameters are the hyperfine interactions $D_z$ and $D_x$ and the $v_{i,q,j}$, the latter of which excites an electron from the $q^{\text{th}}$ local orbital at the probed nuclear species site $i$ into the $j^{\text{th}}$ conduction band while at that site. We note that in most metals, a given element probed by an NMR experiment contains more than one isotope. Although in a crystalline or microcrystalline solid, the positions of the particular probed element are determined by the crystal structure, if two or more isotopes of the probed element are present, the positions of the particular probed isotope form a disordered subset of this ordered crystalline array. In some cases, the probed isotope can exist in more than one sublattice, and can be distinguished by their different crystal field symmetries[1]. But with more than one isotope species, the sites indexed by $i$ on each disordered nuclear isotope sublattice can be treated as random.

In the Anderson model of magnetic impurities in a metal, the local impurity electrons are
assumed to have \(d\)-orbital symmetry, but the conduction band is assumed to be composed of only \(s\) and \(p\) orbitals, so that the local and conduction electron operators automatically anticommute. In the case of the Knight shift, the conduction bands are likely to be composed of a linear combination of a selection of the local electron orbital wave functions, but as soon as an electron is within a conduction band, it is delocalized, travels with an anisotropic Fermi velocity on the order of \(10^{-3}\) the speed of light in vacuum or greater, and can travel to essentially any site in the lattice, including the sites corresponding to different atomic species, so very shortly after the excitation of a local electron into a conduction band, the probability that it is still on the original probed nuclear site \(i\) is vanishingly small. Hence we assume

\[
\begin{align*}
\{\psi_{j,\sigma}(r_j), \psi_{j',\sigma'}(r'_{j'})\} &= \delta_{j,j'}\delta_{\sigma,\sigma'}\delta^{(3)}(r_j - r'_{j'}) \\
\{b_{i,q,\sigma}, \psi_{j,\sigma}(r_j)\} &= \{b_{i,q,\sigma}, \psi_{j,\sigma}(r_j)\} = \{a_{i,\sigma}, \psi_{j,\sigma}(r_j)\} = \{a_{i,q,\sigma}, \psi_{j,\sigma}(r_j)\} = 0.
\end{align*}
\]

In order to evaluate the leading contributions to the Knight shift, we shall use a diagrammatic procedure to write the nuclear and local electronic Green functions to first order in the hyperfine interactions and the local and conduction electron Green functions to second order in the Anderson excitation energy \(v_{i,q,j}\). Thus, we break up \(\tilde{H}\) into two parts, \(\tilde{H} = \tilde{H}_0 + \tilde{V}\), where \(\tilde{V} = \tilde{H}_{hf} + \tilde{H}_{A,1}\), and \(\tilde{H}_0 = \tilde{H}_{n,0} + \tilde{H}_{s,0} + \tilde{H}_{A,0} + \tilde{H}_{A,2}\). We will use the standard equation of motion technique to solve for the bare nuclear and conduction electron Green functions, and a second order equation of motion technique to approximate the correlation effects in the Coulomb interactions \(U_q\) between opposite spin electrons in the same local orbital that will eventually mix with the relevant conduction band(s)[54]. For the conduction electrons with an anisotropic effective mass, we calculate the bare Green function in position space [55, 56], and transform each ellipsoidal Fermi surface to a spherical one using the Klemm-Clem transformations[56].

We note that in the original model of Yosida[22], the only interaction was the last term of \(\tilde{H}_{A,2}\) [Eq. (10)], as the nucleus was completely neglected, and the measured Knight shift in
the nuclear resonance frequency was assumed to be proportional to the conduction electron spin susceptibility probed by the oscillatory magnetic induction $B_1(t)$ in the direction normal to the conduction electron spin quantization axis, and hence $B_0$, the hyperfine interactions, and the time dependence of $B_1$ were neglected. Although in simple metals and superconductors with single isotropic Fermi surfaces and no electronic correlations, this model has been effective in providing a qualitative understanding of the Knight shift measurements, as noted above, there are many examples of layered and quasi-one-dimensional superconductors in which it has led to results inconsistent with thermodynamic upper critical field and/or scanning tunneling microscopy results, leading to a great deal of confusion. Here we study this much more detailed model for the Knight shift in anisotropic and correlated metals, as a preliminary study for the background of a much more quantitative model of the Knight shift in anisotropic and correlated superconductors.

5.2 Equations of Motion

In using the equation of motion technique, it is important to remember that the operators $\tilde{a}_{i,\sigma}$, $\tilde{b}_{i,q,\sigma}$, and $\tilde{c}_{j,k,\sigma}$ all depend explicitly upon the time via their mutual factor $e^{-i\sigma\omega_0t/2}$, so that the total time evolution of each of these operators $\hat{O}$ is given by

$$\frac{d\hat{O}}{dt} = \frac{\partial \hat{O}}{\partial t} - i \left[ \hat{O}, \hat{H}_0 \right].$$

(5.7)
The zero-temperature Green functions for the nuclei and local and conduction electrons are defined respectively as

\[ \tilde{G}_{n,i,i'}^{\sigma,\sigma'}(t, t') \equiv -i \langle T \left[ \tilde{a}_{i,\sigma}(t) \tilde{a}_{i',\sigma'}^\dagger(t') \right] \rangle, \]  
(5.8)

\[ \tilde{G}_{le,i,i'}^{q,q',\sigma,\sigma'}(t, t') \equiv -i \langle \left[ \tilde{b}_{i,q,\sigma}(t) \tilde{b}_{i',q',\sigma'}^\dagger(t') \right] \rangle, \]  
(5.9)

\[ \tilde{G}_{ce,j,j'}^{\sigma,\sigma'}(r_j, t; r_{j'}, t') \equiv -i \langle \left[ \tilde{\psi}_{j,\sigma}(r_j, t) \tilde{\psi}_{j',\sigma'}^\dagger(r_{j'}, t') \right] \rangle, \]  
(5.10)

where \( T \) is the time-ordering operator. We define the zeroth order nuclear spin and conduction electron spin Green functions to be those functions in the absence of the other fermions.

### 5.2.1 Nuclear spin Green function

The nuclear spin Green function then satisfies

\[ \frac{i}{\hbar} \frac{d}{dt} \tilde{G}_{n,i,i',\sigma,\sigma'}^{m,0}(t, t') = \delta_{i,i'} \delta_{\sigma,\sigma'} \delta(t - t') + \frac{\sigma(\omega_0 - \omega_n)}{2} \tilde{G}_{n,i,i',\sigma,\sigma'}^{m,0}(t - t') \]  
\[ - \frac{\Omega_n}{2} \tilde{G}_{n,i,i',-\sigma,\sigma'}^{m,0}(t, t'). \]  
(5.11)

Since the only explicit \( t, t' \) dependencies are in the \( \delta(t - t') \) driving function, \( \tilde{G}_{n,i,i',\sigma,\sigma'}^{m,0} \) must be only a function of \( t - t' \), and Fourier transformation with respect to the time yields

\[ \left( \omega - \frac{\sigma(\omega_0 - \omega_n)}{2} \right) \tilde{G}_{n,i,i',\sigma,\sigma'}^{m,0}(\omega) = \delta_{i,i'} \delta_{\sigma,\sigma'} - \frac{\Omega_n}{2} \tilde{G}_{n,i,i',-\sigma,\sigma'}^{m,0}(\omega), \]  
(5.12)

which is a \( 2 \times 2 \) spin matrix that is easily solved to yield

\[ \tilde{G}_{n,i,i',\sigma,\sigma'}^{m,0}(\omega) = \frac{\delta_{i,i'}}{\omega^2 - R_n^2 + i\delta} \left[ \delta_{\sigma,\sigma'} \left( \omega + \sigma(\omega_0 - \omega_n)/2 \right) - \delta_{\sigma,-\sigma'} \Omega_n/2 \right], \]  
(5.13)
where

\[ R_n = \frac{1}{2} \sqrt{(\omega_0 - \omega_n)^2 + \Omega_n^2} \]  

(5.14)

and \( \delta \to 0^+ \).

As shown in the appendix, this \( 2 \times 2 \) matrix \( \hat{G}_{i,i}^{n,0}(\omega) \) may be written in terms of the Pauli matrices,

\[ \hat{G}_{i,i}^{n,0}(\omega) = \frac{1}{\omega \sigma_0 - \sigma_3(\omega_0 - \omega_n)/2 + \sigma_1 \Omega_n/2}, \]  

(5.15)

allowing us to incorporate the self-energy Feynman diagrams using the matrix form of Dyson’s equation [55],

\[ [\hat{G}_{i,i}(\omega)]^{-1} = \hat{G}_{i,i}^{n,0}(\omega) - \hat{\Sigma}^n(\omega). \]  

(5.16)

5.2.2 Local orbital electron spin Green function

For the local orbital electrons, the analogous procedure leads to

\[ i \frac{d}{dt} \tilde{G}^{le,0}_{i,i',q,q',\sigma,\sigma'}(t,t') = \delta_{i,i',q,q'}(t-t') \left( \frac{\sigma(\omega_0 - \omega_i)}{2} + \epsilon_q \right) \hat{G}^{le,0}_{i,i',q,q',\sigma,\sigma'}(t,t') - \frac{\Omega_e}{2} \tilde{G}^{le,0}_{i,i',q,q',\sigma,\sigma'}(t,t') + U_q \tilde{F}^{le,0}_{i,i',q,q',\sigma,\sigma'}(t,t'), \]  

(5.17)

where

\[ \tilde{F}^{le,0}_{i,i',q,q',\sigma,\sigma'}(t,t') \equiv -i \langle T \tilde{b}_{i,q,\sigma}(t)\tilde{b}_{i',q',\sigma'}^\dagger(t)\tilde{b}_{i,q,\sigma''}(t)\tilde{b}_{i',q',\sigma'''}^\dagger(t') \rangle. \]  

(5.18)
That is, the local electron correlation effects lead to this two-particle Green function. In order to make a reasonable approximation of these correlation effects, we use the equation of motion technique for this two-particle Green function, which in turn leads to a three-particle (six-operator) Green function, etc., so that one needs to find some way to truncate this procedure. If one were to truncate the equation of motion for the single particle Green function in Eq. (19) and Fourier transform it with respect to the time, one would obtain

$$
\tilde{F}_{le,0}^{i,i';q,q';\sigma,\sigma'}(\omega) \approx \langle \hat{n}_{i,q,-\sigma} \rangle \tilde{G}_{le,0}^{i,i';q,q';\sigma,\sigma'}(\omega),
$$

which would lead to a strong underestimation of the correlation effects. However, truncation of the equation of motion for the two-particle Green function $\tilde{F}_{le,0}$ in first order accounted for the electron correlation effects rather well in the strong $U_q$ limit (but with no applied magnetic field), as shown by Lacroix[54]. From the equation of motion method for the two-body Green function, and decoupling the three-body Green functions into two-body functions times an equal-time, equal spin, and equal orbital state density expectation value, after Fourier transformation with respect to the time, one obtains

$$
\left( \omega - \epsilon_q - \frac{\sigma(\omega_q - \omega_0)}{2} \right) \tilde{F}_{le,0}^{i,i';q,q';\sigma,\sigma'}(\omega) \approx \delta_{i,i'} \delta_{q,q'} \delta_{\sigma,\sigma'} \langle \hat{n}_{i,q,-\sigma} \rangle - \frac{\Omega_e}{2} \Delta \tilde{F}_{le,0}(\omega),
$$

where

$$
\Delta \tilde{F}_{le,0}(\omega) = \tilde{F}_{le,0}^{i,i';q,q';\sigma,\sigma'}(\omega) - \tilde{F}_{le,0}^{i,i';q,q';\sigma,-\sigma'}(\omega) + \tilde{F}_{le,0}^{i,i';-q,-q';\sigma,\sigma'}(\omega).
$$

We note that $-(\Omega_e/2)\Delta \tilde{F}_{le,0}(\omega)$ involves spin flips of the correlated orbital electrons, and is thus much smaller than the correlation energies, so that it can be safely neglected. Thus,
the approximation for $\tilde{F}^{le,0}(\omega)$ that takes account of most of the Coulomb correlation effects is

$$\tilde{F}^{le,0}_{i,i',q,q';\sigma,\sigma'}(\omega) \approx \frac{\delta_{i,i'}\delta_{q,q'}\delta_{\sigma,\sigma'}\langle \hat{n}_{i,q,-\sigma} \rangle}{\omega - \epsilon_q - \sigma(\omega_0 - \omega_e)/2 - 2U_q\langle \hat{n}_{i,q,-\sigma} \rangle}, \quad \text{(5.22)}$$

Thus, one may then solve the $2 \times 2$ matrix equation for $\tilde{G}^{le,0}_{i,i',q,q';\sigma,\sigma'}(\omega)$, and obtain

$$\tilde{G}^{le,0}_{i,i',q,q';\sigma,\sigma'}(\omega) \approx \frac{\delta_{i,i'}\delta_{q,q'}Z_{i,q,\sigma'}(\omega)}{(\omega - \epsilon_q)^2 - R_e^2 + i\delta} \left[ \delta_{\sigma,\sigma'}\left( \omega - \epsilon_q + \frac{\sigma(\omega_0 - \omega_e)}{2} \right) - \delta_{\sigma,-\sigma'}\frac{\Omega_e}{2} \right], \quad \text{(5.23)}$$

where

$$Z_{i,q,\sigma'}(\omega) = 1 + \frac{U_q\langle \hat{n}_{i,q,-\sigma'} \rangle}{\omega - \epsilon_q - \sigma(\omega_0 - \omega_e)/2 - 2U_q\langle \hat{n}_{i,q,-\sigma} \rangle + i\delta}, \quad \text{(5.24)}$$

$$R_e = \frac{1}{2}\sqrt{(\omega_0 - \omega_e)^2 + \Omega_e^2}, \quad \text{(5.25)}$$

where $\delta = 0^+$, precisely as for nuclear spin Green function in Eq. (17), except for the energies $\epsilon_q$ of the local orbital electrons and the amplitude renormalization factor $Z_{i,q,\sigma'}(\omega)$. We note that the effects of electron correlation are all contained in this factor, and that as $U_q \to 0$ and $U_q \to \infty$, this renormalization factor approaches the values 1 and $\frac{1}{2}$, respectively.

The mean-field expectation value $\langle \hat{n}_{i,q,-\sigma} \rangle$ is obtained self-consistently from

$$\langle \hat{n}_{i,q,\sigma} \rangle = -\frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \text{Im} \left[ \tilde{G}^{le,0}_{i,i',q,q';\sigma,\sigma'}(\omega) \right]. \quad \text{(5.26)}$$

Performing the contour integral with the aid of symbolic programming software, we obtain
the self-consistency conditions for $\sigma = \pm$,

$$
\langle \hat{n}_{i,q} \rangle = 1 - \frac{U_q \langle \hat{n}_{i,q,-\sigma} \rangle \left[ \sigma (\omega_0 - \omega_b)/2 + R_e \right]}{R_e \left( \sigma (\omega_0 - \omega_b)/2 + 2U_q \langle \hat{n}_{i,q,-\sigma} \rangle - R_e \right)}.
$$

(5.27)

This formula does not apply if the local orbital is completely unoccupied, but accounts for the Coulomb correlations in the case in which it is occupied by one or two electrons. We note that for an occupied orbital in the limit $U_q \to 0$, $\langle \hat{n}_{i,q} \rangle \to 1$, but in the limit $U_q \to \infty$, $\langle \hat{n}_{i,q} \rangle \to \frac{1}{2} - \sigma (\omega_0 - \omega_b)/(4R_e) \approx (1 + \sigma)/2$, since an NMR experiment is performed for $\omega_0 \approx \omega_b \ll \omega_e$. In this limit, the strong Coulomb repulsion greatly deters double occupancy, as expected.

We note that if we had used Eq. (23) for the last term in the Fourier transform of Eq. (21) and solved for $\tilde{G}_{i,i';q,q';\sigma,\sigma'}(\omega)$, one would have obtained Eq. (27) in the limits $Z_{i,q,\sigma'}(\omega) = 1$ but $\epsilon_q \to \epsilon_q + U_q \langle \hat{n}_{i,q,-\sigma} \rangle$, which is just an uncorrelated shift in the orbital energy $\epsilon_q$. As Lacroix pointed out [54], the approximation in Eq. (26) is a much better representation of the strong electron correlation effects than is that in Eq. (23).
5.2.3 *Insulator Feynman diagrams*

Figure 5.1: Feynman diagram of a Knight shift for a nucleus in an insulator.

Figure 5.2: Feynman diagram of a linewidth change for a nucleus in an insulator.
In an insulator, the Feynman diagrams for the corrections to the nuclear magnetic resonance frequency and its linewidth first order in the hyperfine interactions are pictured in Figure 5.1 and Figure 5.2. The solid vertical line is the bare nuclear Green function, the wiggly line is the hyperfine interaction, and the dashed curve is the local electron at the same site as the probed nucleus. The diagrams in may be written in terms of their frequency Fourier transforms as

\[
\tilde{G}_{n,1}^{n,1a}(\omega) = \frac{D_z}{4} \sum_{\sigma''',\sigma''} \sigma'' \sigma'''' \tilde{G}_{n,0}^{n,0}(\omega) \tilde{G}_{i,i;\sigma''',\sigma''}^{m,0}(\omega) \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \tilde{G}_{i,i;\sigma''',\sigma''}^{de,0}(\omega'),
\]

\[
\tilde{G}_{n,1}^{n,1b}(\omega) = \frac{D_x}{2} \sum_{\sigma''',\sigma''} \tilde{G}_{i,i;\sigma''',\sigma''}^{n,0}(\omega) \tilde{G}_{i,i;\sigma''',\sigma''}^{n,0}(\omega) \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \tilde{G}_{i,i;\sigma''',\sigma''}^{de,0}(\omega'),
\]

where we included the factor of -1 for a single fermion loop. In our first quantization perturbation calculation in the absence of the Coulomb correlation effects, there were no such terms. For the diagram shown in Figure 5.1, it is easy to show that

\[
\sum_{\sigma'''} \sigma'' \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \tilde{G}_{i,i;\sigma''',\sigma''}^{de,0}(\omega') = -\frac{i}{2} \sum_{\sigma'''} \sigma''\langle \hat{n}_{i,q;\sigma'''} \rangle,
\]

and the remainder of the diagram is listed in the appendix. The diagram pictured in Figure 5.2 contains the integral

\[
I_{1b}^{le}(\sigma''') = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \tilde{G}_{i,i;\sigma''',\sigma''}^{de,0}(\omega'),
\]
may be evaluated using symbolic programming software, and the result is listed in the appendix. In the limits $U_q \to 0, \infty$,

$$
\sum_{\sigma''} I^{1b}(\omega'') \to 2iK_{1b} \frac{\Omega_e}{R_e},
$$

(5.32)

where $K_{1b} = \frac{1}{4}$ for $U_q = 0$ and $K_{1b} = \frac{1}{8}$ for $U_q \to \infty$. Hence, the diagrams pictured in Figures 5.1-5.2 give rise to the self-energy corrections to the nuclear Green function,

$$
\tilde{\Sigma}^{1a}_{n} = i\sigma_3 \frac{D_z}{8} \sum_{q,\sigma''} \sigma''(\tilde{n}_{i,q,\sigma''}) ,
$$

(5.33)

$$
\tilde{\Sigma}^{1b}_{n} = \frac{D_x}{2} \sigma_1 \sum_{\sigma''} I^{1e}_{1b}(\sigma''') \approx iD_x \sigma_1 K_{1b} \frac{\Omega_e}{R_e},
$$

(5.34)

which provide finite lifetimes to the diagonal and off-diagonal nuclear spin states due to the correlations of its surrounding local orbital electrons when $B_0, B_1 \neq 0$.

5.2.4 Conduction electron Green function

After Fourier transformation with respect to the time, the bare conduction electron Green function similarly satisfies

$$
\left(\omega - \frac{\sigma(\omega_0 - \omega_{j,e})}{2}\right) \tilde{G}_{j,j';\sigma,\sigma'}^{ce,0}(\mathbf{r}_j, \mathbf{r}_{j'}; \omega) = \delta_{j,j'} \delta_{\sigma,\sigma'} \delta^{(3)}(\mathbf{r}_j - \mathbf{r}_{j'})
$$

$$
+ \left( \sum_{\nu} \frac{1}{2m_{j,\nu}} \left( \nabla_{j,\nu}/1 - eA_{j,\nu}(\mathbf{r}_j) \right)^2 - \mu \right)
$$

$$
\times \tilde{G}_{j,j';\sigma,\sigma'}^{ce,0}(\mathbf{r}_j, \mathbf{r}_{j'}; \omega)
$$

$$
- \frac{\Omega_{j,e}}{2} \tilde{G}_{j,j';-\sigma,\sigma'}^{ce,0}(\mathbf{r}_j, \mathbf{r}_{j'}; \omega).
$$

(5.35)
Note that in the present approximation, the vector potential is assumed independent of the time, but the time-dependent part of the applied field can flip the conduction electron spins via the Zeeman interaction.

For a single conduction band with a general ellipsoidal electronic dispersion, the Klemm-Clem transformations were employed to transform the band to one of spherical symmetry,[57] whereby

\[
\sum_{\nu=1}^{3}(\nabla/\iota - eA_{\nu})^{2}/(2m_{\nu}) \rightarrow (\tilde{\nabla}/\iota - e\tilde{A})^{2}/(2m),
\]

where

\[
\tilde{\nabla} = \nabla/\alpha, \\
\tilde{A} = A/\alpha, \\
\tilde{B}_{0} = B_{0}\alpha, \\
m = (m_{1}m_{2}m_{3})^{1/3}, \\
\alpha(\theta, \phi) = [m_{1}\sin^{2}\theta \cos^{2}\phi + m_{2}\sin^{2}\theta \sin^{2}\phi + m_{3}\cos^{2}\theta]^{1/2}, \\
m_{\nu} = m_{\nu}/m.
\]

The eigenstates of the orbital motion of an electron in the presence of a constant \( B_{0} \) are free particle states parallel to \( \tilde{B}_{0} \) and harmonic oscillator states for the Landau orbits in the transformed plane perpendicular to \( \tilde{B}_{0} \), with eigenvalues[57]

\[
\varepsilon(n, \tilde{k}_{\parallel}) = \frac{\tilde{k}_{\parallel}^{2}}{2m} + \frac{e\tilde{B}_{0}(n + 1/2)}{m} = \frac{k_{\parallel}^{2}}{2m\alpha^{2}} + \frac{(n + 1/2)eB_{0}\alpha}{m}.
\]

This transformation involves the anisotropic scale transformation that satisfies the Maxwell
equation $\nabla \cdot B_0 = 0$ while preserving the magnitude of $B_0$, and then a rotation of the transformed $B'_0$ direction to the crystallographic $z$-axis direction. As shown elsewhere, this transformation can be generalized to more than one anisotropic conduction band, each with generally different ellipsoidal electronic band dispersions [57]. Here we employ this powerful technique to transform each of the $j$ ellipsoidal bands separately,

$$\sum_{\nu}(\nabla_{j,\nu}/i - eA_{j,\nu})^2/2m_{j,\nu} \to (\tilde{\nabla}_j/i - e\tilde{A}_j)^2/(2m_j), \quad (5.44)$$

where

$$\tilde{\nabla}_j = \nabla_j/\alpha_j, \quad (5.45)$$
$$\tilde{A}_j = A_j/\alpha_j, \quad (5.46)$$
$$\tilde{B}_{j,0} = B_0\alpha_j, \quad (5.47)$$
$$m_j = (m_{j,1}m_{j,2}m_{j,3})^{1/3}, \quad (5.48)$$
$$\alpha_j = [m_{j,1}\sin^2 \theta \cos^2 \phi + m_{j,2}\sin^2 \theta \sin^2 \phi + m_{j,3}\cos^2 \theta]^{1/2}. \quad (5.49)$$

For band $j$, the orbital eigenstates are then

$$\varepsilon_j(n_j, k_{j,||}) = \frac{k_{j,||}^2}{2m_j \alpha_j^2} + \frac{(n_j + 1/2)eB_0\alpha_j}{m_j}, \quad (5.50)$$

where $n_j = 0, 1, 2, \ldots$ are the two-dimensional harmonic oscillator (Landau level) quantum numbers for band $j$.

We therefore are able to fully transform the conduction electron Green function equation of
motion, obtaining

\[
\omega - \frac{1}{2m_j} \left[ -(\nabla_{j,\|})^2 + (\nabla_{j,\perp}/i - e\tilde{A}_{j,\perp})^2 \right] + \mu \\
- \frac{2}{\sigma(\omega_0 - \tilde{\omega}_{j,e}')} \left\{ \tilde{\omega}_{j,e}' \delta_{j,j'} \delta_{\sigma,\sigma'} \delta_{j,j}' \delta^{(3)}(\tilde{\mathbf{r}}_j - \tilde{\mathbf{r}}'_j) \right\} = \delta_{j,j'} \delta_{\sigma,\sigma'} \delta^{(3)}(\tilde{\mathbf{r}}_j - \tilde{\mathbf{r}}'_j)
\]

(5.51)

where

\[
\tilde{\omega}_{j,e}' = B_0 \beta_j(\theta, \phi), \quad (5.52)
\]

\[
\tilde{\Omega}_{j,e}' = B_1 \gamma_j(\theta, \phi), \quad (5.53)
\]

and \(\beta_j(\theta, \phi)\) and \(\gamma_j(\theta, \phi)\) are given in the appendix.

We note that both the free-particle states \(e^{i\mathbf{k}_{j,\|} \cdot \tilde{\mathbf{r}}_{j,\|}}\) for the motion parallel to the transformed \(\tilde{\mathbf{B}}_{j,0}\) and the two-dimensional harmonic oscillator states \(|u_{n_j}(\tilde{\mathbf{r}}_{j,\perp})\rangle\) in the transformed planes perpendicular to \(\mathbf{B}_{j,0}\) in each of the \(j\) bands are complete orthonormal states, so we write

\[
\delta^{(3)}(\tilde{\mathbf{r}}_j - \tilde{\mathbf{r}}'_j) \rightarrow \frac{eB_0 \alpha_j}{2\pi N_b} \sum_{n_j=0}^{\infty} |u_{n_j}(\tilde{\mathbf{r}}_{j,\perp})\rangle \langle u_{n_j}(\tilde{\mathbf{r}}'_{j,\perp})| \int \frac{d\mathbf{k}_{j,\|}}{2\pi} e^{i\mathbf{k}_{j,\|} \cdot (\mathbf{r}_{j,\|} - \mathbf{r}'_{j,\|})},
\]

(5.54)

where \(eB_0 \alpha_j/(2\pi)\) is the transformed Landau degeneracy factor per spin for band \(j\). Note that \(e\) is the magnitude of the electronic charge. We then expand the bare conduction
electron Green function in terms of these free particle and harmonic oscillator states,

$$
\tilde{G}^{ce,0}_{j,j':\sigma,\sigma'}(\tilde{r}_j,\tilde{r}'_{j'},\omega) \rightarrow \frac{eB_0\alpha_j}{2\pi N_0} \sum_{n_{j}=0}^{\infty} |u_{n_{j}}(\tilde{r}_{j,\perp})\rangle \langle u_{n_{j}}(\tilde{r}'_{j',\perp})| \int \frac{d\tilde{k}_{j,||}}{2\pi} \tilde{e}^{i\tilde{k}_{j,||}(\tilde{r}_{j,||}-\tilde{r}'_{j',||})} \times \tilde{G}^{ce,0}_{j,j':\sigma,\sigma'}(\tilde{k}_{j,||},n_{j},\omega), \quad (5.55)
$$

leading to

$$
\left[ \omega - \varepsilon_j(n_{j},\tilde{k}_{j,||}) + \mu - \frac{\sigma(\omega_0 - \tilde{\omega}'_{j,e})}{2} \right] \tilde{G}^{ce,0}_{j,j':\sigma,\sigma'}(\tilde{k}_{j,||},n_{j},\omega) = \delta_{j,j'}\delta_{\sigma,\sigma'} - \frac{\tilde{\Omega}'_{j,e}}{2} \tilde{G}^{ce,0}_{j,j',-\sigma,\sigma'}(\tilde{k}_{j,||},n_{j},\omega), \quad (5.56)
$$

where \( \varepsilon_j(n_{j},\tilde{k}_{j,||}) \) is given by Eq. (54) with \( \tilde{k}_{j,||} = k_{j,||}/\alpha_j \).

As for the nuclear spin, the resulting 2 × 2 conduction electron spin matrix is easily solved, leading to

$$
\tilde{G}^{ce,0}_{j,j':\sigma,\sigma'}(\tilde{k}_{j,||},n_{j},\omega) = \delta_{j,j'} \frac{\delta_{\sigma,\sigma'}}{(\omega - \varepsilon_j(n_{j},\tilde{k}_{j,||}) + \mu)^2 - (\tilde{R}'_{j,e})^2 + i\delta \text{ sgn}[\varepsilon_j(n_{j},\tilde{k}_{j,||}) - \mu - \sigma \tilde{R}'_{j,e}]} \times \left[ \delta_{\sigma,\sigma'} \left( \omega - \varepsilon_j(n_{j},\tilde{k}_{j,||}) + \mu + \sigma(\omega_0 - \tilde{\omega}'_{j,e})/2 \right) - \delta_{\sigma,-\sigma} \tilde{\Omega}'_{j,e}/2 \right], \quad (5.57)
$$

where

$$
\tilde{R}'_{j,e} = \frac{1}{2}\sqrt{(\omega_0 - \tilde{\omega}'_{j,e})^2 + (\tilde{\Omega}'_{j,e})^2}. \quad (5.58)
$$

Note that the infinitesimal imaginary part of the denominator is different for the Zeeman-split effective chemical potentials. We note that Eq. (61) is remarkably similar to the
expressions for $\tilde{G}_{i,i;\sigma,\sigma'}^{m,0}(\omega)$ and $\tilde{G}_{i,i';q,q';\sigma,\sigma'}^{le,0}(\omega)$ in Eqs. (17) and (27), respectively.

5.3 Knight resonant frequency and linewidth shifts

Figure 5.3: Feynman diagram of the Knight shift for a nucleus in a metal.
Figure 5.4: Feynman diagram of a linewidth change for a nucleus in a metal.

Figure 5.5: Feynman diagram of a linewidth change for a nucleus in a metal.
The three contributions to the Knight resonance frequency and linewidth shifts linear in the hyperfine interactions and quadratic in the excitation energy $v$ are given by the diagrams in Figures 5.3-5.5. The solid vertical line is the bare nuclear Green function, the wiggly line is the hyperfine interaction, the dashed curve is the local electron at the same site as the probed nucleus, the stars represent the excitation into one of the allowed conduction bands, and the solid curves are the conduction electron Green functions. We have

$$
\tilde{G}^{m,2a}_{i,i;\sigma,\sigma'}(\omega) = \frac{D_z}{4} \sum_{\sigma'',\sigma'''q,j} \sigma'' \tilde{G}^{m,0}_{i,i;\sigma,\sigma''}(\omega) \tilde{G}^{m,0}_{i,i;\sigma'',\sigma'}(\omega) \times |v_{i,q,j}|^2 \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} [\tilde{G}^{\text{le},0}_{i,i;q,q;\sigma'',\sigma'''}(\omega')]^2 \\
\times eB_0 \alpha_j \sum_{n_j=0}^{\infty} \frac{\omega'}{2\pi} \int d\tilde{k}_{j||} \tilde{G}^{\text{ce},0}_{j,j;\sigma'',\sigma'''}(\tilde{k}_{j||}, n_j, \omega'), \tag{5.59}
$$

$$
\tilde{G}^{m,2b}_{i,i;\sigma,\sigma'}(\omega) = \frac{D_x}{2} \sum_{\sigma'',\sigma'''q,j} \tilde{G}^{m,0}_{i,i;\sigma,\sigma''}(\omega) \tilde{G}^{m,0}_{i,i;\sigma'',\sigma'}(\omega) \\
\times |v_{i,q,j}|^2 \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \tilde{G}^{\text{le},0}_{i,i;q,q;\sigma'',\sigma'''}(\omega') \tilde{G}^{\text{le},0}_{i,i;\sigma'',\sigma'''}(\omega') \\
\times eB_0 \alpha_j \sum_{n_j=0}^{\infty} \frac{\omega'}{2\pi} \int d\tilde{k}_{j||} \tilde{G}^{\text{ce},0}_{j,j;\sigma'',\sigma'''}(\tilde{k}_{j||}, n_j, \omega'), \tag{5.60}
$$

$$
\tilde{G}^{m,2c}_{i,i;\sigma,\sigma'}(\omega) = \frac{D_x}{2} \sum_{\sigma'',\sigma'''q,j} \tilde{G}^{m,0}_{i,i;\sigma,\sigma''}(\omega) \tilde{G}^{m,0}_{i,i;\sigma'',\sigma'}(\omega) \\
\times |v_{i,q,j}|^2 \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \tilde{G}^{\text{le},0}_{i,i;q,q;\sigma'',\sigma'''}(\omega') \tilde{G}^{\text{le},0}_{i,i;\sigma'',\sigma'''}(\omega') \\
\times eB_0 \alpha_j \sum_{n_j=0}^{\infty} \frac{\omega'}{2\pi} \int d\tilde{k}_{j||} \tilde{G}^{\text{ce},0}_{j,j;\sigma'',\sigma'''}(\tilde{k}_{j||}, n_j, \omega'), \tag{5.61}
$$

where each of these diagrams also contain the overall factor of -1 for a single fermion loop. However, unlike the diagrams shown in Figures 5.1-5.2, the corrections to the nuclear spin Green function for these three diagrams are non-vanishing self-energy corrections that arise explicitly from the conduction electrons, and hence are the leading contributions to the Knight shift in anisotropic and correlated metals.
The first of these gives rise to the actual Knight shift \( K = (\omega_{n,m} - \omega_n) / \omega_n \), or the change in the nuclear resonance frequency \( \omega_n \). The other two combine to give the leading contributions to the change in the resonant linewidth, \( L = (\Omega_{n,m} - \Omega_n) / \Omega_n \), due to the metal. The self-energies corresponding to these diagrams first-order in the hyperfine interactions are

\[
\hat{\Sigma}^{(2a)} = -\frac{(\omega_{n,m} - \omega_n)}{2} \sigma_3 \\
= \frac{D_z}{4N_b} \sigma_3 \sum_{q,j,\sigma''} |v_{i,q,j}|^2 \sigma'' I_{2a}^{le} (\sigma'', I_{0}^{ce,0}) \\
\approx \sigma_3 \frac{D_z K_{2a} \Omega_e^2}{2R_e^3 N_b} \sum_{q,j} |v_{i,q,j}|^2 \frac{e B_0 \alpha_j^2 (\omega_0 - \tilde{\omega}_{j,e})}{8 \pi \tilde{R}_{j,e}} \sum_{\tilde{\sigma}=\pm} \tilde{\sigma} N_{\tilde{\sigma},\tilde{\sigma}},
\]

\[
\hat{\Sigma}^{(2b)} = \frac{D_x}{2N_b} \sigma_1 \sum_{q,j,\sigma''} |v_{i,q,j}|^2 \sigma'' I_{2b}^{le} (\sigma'', I_{0}^{ce,0}) \\
\hat{\Sigma}^{(2c)} = \frac{D_x}{2N_b} \sigma_1 \sum_{q,j,\sigma''} |v_{i,q,j}|^2 \sigma'' I_{2c}^{le} (\sigma'', I_{0}^{ce,0}) \\
\hat{\Sigma}^{(2b)} + \hat{\Sigma}^{(2c)} = -\frac{(\Omega_{n,m} - \Omega_n)}{2} \sigma_1 \\
\approx \frac{D_x}{2N_b} \sigma_1 \sum_{q,j} |v_{i,q,j}|^2 \frac{K_{2b} (\omega_0 - \omega_e) \Omega_e (\omega_0 - \tilde{\omega}_{j,e})}{2 \pi R_e \tilde{R}_{j,e}^3} \\
- K_{2c} [R_e^2 + (\omega_0 - \omega_e)^2 / 4 |\tilde{\Omega}_{j,e}'|] \frac{e B_0 \alpha_j^2}{2 \pi} \sum_{\tilde{\sigma}=\pm} \tilde{\sigma} N_{\tilde{\sigma},\tilde{\sigma}} \tilde{\sigma},
\]

where

\[
N_{\tilde{\sigma},\tilde{\sigma}} = \sqrt{\frac{m_j}{2[\mu + \tilde{\sigma} \tilde{R}_{j,e}' - eB_0 \alpha_j (n_j + 1/2)/m_j]},}
\]

\[
n_{j,\text{max}} = \frac{m_j (\mu + \tilde{\sigma} \tilde{R}_{j,e}')}{eB_0 \alpha_j} - \frac{1}{2}.
\]

These effects are highly anisotropic, and are strongest for the magnetic field applied normal to
the layers of a layered metal, which could be modeled here as having at least one ellipsoidal, cigar-shaped Fermi surface. We note that in experiments to measure the Knight shift, \( \omega_0 \approx \omega_n \ll \omega_e, \tilde{\omega}'_{j,e} \), so that \( \omega_0 - \omega_e \approx -\omega_e \) and \( \omega_0 - \tilde{\omega}'_{j,e} \approx -\tilde{\omega}'_{j,e} \), which appears in Eq. (67) for the Knight shift. In Eq. (71) for the change in the resonant linewidth, the central factor in the large parentheses can be of either sign, depending upon the strength of the Coulomb interaction and upon the field direction and band index. This model incorporates \( N_b \) ellipsoidal metallic Fermi surfaces, leading to strong anisotropies in the Knight shift and in the resonant linewidth changes. The Landau orbits present in the anisotropic metal cause dramatic effects when one orbit crosses the Fermi surface.

We note that due to the differences in the densities of states for up and down spins, the leading contributions to the Knight shift in Eq. (67) (divided by \( \omega_n/2 \)) and to the linewidth change due to the presence of the metal in Eq. (71) (divided by \( \Omega_n/2 \)) for small \( B_1 \) and small \( B_0 \) are

\[
K \propto D_e e(B_1/B_0)^2 \sum_{q,j} |v_{i,q,j}|^2 \alpha_j^2 N'_j(0) \quad \text{and} \quad L \propto D_e e \sum_{q,j} |v_{i,q,j}|^2 [K_{2b} - K_{2c} \gamma_j/(2 \beta_j)] \alpha_j^2 N'_j(0),
\]

where \( N'_j(0) \) is the derivative of the conduction electron density of states for band \( j \), the \( \alpha_j, \beta_j, \) and \( \gamma_j \) are angular functions for band \( j \) given in the appendix, and \( K_{2b} \) and \( K_{2c} \) depend upon the strength of the local electron correlations. Therefore, the changes in the linewidth due to the anisotropic metal can be of either sign, and its anisotropy is influenced by local electron correlations. However, as \( B_0 \) increases, the Landau orbits can cross the spin-split Fermi energies \( E_{F,\sigma} = \mu + \sigma \tilde{R}'_{j,e} \), leading to oscillatory behavior in both formulas. Hence, the orbital and spin components of \( K \) and \( L \) are both contained in single formulas, and do not separate, as has been widely assumed.
CHAPTER 6: CONCLUSION

The model for the Knight shift $K$ and the corresponding change $L$ in the linewidth due to the nuclear species probed in a magnetic resonance experiment being in an anisotropic and correlated metal, as opposed to being in vacuum or in an insulator. The microscopic model couples the nuclear spin 1/2 spins to their local orbital electrons with a diagonal hyperfine interaction tensor of tetragonal symmetry. The local electrons that interact with the nucleus then are excited into the conduction bands, and while in the conduction bands, their orbital motion and spins both interact with the magnetic induction. The Landau levels of their orbital motion can give strong oscillations as a function of the strength of the magnetic induction. Although the model is easy to generalize $K$ and $L$ to finite temperature. This model can then be extended to include the superconducting order parameter of a variety of unconventional spin and orbital symmetries.

The most important inclusion in the new model of the Knight shift is the modified Anderson Hamiltonian term. In all previous applications of this model to local magnetic moments, the only part of the Anderson model that was included was the conduction electron part, whereas spin-orbit coupling was neglected, so that $\varepsilon_{j,\sigma}(k + eA)$ was that of a spherical Fermi surface. However, by flipping the nuclear spins with an applied magnetic field different parts of the Fermi surfaces are probed. A general Fermi surface of elliptical shape is therefore included in this model. Additionally, it is important to note that this model includes a Hamiltonian term that represents the energy of the local singly-occupied atomic orbital. In this way, the only energy level that contributes to the normal state Knight shift is that of energy $\epsilon_{i,q}$. For superconductors with low $T_c$ values such as Sr$_2$RuO$_4$, it is necessary for the singly-occupied atomic levels to lie at or very close to the Fermi level; otherwise they will lead to a normal state Knight shift, with a temperature independent Knight shift.
Of the Knight shift anomalies presented here, the lack of a temperature dependent Knight shift seen in Na$_{0.35}$CoO$_2$·1.3H$_2$O, can be explained by the new model. In this material, all of the Na sites are Na$^+$ ions, which have the atomic configuration of Ne, however they are more tightly bound than those of Ne due to the extra proton. This means that the highest energy orbitals are double filled and lie far below the Fermi energy level. Thus, the Knight shift of $^{23}$Na is zero even in the normal state.
APPENDIX A: THE HAMILTONIAN
A.1 The Model

The electron and nuclear Zeeman interaction is

$$\mathcal{H}_Z = -\gamma B_e \cdot \hat{y}_{B_e} \cdot S_i - \gamma B_N \cdot \hat{y}_{B_N} \cdot I_i$$  \hspace{1cm} (A.1)

where

$$H(t) = \hat{z}H_0 + \hat{x}H_1 \cos \omega t - \hat{y}H_1 \sin \omega t$$  \hspace{1cm} (A.2)

$$B_e = \mu_B H(t)$$  \hspace{1cm} (A.3)

$$B_N = \mu_N H(t)$$  \hspace{1cm} (A.4)

$$I = \frac{1}{2}$$  \hspace{1cm} (A.5)

$$S = \frac{1}{2}$$  \hspace{1cm} (A.6)
\[ H_Z = -\gamma B_e \cdot \hat{g}_{Bi} \cdot S_i - \gamma B_N \cdot \hat{g}_{Ni} \cdot I_i \]  

(A.7) 

\[ = -\gamma g_N (\mu_N H_0 I_z + \mu_N H_1 \cos \omega t I_x - \mu_N H_1 \sin \omega t I_y) \]  

(A.8) 

\[ - \gamma g_B (\mu_B H_0 S_z + \mu_B H_1 \cos \omega t S_x - \mu_B H_1 \sin \omega t S_y) \]  

\[ = -\gamma \left[ H_0 (g_N \mu_N I_z + g_B \mu_B S_z) \right. \]  

(A.9) 

\[ + H_1 \cos \omega t \left( \frac{g_N \mu_N}{2} (I_+ + I_-) + \frac{g_B \mu_B}{2} (S_+ + S_-) \right) \]  

\[ - H_1 \sin \omega t \left( \frac{g_N \mu_N}{2i} (I_+ - I_-) + \frac{g_B \mu_B}{2i} (S_+ - S_-) \right) \]  

\[ = -\gamma \left[ H_0 (g_N \mu_N I_z + g_B \mu_B S_z) \right. \]  

(A.10) 

\[ + I_+ \left( H_1 \cos \omega t \frac{g_N \mu_N}{2} - H_1 \sin \omega t \frac{g_B \mu_B}{2i} \right) + I_- \left( H_1 \cos \omega t \frac{g_N \mu_N}{2} + H_1 \sin \omega t \frac{g_B \mu_B}{2i} \right) \]  

\[ + S_+ \left( H_1 \cos \omega t \frac{g_B \mu_B}{2} - H_1 \sin \omega t \frac{g_B \mu_B}{2i} \right) + S_- \left( H_1 \cos \omega t \frac{g_B \mu_B}{2} + H_1 \sin \omega t \frac{g_B \mu_B}{2i} \right) \]  

\[ = -\gamma \left[ H_0 (g_N \mu_N I_z + g_B \mu_B S_z) \right. \]  

(A.11) 

\[ + \frac{g_N \mu_N}{2} H_1 I_+ \left( \cos \omega t + i \sin \omega t \right) + \frac{g_N \mu_N}{2} H_1 I_- \left( \cos \omega t - i \sin \omega t \right) \]  

\[ + \frac{g_B \mu_B}{2} H_1 S_+ \left( \cos \omega t + i \sin \omega t \right) + \frac{g_B \mu_B}{2} H_1 S_- \left( \cos \omega t - i \sin \omega t \right) \]  

\[ = -\gamma \left[ H_0 (g_N \mu_N I_z + g_B \mu_B S_z) \right. \]  

(A.12) 

\[ + \frac{g_N \mu_N}{2} H_1 I_+ \left( e^{i\omega t} \right) + \frac{g_N \mu_N}{2} H_1 I_- \left( e^{-i\omega t} \right) \]  

\[ + \frac{g_B \mu_B}{2} H_1 S_+ \left( e^{i\omega t} \right) + \frac{g_B \mu_B}{2} H_1 S_- \left( e^{-i\omega t} \right) \]  

\[ = -\gamma \left[ H_0 (g_N \mu_N I_z + g_B \mu_B S_z) \right. \]  

(A.13) 

\[ + \frac{1}{2} g_B \mu_B H_1 \left( e^{i\omega t} S_+ + e^{-i\omega t} S_- \right) + \frac{1}{2} g_N \mu_N H_1 \left( e^{i\omega t} I_+ + e^{-i\omega t} I_- \right) \]  

67
Now let,

\[
\begin{align*}
\omega_B &= \gamma g_B \mu_B H_0 \\
\omega_N &= \gamma g_N \mu_N H_0 \\
\Omega_B &= \frac{1}{2} \gamma g_B \mu_B H_1 \\
\Omega_N &= \frac{1}{2} \gamma g_N \mu_N H_1
\end{align*}
\] (A.14)

Then

\[
\mathcal{H}_Z = -\omega_B S_z - \omega_N I_z - \Omega_B \left( e^{i\omega t} S_+ + e^{-i\omega t} S_- \right) - \Omega_N \left( e^{i\omega t} I_+ + e^{-i\omega t} I_- \right)
\] (A.16)

The hyperfine interaction is

\[
\mathcal{H}_{hf} = \mathbf{I} \cdot \mathbf{\hat{D}} \cdot \mathbf{S} = \mathbf{\hat{D}} \mathbf{S} \cdot \mathbf{I}
\] (A.17)

\[
= \mathbf{\hat{D}} I_z S_z + \frac{\mathbf{\hat{D}}}{2} I_- S_+ + \frac{\mathbf{\hat{D}}}{2} I_+ S_-
\] (A.18)

And finally, the complete Hamiltonian is

\[
\mathcal{H} = -\omega_B S_z - \omega_N I_z
\]

\[
- \Omega_B \left( e^{i\omega t} S_+ + e^{-i\omega t} S_- \right) - \Omega_N \left( e^{i\omega t} I_+ + e^{-i\omega t} I_- \right)
\]

\[
+ \mathbf{\hat{D}} I_z S_z + \frac{\mathbf{\hat{D}}}{2} I_- S_+ + \frac{\mathbf{\hat{D}}}{2} I_+ S_-
\] (A.19)
A.2 Quartic Equation for $\gamma_i$

To obtain the matrix of equation, I operate with the Hamiltonian on each state as follows.

$$\mathcal{H} \left| \frac{1}{2}, -\frac{1}{2} \right\rangle =
\begin{pmatrix}
\left(-\omega_B \frac{1}{2} + \omega_N \frac{1}{2}\right) & \left| \frac{1}{2}, -\frac{1}{2} \right\rangle - \Omega_B e^{i\omega t} & -\frac{1}{2}, \frac{1}{2} \right\rangle - \Omega_N e^{-i\omega t} \left| \frac{1}{2}, \frac{1}{2} \right\rangle \\
-\frac{\tilde{D}}{4} & \left| 1, \frac{1}{2} \right\rangle + \frac{\tilde{D}}{2} & \left| \frac{1}{2}, \frac{1}{2} \right\rangle 
\end{pmatrix}
$$

(A.20)

$$\mathcal{H} \left| -\frac{1}{2}, \frac{1}{2} \right\rangle =
\begin{pmatrix}
\left(\omega_B \frac{1}{2} - \omega_N \frac{1}{2}\right) & \left| -1, \frac{1}{2} \right\rangle - \Omega_B e^{-i\omega t} & \frac{1}{2}, \frac{1}{2} \right\rangle - \Omega_N e^{i\omega t} \left| -\frac{1}{2}, -\frac{1}{2} \right\rangle \\
-\frac{\tilde{D}}{4} & \left| -1, \frac{1}{2} \right\rangle + \frac{\tilde{D}}{2} & \left| 1, -\frac{1}{2} \right\rangle 
\end{pmatrix}
$$

(A.21)

$$\sqrt{2} \mathcal{H} \left| 00 \right\rangle = \mathcal{H} \left| \frac{1}{2}, -\frac{1}{2} \right\rangle - \mathcal{H} \left| -\frac{1}{2}, \frac{1}{2} \right\rangle$$

(A.22)

$$= \begin{pmatrix}
\left(-\omega_B \frac{1}{2} + \omega_N \frac{1}{2}\right) & \left| \frac{1}{2}, -\frac{1}{2} \right\rangle - \Omega_B e^{i\omega t} & -\frac{1}{2}, \frac{1}{2} \right\rangle - \Omega_N e^{-i\omega t} \left| \frac{1}{2}, \frac{1}{2} \right\rangle \\
-\frac{\tilde{D}}{4} & \left| 1, \frac{1}{2} \right\rangle + \frac{\tilde{D}}{2} & \left| \frac{1}{2}, \frac{1}{2} \right\rangle 
\end{pmatrix}
$$

(A.23)

$$= \begin{pmatrix}
\left(\omega_N - \omega_B\right) & \left| \frac{1}{2}, -\frac{1}{2} \right\rangle - \Omega_B e^{i\omega t} & -\frac{1}{2}, \frac{1}{2} \right\rangle - \Omega_N e^{-i\omega t} \left| \frac{1}{2}, \frac{1}{2} \right\rangle \\
\frac{\tilde{D}}{4} & \left| 1, \frac{1}{2} \right\rangle + \frac{\tilde{D}}{2} & \left| \frac{1}{2}, \frac{1}{2} \right\rangle 
\end{pmatrix}
$$

(A.24)
\[ H |00\rangle = \frac{1}{2} (\omega_N - \omega_B) \frac{1}{\sqrt{2}} \left( \left| \frac{1}{2}, -\frac{1}{2} \right\rangle + \left| -\frac{1}{2}, \frac{1}{2} \right\rangle \right) - \frac{3\tilde{D}}{4} \frac{1}{\sqrt{2}} \left( \left| \frac{1}{2}, -\frac{1}{2} \right\rangle - \left| \frac{1}{2}, \frac{1}{2} \right\rangle \right) \\
+ \frac{1}{\sqrt{2}} (\Omega_N - \Omega_B) e^{i\omega t} \left| -\frac{1}{2}, -\frac{1}{2} \right\rangle + \frac{1}{\sqrt{2}} (\Omega_B - \Omega_N) e^{-i\omega t} \left| \frac{1}{2}, \frac{1}{2} \right\rangle \]

(A.25)

This gives the first equation for the state a.

\[ H a = \frac{1}{2} (\omega_N - \omega_B) b - \frac{3\tilde{D}}{4} a + \frac{1}{\sqrt{2}} (\Omega_N - \Omega_B) e^{i\omega t} c + \frac{1}{\sqrt{2}} (\Omega_B - \Omega_N) e^{-i\omega t} d \]  

(A.26)

\[ \sqrt{2} H |01\rangle = \sqrt{2} H \left| \frac{1}{2}, -\frac{1}{2} \right\rangle + H \left| -\frac{1}{2}, \frac{1}{2} \right\rangle \]

(A.27)

\[ = \left( -\frac{\Omega_B}{2} + \frac{\omega_N}{2} - \tilde{D} \frac{4}{2} + \tilde{D} \frac{2}{2} \right) \left| \frac{1}{2}, -\frac{1}{2} \right\rangle + \left( \frac{\omega_B}{2} - \frac{\omega_N}{2} - \tilde{D} \frac{4}{2} + \tilde{D} \frac{2}{2} \right) \left| -\frac{1}{2}, \frac{1}{2} \right\rangle \\
+ (-\Omega_B e^{i\omega t} - \Omega_N e^{i\omega t}) \left| -\frac{1}{2}, -\frac{1}{2} \right\rangle + (-\Omega_N e^{-i\omega t} - \Omega_B e^{-i\omega t}) \left| \frac{1}{2}, \frac{1}{2} \right\rangle \]

(A.28)

\[ H |01\rangle = \frac{1}{2} (\omega_N - \omega_B) \frac{1}{\sqrt{2}} \left( \left| \frac{1}{2}, -\frac{1}{2} \right\rangle - \left| -\frac{1}{2}, \frac{1}{2} \right\rangle \right) + \frac{3\tilde{D}}{4} \frac{1}{\sqrt{2}} \left( \left| \frac{1}{2}, -\frac{1}{2} \right\rangle + \left| -\frac{1}{2}, \frac{1}{2} \right\rangle \right) \\
- \frac{1}{\sqrt{2}} (\Omega_N + \Omega_B) e^{i\omega t} \left| -\frac{1}{2}, -\frac{1}{2} \right\rangle - \frac{1}{\sqrt{2}} (\Omega_B + \Omega_N) e^{-i\omega t} \left| \frac{1}{2}, \frac{1}{2} \right\rangle \]

(A.29)

This gives the second equation for the state b.

\[ H b = \frac{1}{2} (\omega_N - \omega_B) a + \frac{3\tilde{D}}{4} b - \frac{1}{\sqrt{2}} (\Omega_N + \Omega_B) e^{i\omega t} c - \frac{1}{\sqrt{2}} (\Omega_B + \Omega_N) e^{-i\omega t} d \]  

(A.30)
\( \mathcal{H} c = \mathcal{H} |1, -1\rangle = \mathcal{H} \left| \frac{1}{2}, -\frac{1}{2}\right\rangle \)  
\( = \frac{1}{2}(\omega_B + \omega_N) \left| \frac{1}{2}, -\frac{1}{2}\right\rangle - \Omega_B e^{-i\omega t} \left| \frac{1}{2}, -\frac{1}{2}\right\rangle - \Omega_N e^{-i\omega t} \left| -\frac{1}{2}, \frac{1}{2}\right\rangle + \frac{\tilde{D}}{4} \left| -\frac{1}{2}, -\frac{1}{2}\right\rangle \)  
\( \tag{A.31} \)

\( = \frac{1}{2}(\omega_B + \omega_N) c - \Omega_B e^{-i\omega t} \frac{1}{\sqrt{2}}(a + b) - \Omega_N e^{-i\omega t} \frac{1}{\sqrt{2}}(b - a) + \frac{\tilde{D}}{4} c \)  
\( \tag{A.32} \)

This gives the third equation for the state c.

\( \mathcal{H} c = \frac{1}{\sqrt{2}}(\Omega_N - \Omega_B) e^{-i\omega t} a - \frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) e^{-i\omega t} b + \left( \frac{\tilde{D}}{4} + \frac{1}{2}(\omega_N + \omega_B) \right) c \)  
\( \tag{A.33} \)

\( \mathcal{H} d = \mathcal{H} |1, 1\rangle = \mathcal{H} \left| \frac{1}{2}, \frac{1}{2} \right\rangle \)  
\( = -\frac{1}{2}(\omega_B + \omega_N) \left| \frac{1}{2}, \frac{1}{2} \right\rangle - \Omega_B e^{i\omega t} \left| -\frac{1}{2}, \frac{1}{2} \right\rangle - \Omega_N e^{i\omega t} \left| \frac{1}{2}, -\frac{1}{2} \right\rangle + \frac{\tilde{D}}{4} \left| \frac{1}{2}, \frac{1}{2} \right\rangle \)  
\( \tag{A.35} \)

\( = -\frac{1}{2}(\omega_B + \omega_N) d - \Omega_B e^{i\omega t} \frac{1}{\sqrt{2}}(b - a) - \Omega_N e^{i\omega t} \frac{1}{\sqrt{2}}(a + b) + \frac{\tilde{D}}{4} d \)  
\( \tag{A.36} \)

This gives the second equation for the state b.

\( \mathcal{H} d = \frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) e^{i\omega t} a - \frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) e^{i\omega t} b + \left( \frac{\tilde{D}}{4} - \frac{1}{2}(\omega_N + \omega_B) \right) d \)  
\( \tag{A.37} \)

And finally, we have a system of four equations.

\[
\mathcal{H} \begin{pmatrix} a \\ b \\ c \\ d \end{pmatrix} = \begin{pmatrix} -\frac{\tilde{D}}{4} & \frac{1}{2}(\omega_N - \omega_B) & \frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) e^{i\omega t} & \frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) e^{-i\omega t} \\ \frac{1}{2}(\omega_N - \omega_B) & \frac{\tilde{D}}{4} & \frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) e^{i\omega t} & \frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) e^{-i\omega t} \\ \frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) e^{-i\omega t} & \frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) e^{-i\omega t} & \frac{\tilde{D}}{4} + \frac{1}{2}(\omega_N + \omega_B) & 0 \\ \frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) e^{i\omega t} & \frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) e^{i\omega t} & 0 & \frac{\tilde{D}}{4} - \frac{1}{2}(\omega_N + \omega_B) \end{pmatrix} \begin{pmatrix} a \\ b \\ c \\ d \end{pmatrix} \]
\( \tag{A.39} \)
We begin to solve the system using the Schrödinger equation.

\[
\begin{align*}
\hbar i \frac{d\Psi}{dt} &= \mathcal{H}\,|\Psi\rangle \quad \text{(A.40)}
\end{align*}
\]

\[
\begin{pmatrix}
\dot{a} \\
\dot{b} \\
\dot{c} \\
\dot{d}
\end{pmatrix} =
\begin{pmatrix}
\frac{-3\tilde{D}}{4} & \frac{1}{2}(\omega_N - \omega_B) & -\frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) e^{i\omega t} & -\frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) e^{-i\omega t} \\
\frac{1}{2}(\omega_N - \omega_B) & \frac{\tilde{D}}{4} & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) e^{i\omega t} & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) e^{-i\omega t} \\
\frac{1}{\sqrt{2}}(\Omega_N - \Omega_B) e^{-i\omega t} & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) e^{-i\omega t} & \frac{\tilde{D}}{4} + \frac{1}{2}(\omega_N + \omega_B) & 0 \\
\frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) e^{i\omega t} & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) e^{i\omega t} & 0 & \frac{\tilde{D}}{4} - \frac{1}{2}(\omega_N + \omega_B)
\end{pmatrix}
\begin{pmatrix}
a \\
b \\
c \\
d
\end{pmatrix} \quad \text{(A.41)}
\]

Now let

\[
\begin{align*}
a &= Ae^{-\frac{i\tilde{D}t}{4}} & \dot{a} &= \dot{A}e^{-\frac{i\tilde{D}t}{4}} - A\frac{i\tilde{D}}{4} e^{-\frac{i\tilde{D}t}{4}} \quad \text{(A.42)} \\
b &= Be^{-\frac{i\tilde{D}t}{4}} & \dot{b} &= \dot{B}e^{-\frac{i\tilde{D}t}{4}} - B\frac{i\tilde{D}}{4} e^{-\frac{i\tilde{D}t}{4}} \quad \text{(A.43)} \\
c &= Ce^{-i\omega t - \frac{i\tilde{D}t}{4}} & \dot{c} &= \dot{C}e^{-i\omega t - \frac{i\tilde{D}t}{4}} + C(-i\omega - \frac{i\tilde{D}}{4}) e^{-i\omega t - \frac{i\tilde{D}t}{4}} \quad \text{(A.44)} \\
d &= De^{i\omega t - \frac{i\tilde{D}t}{4}} & \dot{d} &= \dot{D}e^{i\omega t - \frac{i\tilde{D}t}{4}} + D(i\omega - \frac{i\tilde{D}}{4}) e^{i\omega t - \frac{i\tilde{D}t}{4}} \quad \text{(A.45)}
\end{align*}
\]
Now I get

\[
e^{-i\frac{\theta}{4}} \begin{pmatrix} i\dot{A} + A\frac{\dot{D}}{4} \\ i\dot{B} + B\frac{\dot{D}}{4} \\ (i\dot{C} + C\omega + C\frac{\dot{D}}{4})e^{-i\omega t} \\ (i\dot{D} - D\omega + D\frac{\dot{D}}{4})e^{i\omega t} \end{pmatrix} = \\
\begin{pmatrix} -\frac{3\dot{D}}{4} & \frac{1}{2}(\omega_N - \omega_B) & -\frac{1}{\sqrt{2}}(\Omega_B - \Omega_N)e^{i\omega t} & -\frac{1}{\sqrt{2}}(\Omega_B - \Omega_N)e^{-i\omega t} \\ \frac{1}{2}(\omega_N - \omega_B) & \frac{\dot{D}}{4} & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B)e^{i\omega t} & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B)e^{-i\omega t} \\ \frac{1}{\sqrt{2}}(\Omega_N - \Omega_B)e^{i\omega t} & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B)e^{i\omega t} & \frac{\dot{D}}{4} + \frac{1}{2}(\omega_N + \omega_B) & 0 \\ \frac{1}{\sqrt{2}}(\Omega_N - \Omega_B)e^{i\omega t} & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B)e^{i\omega t} & 0 & \frac{\dot{D}}{4} - \frac{1}{2}(\omega_N + \omega_B) \end{pmatrix} \begin{pmatrix} A \\ B \\ Ce^{-i\omega t} \\ De^{i\omega t} \end{pmatrix} \]

This simplifies to

\[
i \begin{pmatrix} \dot{A} \\ \dot{B} \\ \dot{C} \\ \dot{D} \end{pmatrix} = \begin{pmatrix} -\tilde{D} & \frac{1}{2}(\omega_N - \omega_B) & -\frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) & \frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) \\ \frac{1}{2}(\omega_N - \omega_B) & 0 & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) \\ \frac{1}{\sqrt{2}}(\Omega_N - \Omega_B) & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) & -\omega + \frac{1}{2}(\omega_N + \omega_B) & 0 \\ \frac{1}{\sqrt{2}}(\Omega_B - \Omega_N) & -\frac{1}{\sqrt{2}}(\Omega_N + \Omega_B) & 0 & \omega - \frac{1}{2}(\omega_N + \omega_B) \end{pmatrix} \begin{pmatrix} A \\ B \\ C \\ D \end{pmatrix}
\]

(A.47)

Now let

\[
\Gamma = \frac{D - C}{\sqrt{2}} \quad \Delta = \frac{D + C}{\sqrt{2}}
\]

(A.48)
Which gives

\[
\begin{pmatrix}
\dot{A} \\
\dot{B} \\
\dot{\Gamma} \\
\dot{\Delta}
\end{pmatrix} =
\begin{pmatrix}
-\ddot{D} & \frac{1}{2}(\omega_N - \omega_B) & (\Omega_B - \Omega_N) & 0 \\
\frac{1}{2}(\omega_N - \omega_B) & 0 & 0 & -(\Omega_N + \Omega_B) \\
-(\Omega_N - \Omega_B) & 0 & 0 & \omega - \frac{1}{2}(\omega_N + \omega_B) \\
0 & -(\Omega_N + \Omega_B) & \omega - \frac{1}{2}(\omega_N + \omega_B) & 0
\end{pmatrix}
\begin{pmatrix}
A \\
B \\
\Gamma \\
\Delta
\end{pmatrix}
\]  

(A.49)

Simplying the algebra even further, I let

\[
\begin{align*}
\alpha &= \frac{1}{2}(\omega_N - \omega_B) \\
\beta &= \omega - \frac{1}{2}(\omega_N + \omega_B) \\
\delta &= \Omega_N + \Omega_B \\
\epsilon &= \Omega_B - \Omega_N
\end{align*}
\]

(A.50) (A.51) (A.52) (A.53)

Which gives

\[
\begin{pmatrix}
\dot{A} \\
\dot{B} \\
\dot{\Gamma} \\
\dot{\Delta}
\end{pmatrix} =
\begin{pmatrix}
-\ddot{D} & \alpha & \epsilon & 0 \\
\alpha & 0 & 0 & -\delta \\
\epsilon & 0 & 0 & \beta \\
0 & -\delta & \beta & 0
\end{pmatrix}
\begin{pmatrix}
A \\
B \\
\Gamma \\
\Delta
\end{pmatrix}
\]  

(A.54)
Then, since the equations do not depend explicitly upon the time, I make the following substitutions

\[ A = A_0 e^{i\gamma t} \quad (A.55) \]
\[ B = B_0 e^{i\gamma t} \quad (A.56) \]
\[ \Gamma = \Gamma_0 e^{i\gamma t} \quad (A.57) \]
\[ \Delta = \Delta_0 e^{i\gamma t} \quad (A.58) \]

Which gives

\[
\begin{pmatrix}
-\gamma_0 A_0 \\
-\gamma_0 B_0 \\
-\gamma_0 \Gamma_0 \\
-\gamma_0 \Delta_0
\end{pmatrix} e^{i\gamma t} =
\begin{pmatrix}
-\tilde{D} & \alpha & \epsilon & 0 \\
\alpha & 0 & 0 & -\delta \\
\epsilon & 0 & 0 & \beta \\
0 & -\delta & \beta & 0
\end{pmatrix}
\begin{pmatrix}
A_0 \\
B_0 \\
\Gamma_0 \\
\Delta_0
\end{pmatrix} e^{i\gamma t}
\] (A.59)

And so I get the following system of algebraic equations,

\[-\gamma_0 A_0 = -\tilde{D} A_0 + \alpha B_0 + \epsilon \Gamma_0 \quad (A.60)\]
\[-\gamma_0 B_0 = \alpha A_0 - \delta \Delta_0 \quad (A.61)\]
\[-\gamma_0 \Gamma_0 = \epsilon A_0 + \beta \Delta_0 \quad (A.62)\]
\[-\gamma_0 \Delta_0 = -\delta B_0 + \beta \Gamma_0 \quad (A.63)\]
I now solve this system of equations to obtain a quartic equation for $\gamma_i$.

\[
\begin{align*}
\Delta_0 &= \frac{\alpha A_0 + \gamma_i B_0}{\delta} \quad \text{(A.64)} \\
-\gamma_i \Gamma_0 &= \epsilon A_0 + \beta \frac{\alpha A_0 + \gamma_i B_0}{\delta} \quad \text{(A.65)} \\
\Gamma_0 &= (\epsilon - \frac{\alpha \beta}{\gamma_i \delta}) A_0 - \frac{\beta}{\delta} B_0 \quad \text{(A.66)} \\
-\gamma_i \frac{\alpha A_0 + \gamma_i B_0}{\delta} &= -\delta B_0 + \beta \left[ (\epsilon - \frac{\alpha \beta}{\gamma_i \delta}) A_0 - \frac{\beta}{\delta} B_0 \right] \quad \text{(A.67)} \\
\left[ -\frac{\gamma_i \alpha}{\delta} + \beta \left( \frac{\epsilon}{\gamma_i} + \frac{\alpha \beta}{\gamma_i \delta} \right) \right] A_0 &= \left[ \frac{\gamma_i^2}{\delta} - \delta^2 - \frac{\beta^2}{\delta} \right] B_0 \quad \text{(A.68)} \\
\left[ -\gamma_i \alpha + \beta \left( \frac{\delta \epsilon}{\gamma_i} + \frac{\alpha \beta}{\gamma_i} \right) \right] A_0 &= \left[ \gamma_i^2 - \delta^2 - \beta^2 \right] B_0 \quad \text{(A.69)} \\
\left[ -\gamma_i^2 \alpha + \beta \delta \epsilon + \alpha \beta^2 \right] A_0 &= \left[ \gamma_i^3 - \gamma_i \delta^2 - \gamma_i \beta^2 \right] B_0 \quad \text{(A.70)} \\
-\gamma_i A_0 &= -\tilde{D} A_0 + \alpha B_0 + \epsilon \left[ (\epsilon - \frac{\alpha \beta}{\gamma_i \delta}) A_0 - \frac{\beta}{\delta} B_0 \right] \\
\left[ -\gamma_i + \tilde{D} + \frac{\epsilon^2}{\gamma_i} + \frac{\alpha \beta \epsilon}{\gamma_i \delta} \right] A_0 &= \left[ \alpha - \frac{\beta \epsilon}{\delta} \right] B_0 \quad \text{(A.72)} \\
\left[ -\gamma_i^2 + \gamma_i \tilde{D} + \epsilon^2 + \frac{\alpha \beta \epsilon}{\delta} \right] A_0 &= \left[ \alpha \gamma_i - \frac{\gamma_i \beta \epsilon}{\delta} \right] B_0 \quad \text{(A.73)} \\
\left[ -\gamma_i^2 \delta + \gamma_i \delta \tilde{D} + \delta \epsilon^2 + \alpha \beta \epsilon \right] A_0 &= \left[ \delta \alpha \gamma_i - \gamma_i \beta \epsilon \right] B_0 \quad \text{(A.74)}
\end{align*}
\]
Now these two equations give

\[
\begin{align*}
-\gamma^2 \alpha + \beta \delta \epsilon + \alpha \beta^2]A_0 &= [\gamma^3 - \gamma_i \delta^2 - \gamma_i \beta^2]B_0 \\
-\gamma^2 \alpha + \beta \delta \epsilon + \alpha \beta^2][\delta \alpha \gamma_i - \gamma_i \beta \epsilon]B_0 &= (A.75)
\end{align*}
\]

\[
\begin{align*}
\gamma^3 - \gamma_i \delta^2 - \gamma_i \beta^2][-\gamma^2 \delta + \gamma_i \delta D + \delta \epsilon^2 + \alpha \beta \epsilon] = \\
-\gamma^2 \alpha + \beta \delta \epsilon + \alpha \beta^2][\delta \alpha \gamma_i - \gamma_i \beta \epsilon] = (A.76)
\end{align*}
\]

\[
\begin{align*}
-\gamma^2 \delta^2 + \gamma_i \delta D + \delta \epsilon^2 + \alpha \beta \epsilon] = \\
\gamma^3 - \gamma_i \delta^2 - \gamma_i \beta^2][-\gamma^2 \delta + \gamma_i \delta D + \delta \epsilon^2 + \alpha \beta \epsilon] = (A.77)
\end{align*}
\]

\[
\begin{align*}
-\gamma^2 \alpha + \beta \delta \epsilon + \alpha \beta^2][\delta \alpha - \beta \epsilon] = \\
\gamma^3 - \gamma_i \delta^2 - \gamma_i \beta^2][-\gamma^2 \delta + \gamma_i \delta D + \delta \epsilon^2 + \alpha \beta \epsilon] = (A.78)
\end{align*}
\]

\[
\begin{align*}
-\gamma^2 \delta^2 + \gamma_i \alpha \beta \epsilon + \beta \delta^2 \epsilon \alpha \\
-\beta^2 \delta \epsilon^2 + \alpha^2 \beta^2 \delta - \alpha \beta^3 \epsilon
\end{align*}
\]

\[
\begin{align*}
-\gamma^2 \delta^2 + \gamma_i \alpha \beta \epsilon + \beta \delta^2 \epsilon \alpha \\
-\beta^2 \delta \epsilon^2 + \alpha^2 \beta^2 \delta - \alpha \beta^3 \epsilon
\end{align*}
\]

\[
\begin{align*}
+\alpha^2 \beta^3 \epsilon - [\gamma^2 - \delta^2 - \beta^2] \alpha \beta \epsilon = \\
[\gamma^2 - \delta^2 - \beta^2](\gamma \alpha \epsilon) = (A.79)
\end{align*}
\]

\[
\begin{align*}
-\gamma^2 \delta^2 + \gamma_i \alpha \beta \epsilon + \beta \delta^2 \epsilon \alpha \\
-\beta^2 \delta \epsilon^2 + \alpha^2 \beta^2 \delta - \alpha \beta^3 \epsilon
\end{align*}
\]

\[
\begin{align*}
+\alpha^2 \beta^3 \epsilon - [\gamma^2 - \delta^2 - \beta^2] \alpha \beta \epsilon = \\
[\gamma^2 - \delta^2 - \beta^2](\gamma \alpha \epsilon) = (A.80)
\end{align*}
\]

\[
\begin{align*}
-\gamma^2 \delta^2 + \gamma_i \alpha \beta \epsilon + \beta \delta^2 \epsilon \alpha \\
-\beta^2 \delta \epsilon^2 + \alpha^2 \beta^2 \delta - \alpha \beta^3 \epsilon
\end{align*}
\]

\[
\begin{align*}
+\delta^2 \alpha \beta \epsilon + \alpha \beta^3 \epsilon
\end{align*}
\]

\[
\begin{align*}
[\gamma^2 - \delta^2 - \beta^2](\gamma \alpha \epsilon) = (A.81)
\end{align*}
\]

\[
\begin{align*}
-\gamma^2 \delta^2 + \beta^2 \epsilon^2 + \alpha^2 \beta^2 \delta + 2 \delta^2 \alpha \beta \epsilon = [\gamma^2 - \delta^2 - \beta^2](\gamma \alpha \epsilon) = (A.82)
\end{align*}
\]
\[
\gamma_i^2 \alpha^2 + \beta^2 \epsilon^2 - \alpha^2 \beta^2 - 2 \delta \alpha \beta \epsilon = \left[ \gamma_i^2 - \delta^2 - \beta^2 \right]\left[ \gamma_i^2 - \gamma_i \tilde{D} - \epsilon^2 \right]
\] (A.83)

\[
\gamma_i^2 \alpha^2 - \alpha^2 \beta^2 - \alpha^2 \delta^2 
+ \alpha^2 \delta^2 + \beta^2 \epsilon^2 - 2 \delta \alpha \beta \epsilon = \left[ \gamma_i^2 - \delta^2 - \beta^2 \right]\left[ \gamma_i^2 - \gamma_i \tilde{D} - \epsilon^2 \right]
\] (A.84)

\[
\alpha^2 \left[ \gamma_i^2 - \beta^2 - \delta^2 \right] + (\alpha \delta - \beta \epsilon)^2 = \left[ \gamma_i^2 - \delta^2 - \beta^2 \right]\left[ \gamma_i^2 - \gamma_i \tilde{D} - \epsilon^2 \right]
\] (A.85)

\[
0 = \left[ \gamma_i^2 - \delta^2 - \beta^2 \right]\left[ \gamma_i^2 - \gamma_i \tilde{D} - \epsilon^2 - \alpha^2 \right] - (\alpha \delta - \beta \epsilon)^2
\] (A.86)

And substituting back in

\[
0 = \left[ \gamma_i^2 - (\Omega_N + \Omega_B)^2 - (\omega - \frac{1}{2}(\omega_N + \omega_B))^2 \right]\left[ \gamma_i^2 - \gamma_i \tilde{D} - (\Omega_B - \Omega_N)^2 - \frac{1}{2}(\omega_N - \omega_B)^2 \right]
- \left[ \frac{1}{2}(\omega_N - \omega_B) \right]\left( \omega_N + \Omega_B \right) - \left( \omega - \frac{1}{2}(\omega_N + \omega_B) \right) (\Omega_B - \Omega_N)^2
\] (A.87)
APPENDIX B: COEFFICIENT EQUATIONS
B.1 Set Up of Equations

To begin, I am using the a, b, c, and d notation of spin states, where

\[ a = \frac{e - f}{\sqrt{2}} \]  
\[ b = \frac{e + f}{\sqrt{2}} \]  

Later I use the following transformation

\[ a = Ae^{-i\beta t} \]  
\[ b = Be^{-i\beta t} \]  
\[ c = Ce^{i\omega t - i\beta t} \]  
\[ d = De^{-i\omega t - i\beta t} \]  

And

\[ \Gamma = \frac{D - C}{\sqrt{2}} \]  
\[ \Delta = \frac{D + C}{\sqrt{2}} \]  

(B.1)  
(B.2)  
(B.3)  
(B.4)  
(B.5)  
(B.6)  
(B.7)
Now, in order to find the coefficients $a_1, a_2, a_3, a_4$ and similarly for $b, c, d$, I start with the differential equations for $A_1, A_2, A_3, A_4$ and so on. The equations for $a(t)$ etc. are

\[ a(t) = e^{-i\Delta t} (a_1 e^{i\gamma_1 t} + a_2 e^{i\gamma_2 t} + a_3 e^{i\gamma_3 t} + a_4 e^{i\gamma_4 t}) = e^{-i\Delta t} \sum_{j=1}^{4} a_j e^{i\alpha_j t} = \sum_{j=1}^{4} a_j e^{i\alpha_j t} \quad (B.8) \]

\[ b(t) = e^{-i\Delta t} (b_1 e^{i\gamma_1 t} + b_2 e^{i\gamma_2 t} + b_3 e^{i\gamma_3 t} + b_4 e^{i\gamma_4 t}) = e^{-i\Delta t} \sum_{j=1}^{4} b_j e^{i\beta_j t} = \sum_{j=1}^{4} b_j e^{i\beta_j t} \quad (B.9) \]

\[ c(t) = e^{i\omega t - i\Delta t} (c_1 e^{i\gamma_1 t} + c_2 e^{i\gamma_2 t} + c_3 e^{i\gamma_3 t} + c_4 e^{i\gamma_4 t}) = e^{i\omega t - i\Delta t} \sum_{j=1}^{4} c_j e^{i\gamma_j t} = \sum_{j=1}^{4} c_j e^{i\gamma_j t} \quad (B.10) \]

\[ d(t) = e^{-i\omega t - i\Delta t} (d_1 e^{i\gamma_1 t} + d_2 e^{i\gamma_2 t} + d_3 e^{i\gamma_3 t} + d_4 e^{i\gamma_4 t}) = e^{-i\omega t - i\Delta t} \sum_{j=1}^{4} d_j e^{i\gamma_j t} = \sum_{j=1}^{4} d_j e^{i\gamma_j t} \quad (B.11) \]

To simplify notation, let $\sum_{j=1}^{4} = \sum$

\[ a(t) = \sum a_j e^{i\alpha_j t} \quad (B.12) \]

\[ b(t) = \sum b_j e^{i\beta_j t} \quad (B.13) \]

\[ c(t) = \sum c_j e^{i\gamma_j t} \quad (B.14) \]

\[ d(t) = \sum d_j e^{i\delta_j t} \quad (B.15) \]

I can now obtain the following higher derivative equations.

\[ \dot{a}(t) = i \sum \tilde{a}_j a_j e^{i\alpha_j t} \quad \ddot{a}(t) = -i \sum \tilde{a}_j^2 a_j e^{i\alpha_j t} \quad 
\dot{b}(t) = i \sum \tilde{b}_j b_j e^{i\beta_j t} \quad \ddot{b}(t) = -i \sum \tilde{b}_j^2 b_j e^{i\beta_j t} \quad 
\dot{c}(t) = i \sum \tilde{c}_j c_j e^{i\gamma_j t} \quad \ddot{c}(t) = -i \sum \tilde{c}_j^2 c_j e^{i\gamma_j t} \quad 
\dot{d}(t) = i \sum \tilde{d}_j d_j e^{i\delta_j t} \quad \ddot{d}(t) = -i \sum \tilde{d}_j^2 d_j e^{i\delta_j t} \quad \]
This gives me sixteen differential equations with are to be solved using the set of equations from the previous section. These are

\[
\begin{align*}
    i\dot{A}(t) &= -\tilde{D}A(t) + \alpha B(t) + \epsilon \Gamma(t) \\
    i\dot{B}(t) &= \alpha A(t) - \delta \Delta(t) \\
    i\dot{\Gamma}(t) &= \epsilon A(t) + \beta \Delta(t) \\
    i\dot{\Delta}(t) &= -\delta B(t) + \beta \Gamma(t)
\end{align*}
\] (B.20)

B.2 Initial State \(|\frac{1}{2}, \frac{1}{2}\rangle\)

First we need to pick an initial condition, so let’s assume we are in the \(|\uparrow\uparrow\rangle\) state. This means \(a(0) = b(0) = c(0) = 0\) and \(d(0) = 1\).

\[
\begin{align*}
    a(0) &= \sum a_j = 0 & \dot{a}(0) &= i \sum \tilde{a}_j a_j & \ddot{a}(0) &= -\sum \tilde{a}_j^2 a_j & \dot{\dddot{a}}(0) &= -i \sum \tilde{a}_j^3 a_j \\
    b(0) &= \sum b_j = 0 & \dot{b}(0) &= i \sum \tilde{b}_j b_j & \ddot{b}(0) &= -\sum \tilde{b}_j^2 b_j & \dot{\dddot{b}}(0) &= -i \sum \tilde{b}_j^3 b_j \\
    c(0) &= \sum c_j = 0 & \dot{c}(0) &= i \sum \tilde{c}_j c_j & \ddot{c}(0) &= -\sum \tilde{c}_j^2 c_j & \dot{\dddot{c}}(0) &= -i \sum \tilde{c}_j^3 c_j \\
    d(0) &= \sum d_j = 1 & \dot{d}(0) &= i \sum \tilde{d}_j d_j & \ddot{d}(0) &= -\sum \tilde{d}_j^2 d_j & \dot{\dddot{d}}(0) &= -i \sum \tilde{d}_j^3 d_j
\end{align*}
\] (B.24)

And I have the following differential equations

\[
\begin{align*}
    i\dot{A}(t) &= -\tilde{D}A(t) + \alpha B(t) + \epsilon \Gamma(t) \\
    i\dot{B}(t) &= \alpha A(t) - \delta \Delta(t) \\
    i\dot{\Gamma}(t) &= \epsilon A(t) + \beta \Delta(t) \\
    i\dot{\Delta}(t) &= -\delta B(t) + \beta \Gamma(t)
\end{align*}
\] (B.28)
Which become the following when transformed back:

\[ i \dot{A}(t) = -\tilde{D}A(t) + \alpha B(t) + \frac{\epsilon}{\sqrt{2}}(D(t) - C(t)) \] (B.32)
\[ i \dot{B}(t) = \alpha A(t) - \frac{\delta}{\sqrt{2}}(D(t) + C(t)) \] (B.33)
\[ i \dot{C}(t) = \frac{1}{\sqrt{2}}(-\epsilon A(t) - \delta B(t)) - \beta C(t) \] (B.34)
\[ i \dot{D}(t) = \frac{1}{\sqrt{2}}(\epsilon A(t) - \delta B(t)) + \beta D(t) \] (B.35)

\[ i(\dot{a}(t)e^{it\frac{\omega}{4}} + \frac{i}{4}\tilde{D}a(t)e^{it\frac{\omega}{4}}) = -\tilde{D}a(t)e^{it\frac{\omega}{4}} + \alpha b(t)e^{it\frac{\omega}{4}} + \frac{\epsilon}{\sqrt{2}}(d(t)e^{i\omega t - i\frac{\omega}{4}} - c(t)e^{-i\omega t + i\frac{\omega}{4}}) \] (B.36)
\[ i(\dot{b}(t)e^{-it\frac{\omega}{4}} + \frac{i}{4}\tilde{D}b(t)e^{-it\frac{\omega}{4}}) = \alpha a(t)e^{it\frac{\omega}{4}} - \frac{\delta}{\sqrt{2}}(d(t)e^{i\omega t + i\frac{\omega}{4}} + c(t)e^{-i\omega t + i\frac{\omega}{4}}) \] (B.37)
\[ i(\dot{c}(t)e^{-i\omega t + it\frac{\omega}{4}} + (-i\omega + \frac{i}{4}\tilde{D})c(t)e^{-i\omega t + it\frac{\omega}{4}}) = \frac{1}{\sqrt{2}}(-\epsilon a(t)e^{it\frac{\omega}{4}} - \delta b(t)e^{it\frac{\omega}{4}} - \beta c(t)e^{-i\omega t + it\frac{\omega}{4}}) \] (B.38)
\[ i(\dot{d}(t)e^{i\omega t + it\frac{\omega}{4}} + (i\omega + \frac{i}{4}\tilde{D})d(t)e^{i\omega t + it\frac{\omega}{4}}) = \frac{1}{\sqrt{2}}(\epsilon a(t)e^{it\frac{\omega}{4}} - \delta b(t)e^{it\frac{\omega}{4}} + \beta d(t)e^{i\omega t + it\frac{\omega}{4}}) \] (B.39)
\[ \begin{align*}
i(\dot{a}(t) + \frac{i\tilde{D}}{4}a(t)) &= -\tilde{D}a(t) + ab(t) + \frac{\epsilon}{\sqrt{2}}(d(t)e^{i\omega t} - c(t)e^{-i\omega t}) \quad (B.40) \\
i(\dot{b}(t) + \frac{i\tilde{D}}{4}b(t)) &= \alpha a(t) - \frac{\delta}{\sqrt{2}}(d(t)e^{i\omega t} + c(t)e^{-i\omega t}) \quad (B.41) \\
i(\dot{c}(t)e^{-i\omega t} + (-i\omega + \frac{i\tilde{D}}{4})c(t)e^{-i\omega t}) &= \frac{1}{\sqrt{2}}(-\epsilon a(t) - \delta b(t)) - \beta c(t)e^{-i\omega t} \quad (B.42) \\
i(\dot{d}(t)e^{i\omega t} + (i\omega + \frac{i\tilde{D}}{4})d(t)e^{i\omega t}) &= \frac{1}{\sqrt{2}}(\epsilon a(t) - \delta b(t)) + \beta d(t)e^{i\omega t} \quad (B.43)
\end{align*} \]

\[ \begin{align*}
i\dot{a}(t) &= (-\frac{3\tilde{D}}{4})a(t) + ab(t) + \frac{\epsilon}{\sqrt{2}}e^{i\omega t}d(t) - \frac{\epsilon}{\sqrt{2}}e^{-i\omega t}c(t) \quad (B.44) \\
i\dot{b}(t) &= \alpha a(t) + \frac{\tilde{D}}{4}b(t) - \frac{\delta}{\sqrt{2}}e^{i\omega t}d(t) - \frac{\delta}{\sqrt{2}}e^{-i\omega t}c(t) \quad (B.45) \\
i\dot{c}(t)e^{-i\omega t} &= -\frac{\epsilon}{\sqrt{2}}a(t) - \frac{\delta}{\sqrt{2}}b(t) + (-\beta - \omega + \frac{\tilde{D}}{4})e^{-i\omega t}c(t) \quad (B.46) \\
i\dot{d}(t)e^{i\omega t} &= \frac{\epsilon}{\sqrt{2}}a(t) - \frac{\delta}{\sqrt{2}}b(t) + (\beta + \omega + \frac{\tilde{D}}{4})e^{i\omega t}d(t) \quad (B.47)
\end{align*} \]

Now let

\[ \frac{\delta}{\sqrt{2}} = \delta' \quad (B.48) \]
\[ \frac{\epsilon}{\sqrt{2}} = \epsilon' \quad (B.49) \]

This simplifies to

\[ \begin{align*}
i\dot{a}(t) &= (-\frac{3\tilde{D}}{4})a(t) + ab(t) + \epsilon'e^{i\omega t}d(t) - \epsilon'e^{-i\omega t}c(t) \quad (B.50) \\
i\dot{b}(t) &= \alpha a(t) + \frac{\tilde{D}}{4}b(t) - \delta'e^{i\omega t}d(t) - \delta'e^{-i\omega t}c(t) \quad (B.51) \\
i\dot{c}(t)e^{-i\omega t} &= -\epsilon'e^{i\omega t}a(t) - \delta'e^{i\omega t}b(t) + (-\beta - \omega + \frac{\tilde{D}}{4})c(t) \quad (B.52) \\
i\dot{d}(t)e^{i\omega t} &= \epsilon'e^{-i\omega t}a(t) - \delta'e^{-i\omega t}b(t) + (\beta + \omega + \frac{\tilde{D}}{4})d(t) \quad (B.53)
\end{align*} \]
\[ i\ddot{a}(t) = (-\frac{3\bar{D}}{4})\dot{a}(t) + \alpha\dot{a}(t) + \epsilon' e^{i\omega t} \dot{d}(t) + i\omega e' e^{i\omega t} d(t) - \epsilon' e^{-i\omega t} \dot{c}(t) + i\omega e' e^{-i\omega t} c(t) \] (B.54)

\[ i\ddot{b}(t) = \alpha\dot{a}(t) + \frac{\bar{D}}{4}\dot{b}(t) - \delta' e^{i\omega t} \dot{d}(t) - i\omega\delta' e^{i\omega t} d(t) - \delta' e^{-i\omega t} \dot{c}(t) + i\omega\delta' e^{-i\omega t} c(t) \] (B.55)

\[ i\ddot{c}(t) = -\epsilon' e^{i\omega t} \dot{a}(t) - i\omega e' e^{i\omega t} a(t) - \delta' e^{i\omega t} \dot{b}(t) - i\omega\delta' e^{i\omega t} b(t) + (-\beta - \omega + \frac{\bar{D}}{4})\dot{c}(t) \] (B.56)

\[ i\ddot{d}(t) = \epsilon' e^{-i\omega t} \dot{a}(t) - i\omega e^{-i\omega t} a(t) - \delta' e^{-i\omega t} \dot{b}(t) + i\omega\delta' e^{-i\omega t} b(t) + (\beta + \omega + \frac{\bar{D}}{4})\dot{d}(t) \] (B.57)

\[ i\dddot{a}(t) = \] (B.58)

\[ \left(-\frac{3\bar{D}}{4}\right)\ddot{a}(t) + \alpha\ddot{a}(t) + \epsilon' e^{i\omega t} \ddot{d}(t) + 2i\omega e' e^{i\omega t} \ddot{d}(t) - \omega^2 \epsilon' e^{i\omega t} \dot{c}(t) + 2i\omega e' e^{-i\omega t} \dot{c}(t) + \omega^2 \epsilon' e^{-i\omega t} c(t) \]

\[ i\dddot{b}(t) = \] (B.59)

\[ \alpha\ddot{a}(t) + \frac{\bar{D}}{4}\ddot{b}(t) - \delta' e^{i\omega t} \ddot{d}(t) - 2i\omega\delta' e^{i\omega t} \ddot{d}(t) + \omega^2 \delta' e^{i\omega t} d(t) - \delta' e^{-i\omega t} \ddot{c}(t) + 2i\omega\delta' e^{-i\omega t} \ddot{c}(t) + \omega^2 \delta' e^{-i\omega t} c(t) \]

\[ i\dddot{c}(t) = \] (B.60)

\[ -\epsilon' e^{i\omega t} \dddot{a}(t) - 2i\omega e' e^{i\omega t} \dddot{a}(t) + \omega^2 \epsilon' e^{i\omega t} a(t) - \delta' e^{i\omega t} \dddot{b}(t) - 2i\omega\delta' e^{i\omega t} \dddot{b}(t) + \omega^2 \delta' e^{i\omega t} b(t) + (-\beta - \omega + \frac{\bar{D}}{4})\dddot{c}(t) \]

\[ i\dddot{d}(t) = \] (B.61)

\[ \epsilon' e^{-i\omega t} \dddot{a}(t) - 2i\omega e^{-i\omega t} \dddot{a}(t) - \omega^2 \epsilon' e^{-i\omega t} a(t) - \delta' e^{-i\omega t} \dddot{b}(t) + 2i\omega\delta' e^{-i\omega t} \dddot{b}(t) + \omega^2 \delta' e^{-i\omega t} b(t) + (\beta + \omega + \frac{\bar{D}}{4})\dddot{d}(t) \]
Now at $t = 0$

\[ i\dot{a}(0) = \epsilon' \rightarrow \dot{a}(0) = -i\epsilon' \]  
(B.62)

\[ i\dot{b}(0) = -\delta' \rightarrow \dot{b}(0) = i\delta' \]  
(B.63)

\[ i\dot{c}(0) = 0 \rightarrow \dot{c}(0) = 0 \]  
(B.64)

\[ i\ddot{d}(0) = \beta + \omega + \frac{\tilde{D}}{4} \rightarrow \ddot{d}(0) = i(-\beta - \omega - \frac{\tilde{D}}{4}) \]  
(B.65)

\[ i\dddot{a}(0) = \]  
\[ \left(-\frac{3\tilde{D}}{4}\right)(-i\epsilon') + \alpha(i\delta') + \epsilon'i(-\beta - \omega - \frac{\tilde{D}}{4}) + i\omega' \]  
\[ \rightarrow \dddot{a}(0) = \frac{\tilde{D}}{2}\epsilon' + \alpha\delta' - \beta\epsilon' \]  
(B.66)

\[ i\ddot{b}(0) = \]  
\[ \alpha(-i\epsilon') + \frac{\tilde{D}}{4}(i\delta') - \delta'i(-\beta - \omega - \frac{\tilde{D}}{4}) - i\omega\delta' \]  
\[ \rightarrow \ddot{b}(0) = \frac{\tilde{D}}{2}\delta' - \alpha\epsilon' + \beta\delta' \]  
(B.67)

\[ i\dddot{c}(0) = \]  
\[ -\epsilon'(-i\epsilon') - \delta'(i\delta') \rightarrow \dddot{c}(0) = \epsilon'^2 - \delta'^2 \]  
(B.68)

\[ i\dddot{d}(0) = \]  
\[ \epsilon'(-i\epsilon') - \delta'(i\delta') + (\beta + \omega + \frac{\tilde{D}}{4})i(-\beta - \omega - \frac{\tilde{D}}{4}) \]  
\[ \rightarrow \dddot{d}(0) = -\epsilon'^2 - \delta'^2 - (\beta - \omega - \frac{\tilde{D}}{4})^2 \]  
(B.69)
\[ i\dot{a}(0) = \]
\[ = \left( -\frac{3\tilde{D}}{4} \right) \left( \frac{\tilde{D}}{2} \epsilon' + \alpha \delta' - \beta \epsilon' \right) + \alpha \left( \frac{\tilde{D}}{2} \delta' - \alpha \epsilon' + \beta \delta' \right) \]
\[ + \epsilon' \left( -\epsilon'^2 - \delta'^2 - (-\beta - \omega - \frac{\tilde{D}}{4})^2 \right) \]
\[ + 2i\omega \delta' \left( -\beta - \omega - \frac{\tilde{D}}{4} \right) - \omega^2 \epsilon' \]
\[ - \epsilon'(\epsilon'^2 - \delta'^2) \]
\[ \text{(B.70)} \]

\[ i\dot{b}(0) = \]
\[ = \alpha \left( \frac{\tilde{D}}{2} \epsilon' + \alpha \delta' - \beta \epsilon' \right) \]
\[ + \tilde{D} \left( \frac{\tilde{D}}{2} \delta' - \alpha \epsilon' + \beta \delta' \right) \]
\[ - \delta' \left( -\epsilon'^2 - \delta'^2 - (-\beta - \omega - \frac{\tilde{D}}{4})^2 \right) \]
\[ - 2i\omega \delta' i(-\beta - \omega - \frac{\tilde{D}}{4}) \]
\[ + \omega^2 \delta' - \delta'(\epsilon'^2 - \delta'^2) \]
\[ \text{(B.71)} \]

\[ i\dot{c}(0) = \]
\[ = -\epsilon' \left( \frac{\tilde{D}}{2} \epsilon' + \alpha \delta' - \beta \epsilon' \right) - 2i\omega \epsilon' (-i\epsilon') \]
\[ - \delta' \left( \frac{\tilde{D}}{2} \delta' - \alpha \epsilon' + \beta \delta' \right) - 2i\omega \delta'(i\delta') \]
\[ + (-\beta - \omega + \frac{\tilde{D}}{4})(\epsilon'^2 - \delta'^2) \]
\[ \text{(B.72)} \]

\[ i\dot{d}(0) = \]
\[ = \epsilon' \left( \frac{\tilde{D}}{2} \epsilon' + \alpha \delta' - \beta \epsilon' \right) - 2i\omega \epsilon' (-i\epsilon') \]
\[ - \delta' \left( \frac{\tilde{D}}{2} \delta' - \alpha \epsilon' + \beta \delta' \right) + 2i\omega \delta'(i\delta') \]
\[ + (\beta + \omega + \frac{\tilde{D}}{4})(-\epsilon'^2 - \delta'^2 - (-\beta - \omega - \frac{\tilde{D}}{4})^2) \]
\[ \text{(B.73)} \]
So

\begin{align*}
a(0) &= 0 \quad \text{(B.74)} \\
b(0) &= 0 \quad \text{(B.75)} \\
c(0) &= 0 \quad \text{(B.76)} \\
d(0) &= 1 \quad \text{(B.77)} \\
\dot{a}(0) &= -i\epsilon' \quad \text{(B.78)} \\
\dot{b}(0) &= i\delta' \quad \text{(B.79)} \\
\dot{c}(0) &= 0 \quad \text{(B.80)} \\
\ddot{a}(0) &= i(-\beta - \omega - \frac{\tilde{D}}{4}) \quad \text{(B.81)} \\
\ddot{b}(0) &= i\frac{\tilde{D}}{2}\epsilon' + \alpha\delta' - \beta\epsilon' \quad \text{(B.82)} \\
\ddot{c}(0) &= \epsilon'^2 - \delta'^2 \quad \text{(B.84)} \\
\ddot{d}(0) &= -\epsilon'^2 - \delta'^2 - \frac{(-\beta - \omega - \frac{\tilde{D}}{4})^2}{4} \quad \text{(B.85)}
\end{align*}

\begin{align*}
\dddot{a}(0) &= -i\left[-\frac{7\tilde{D}^2}{16}\epsilon' - \frac{\tilde{D}}{4}\alpha\delta' + \frac{\tilde{D}}{4}\beta\epsilon' - \alpha^2\epsilon + \alpha\beta\delta' - \beta^2\epsilon' - 2\epsilon'^3\right] \quad \text{(B.86)} \\
\dddot{b}(0) &= -i\left[\frac{\tilde{D}}{4}\epsilon' + \alpha^2\delta' - \alpha\beta\epsilon' + \frac{3\tilde{D}}{4}\beta\delta' + 2\delta'^3 + \delta'\beta'^2 + \frac{3\tilde{D}^2}{16}\delta'ight] \quad \text{(B.87)} \\
\dddot{c}(0) &= -i\left[-\frac{\tilde{D}}{4}\epsilon'^2 - \frac{3\tilde{D}}{4}\delta'^2 - 3\omega\epsilon'^2 + 3\omega\delta'^2\right] \quad \text{(B.88)} \\
\dddot{d}(0) &= -i\left[-\frac{\tilde{D}}{4}\epsilon'^2 - \frac{3\tilde{D}}{4}\delta'^2 - 2\beta\epsilon'^2 - 2\beta\delta'^2 - 3\omega\epsilon'^2 - 3\omega\delta'^2 + 2\alpha\delta'\epsilon' + \beta^3 + 3\beta^2\omega + 3\beta\omega^2 \right. \\
&\quad + \left. \frac{3\tilde{D}}{2}\omega\beta + \frac{3\tilde{D}^2}{16}\beta + \omega^3 + \frac{3\tilde{D}^2}{16}\omega + \frac{3\tilde{D}}{4}\beta^2 + \frac{3\tilde{D}}{4}\omega^2 + \frac{\tilde{D}^3}{64}\right] \quad \text{(B.89)}
\end{align*}
\[ \sum a_j = 0 \quad \text{(B.90)} \]
\[ \sum \tilde{a}_j a_j = -\epsilon' \quad \text{(B.91)} \]
\[ \sum \tilde{a}_j^2 a_j = -\frac{\tilde{D}}{2} \epsilon' - \alpha \delta' + \beta \epsilon' \quad \text{(B.92)} \]
\[ \sum \tilde{a}_j^3 a_j = -\frac{7\tilde{D}^2}{16} \epsilon' - \frac{\tilde{D}}{4} \alpha \delta' + \frac{\tilde{D}}{4} \beta \epsilon' - \alpha^2 \epsilon' + \alpha \beta \delta' - \beta^2 \epsilon' - 2\epsilon'^3 \quad \text{(B.93)} \]

\[ \sum b_j = 0 \quad \text{(B.94)} \]
\[ \sum \tilde{b}_j b_j = \delta' \quad \text{(B.95)} \]
\[ \sum \tilde{b}_j^2 b_j = -\frac{\tilde{D}}{2} \delta' + \alpha \epsilon' - \beta \delta' \quad \text{(B.96)} \]
\[ \sum \tilde{b}_j^3 b_j = \frac{\tilde{D}}{4} \alpha \epsilon' + \alpha^2 \delta' - \alpha \beta \epsilon' + \frac{3\tilde{D}}{4} \beta \delta' + 2\delta'^2 + \frac{3\tilde{D}^2}{16} \delta' \quad \text{(B.97)} \]

\[ \sum c_j = 0 \quad \text{(B.98)} \]
\[ \sum \tilde{c}_j c_j = 0 \quad \text{(B.99)} \]
\[ \sum \tilde{c}_j^2 c_j = -\epsilon'^2 + \delta'^2 \quad \text{(B.100)} \]
\[ \sum \tilde{c}_j^3 c_j = -\frac{\tilde{D}}{4} \epsilon'^2 - \frac{3\tilde{D}}{4} \delta'^2 - 3\omega \epsilon'^2 + 3\omega \delta'^2 \quad \text{(B.101)} \]
\[ \sum d_j = 1 \tag{B.102} \]

\[ \sum \tilde{d}_j d_j = -\beta - \omega - \frac{\tilde{D}}{4} \tag{B.103} \]

\[ \sum \tilde{d}_j^2 d_j = \epsilon'^2 + \delta'^2 + (-\beta - \omega - \frac{\tilde{D}}{4})^2 \tag{B.104} \]

\[ \sum \tilde{d}_j^3 d_j = -\frac{\tilde{D}}{4} \epsilon'^2 - \frac{3\tilde{D}^2}{16} \delta + \frac{3\tilde{D}^2}{16} \beta + \frac{3\tilde{D}^2}{16} \omega + \frac{3\tilde{D}}{4} \beta^2 + \frac{3\tilde{D}}{4} \omega^2 + \tilde{D}^3 \tag{B.105} \]

Now I need \( e = \frac{a+b}{\sqrt{2}} \).

\[ \sum e_j = 0 \tag{B.106} \]

\[ \sum \tilde{e}_j e_j = \frac{1}{\sqrt{2}} (-\epsilon' + \delta') \tag{B.107} \]

\[ \sum \tilde{e}_j^2 e_j = \frac{1}{\sqrt{2}} (-\frac{\tilde{D}}{2} \epsilon' - \alpha \delta' + \beta \epsilon' - \frac{\tilde{D}}{2} \delta' + \alpha \epsilon' - \beta \delta') \tag{B.108} \]

\[ \sum \tilde{e}_j^3 e_j = \frac{1}{\sqrt{2}} (-\frac{7\tilde{D}^2}{16} \epsilon' - \frac{\tilde{D}}{4} \alpha \delta' + \frac{\tilde{D}}{4} \beta \epsilon' - \alpha^2 \epsilon + \alpha \beta \epsilon' - \beta^2 \epsilon' - 2 \epsilon'^3 \]
\[ + \frac{\tilde{D}}{4} \alpha \epsilon' + \alpha^2 \delta' - \alpha \beta \epsilon' + \frac{3\tilde{D}}{4} \beta \delta' + 2 \delta'^3 + \delta' \beta^2 + \frac{3\tilde{D}^2}{16} \delta' \tag{B.109} \]

And I need \( f = \frac{b-a}{\sqrt{2}} \).

\[ \sum f_j = 0 \tag{B.110} \]

\[ \sum \tilde{f}_j f_j = \frac{1}{\sqrt{2}} (\epsilon' + \delta') \tag{B.111} \]

\[ \sum \tilde{f}_j^2 f_j = \frac{1}{\sqrt{2}} (\frac{\tilde{D}}{2} \epsilon' + \alpha \delta' - \beta \epsilon' - \frac{\tilde{D}}{2} \delta' + \alpha \epsilon' - \beta \delta') \tag{B.112} \]

\[ \sum \tilde{f}_j^3 f_j = \frac{1}{\sqrt{2}} (\frac{7\tilde{D}^2}{16} \epsilon' + \frac{\tilde{D}}{4} \alpha \delta' - \frac{\tilde{D}}{4} \beta \epsilon' + \alpha^2 \epsilon - \alpha \beta \delta' + \beta^2 \epsilon' + 2 \epsilon'^3 \]
\[ + \frac{\tilde{D}}{4} \alpha \epsilon' + \alpha^2 \delta' - \alpha \beta \epsilon' + \frac{3\tilde{D}}{4} \beta \delta' + 2 \delta'^3 + \delta' \beta^2 + \frac{3\tilde{D}^2}{16} \delta' \tag{B.113} \]
So finally I have

\[ \sum e_j = 0 \] \text{ (B.114)}

\[ \sum (\gamma_j - \frac{\tilde{D}}{4})e_j = \frac{1}{2}(-\epsilon + \delta) \] \text{ (B.115)}

\[ \sum (\gamma_j - \frac{\tilde{D}}{4})^2e_j = \frac{1}{2}(-\frac{\tilde{D}}{2} - \alpha \delta + \beta \epsilon - \frac{\tilde{D}}{2} \delta + \alpha \epsilon - \beta \delta) \] \text{ (B.116)}

\[ \sum (\gamma_j - \frac{\tilde{D}}{4})^3e_j = \frac{1}{2}(-\frac{7\tilde{D}^2}{16} - \frac{\tilde{D}}{4} \alpha \delta + \frac{\tilde{D}}{4} \beta \epsilon - \frac{\tilde{D}}{4} \alpha^2 \epsilon + \alpha \beta \delta - \beta^2 \epsilon - \epsilon^3 + \frac{\tilde{D}}{4} \alpha \epsilon + \frac{3\tilde{D}}{4} \beta \delta + \delta^3 + \delta \beta^2 + \frac{3\tilde{D}^2}{16} \delta) \] \text{ (B.117)}

\[ \sum f_j = 0 \] \text{ (B.118)}

\[ \sum (\gamma_j - \frac{\tilde{D}}{4})f_j = \frac{1}{2}(\epsilon + \delta) \] \text{ (B.119)}

\[ \sum (\gamma_j - \frac{\tilde{D}}{4})^2f_j = \frac{1}{2}(\frac{\tilde{D}}{2} \epsilon + \alpha \delta - \beta \epsilon - \frac{\tilde{D}}{2} \delta + \alpha \epsilon - \beta \delta) \] \text{ (B.120)}

\[ \sum (\gamma_j - \frac{\tilde{D}}{4})^3f_j = \frac{1}{2}(\frac{7\tilde{D}^2}{16} \epsilon + \frac{\tilde{D}}{4} \alpha \delta - \frac{\tilde{D}}{4} \beta \epsilon + \frac{\tilde{D}}{4} \alpha^2 \epsilon - \alpha \beta \delta + \beta^2 \epsilon + \epsilon^3 + \frac{\tilde{D}}{4} \alpha \epsilon + \frac{3\tilde{D}}{4} \beta \delta + \delta^3 + \delta \beta^2 + \frac{3\tilde{D}^2}{16} \delta) \] \text{ (B.121)}
B.3 Initial State $c\left(-\frac{1}{2},-\frac{1}{2}\right)$

Now lets assume we are instead in the $|\downarrow\downarrow\rangle$ state. This means $a(0) = b(0) = d(0) = 0$ and $c(0) = 1$.

$$a(0) = \sum a_j = 0 \quad \dot{a}(0) = i \sum \tilde{a}_j a_j \quad \ddot{a}(0) = -\sum \tilde{a}_j^2 a_j \quad \dddot{a}(0) = -i \sum \tilde{a}_j^3 a_j \quad (B.122)$$

$$b(0) = \sum b_j = 0 \quad \dot{b}(0) = i \sum \tilde{b}_j b_j \quad \ddot{b}(0) = -\sum \tilde{b}_j^2 b_j \quad \dddot{b}(0) = -i \sum \tilde{b}_j^3 b_j \quad (B.123)$$

$$c(0) = \sum c_j = 1 \quad \dot{c}(0) = i \sum \tilde{c}_j c_j \quad \ddot{c}(0) = -\sum \tilde{c}_j^2 c_j \quad \dddot{c}(0) = -i \sum \tilde{c}_j^3 c_j \quad (B.124)$$

$$d(0) = \sum d_j = 0 \quad \dot{d}(0) = i \sum \tilde{d}_j d_j \quad \ddot{d}(0) = -\sum \tilde{d}_j^2 d_j \quad \dddot{d}(0) = -i \sum \tilde{d}_j^3 d_j \quad (B.125)$$

And I have the following differential equations

$$i\dot{A}(t) = -\tilde{D}A(t) + \alpha B(t) + \epsilon \Gamma(t) \quad (B.126)$$

$$i\dot{B}(t) = \alpha A(t) - \delta \Delta(t) \quad (B.127)$$

$$i\dot{\Gamma}(t) = \epsilon A(t) + \beta \Delta(t) \quad (B.128)$$

$$i\dot{\Delta}(t) = -\delta B(t) + \beta \Gamma(t) \quad (B.129)$$

Which become the following when transformed back:

$$i\dot{A}(t) = -\tilde{D}A(t) + \alpha B(t) + \frac{\epsilon}{\sqrt{2}}(D(t) - C(t)) \quad (B.130)$$

$$i\dot{B}(t) = \alpha A(t) - \frac{\delta}{\sqrt{2}}(D(t) + C(t)) \quad (B.131)$$

$$i\dot{C}(t) = \frac{1}{\sqrt{2}}(-\epsilon A(t) - \delta B(t)) - \beta C(t) \quad (B.132)$$

$$i\dot{D}(t) = \frac{1}{\sqrt{2}}(\epsilon A(t) - \delta B(t)) + \beta D(t) \quad (B.133)$$
\[ \begin{align*}
  i(\dot{a}(t)e^{i\Omega t/4} + i\tilde{D}/4 a(t)e^{i\Omega t/4}) &= -\tilde{D}a(t)e^{i\Omega t/4} + \alpha b(t)e^{i\Omega t/4} + \epsilon/\sqrt{2}(d(t)e^{i\omega t + i\Omega t/4} - c(t)e^{-i\omega t + i\Omega t/4}) \\
  i(\dot{b}(t)e^{i\Omega t/4} + i\tilde{D}/4 b(t)e^{i\Omega t/4}) &= \alpha a(t)e^{i\Omega t/4} - \delta/\sqrt{2}(d(t)e^{i\omega t + i\Omega t/4} + c(t)e^{-i\omega t + i\Omega t/4}) \\
  i(\dot{c}(t)e^{-i\omega t + i\Omega t/4} + (-i\omega + i\tilde{D}/4)c(t)e^{-i\omega t + i\Omega t/4}) &= 1/\sqrt{2}(-\epsilon a(t)e^{i\Omega t/4} - \delta b(t)e^{i\Omega t/4}) \\
  i(\dot{d}(t)e^{i\omega t + i\Omega t/4} + (i\omega + i\tilde{D}/4)d(t)e^{i\omega t + i\Omega t/4}) &= 1/\sqrt{2}(\epsilon a(t)e^{i\Omega t/4} - \delta b(t)e^{i\Omega t/4}) + \beta d(t)e^{i\Omega t/4}
\end{align*} \]
Now let

\[ \frac{\delta}{\sqrt{2}} = \delta' \]  \hspace{1cm} (B.146)
\[ \frac{\epsilon}{\sqrt{2}} = \epsilon' \]  \hspace{1cm} (B.147)

\[ i \dot{a}(t) = \left( - \frac{3\tilde{D}}{4} \right) a(t) + \alpha b(t) + \epsilon' e^{i\omega t} d(t) - \epsilon' e^{-i\omega t} c(t) \]  \hspace{1cm} (B.148)
\[ i \dot{b}(t) = \alpha a(t) + \frac{\tilde{D}}{4} b(t) - \delta' e^{i\omega t} d(t) - \delta' e^{-i\omega t} c(t) \]  \hspace{1cm} (B.149)
\[ i \dot{c}(t) = -\epsilon' e^{i\omega t} a(t) - \delta' e^{i\omega t} b(t) + (-\beta - \omega + \frac{\tilde{D}}{4}) c(t) \]  \hspace{1cm} (B.150)
\[ i \dot{d}(t) = \epsilon' e^{-i\omega t} a(t) - \delta' e^{-i\omega t} b(t) + (\beta + \omega + \frac{\tilde{D}}{4}) d(t) \]  \hspace{1cm} (B.151)

\[ i \ddot{a}(t) = \left( - \frac{3\tilde{D}}{4} \right) \ddot{a}(t) + \alpha \dot{b}(t) + \epsilon' e^{i\omega t} \dot{d}(t) + i\omega \epsilon' e^{i\omega t} d(t) - \epsilon' e^{-i\omega t} \dot{c}(t) + i\omega \epsilon' e^{-i\omega t} c(t) \]  \hspace{1cm} (B.152)
\[ i \ddot{b}(t) = \alpha \ddot{a}(t) + \frac{\tilde{D}}{4} \ddot{b}(t) - \delta' e^{i\omega t} \dot{d}(t) - i\omega \delta' e^{i\omega t} d(t) - \delta' e^{-i\omega t} \dot{c}(t) + i\omega \delta' e^{-i\omega t} c(t) \]  \hspace{1cm} (B.153)
\[ i \ddot{c}(t) = -\epsilon' e^{i\omega t} \ddot{a}(t) - i\omega \epsilon' e^{i\omega t} a(t) - \delta' e^{i\omega t} \ddot{b}(t) - i\omega \delta' e^{i\omega t} b(t) + (-\beta - \omega + \frac{\tilde{D}}{4}) \ddot{c}(t) \]  \hspace{1cm} (B.154)
\[ i \ddot{d}(t) = \epsilon' e^{-i\omega t} \ddot{a}(t) - i\omega \epsilon' e^{-i\omega t} a(t) - \delta' e^{-i\omega t} \ddot{b}(t) + i\omega \delta' e^{-i\omega t} b(t) + (\beta + \omega + \frac{\tilde{D}}{4}) \dddot{d}(t) \]  \hspace{1cm} (B.155)
\[ i \dddot{a}(t) = \left(- \frac{3\tilde{D}}{4}\right) \dddot{a}(t) + \alpha \dddot{b}(t) + \epsilon' e^{i\omega t} \dddot{d}(t) + 2i \omega \epsilon' e^{i\omega t} \dot{d}(t) \]
\[ - \omega^2 \epsilon' e^{i\omega t} d(t) - \epsilon' e^{-i\omega t} \dddot{c}(t) + 2i \omega \epsilon' e^{-i\omega t} \dot{c}(t) + \omega^2 \epsilon' e^{-i\omega t} c(t) \]
\[ i \dddot{b}(t) = \alpha \dddot{a}(t) + \frac{\tilde{D}}{4} \dddot{b}(t) - \delta' e^{i\omega t} \dddot{d}(t) - 2i \omega \delta' e^{i\omega t} \dot{d}(t) \]
\[ + \omega^2 \delta' e^{i\omega t} d(t) - \delta' e^{-i\omega t} \dddot{c}(t) + 2i \omega \delta' e^{-i\omega t} \dot{c}(t) + \omega^2 \delta' e^{-i\omega t} c(t) \]
\[ i \dddot{c}(t) = -\epsilon' e^{i\omega t} \dddot{a}(t) - 2i \omega \epsilon' e^{i\omega t} \dddot{a}(t) + \omega^2 \epsilon' e^{i\omega t} \dddot{a}(t) \]
\[ - \delta' e^{i\omega t} \dddot{b}(t) - 2i \omega \delta' e^{i\omega t} \dddot{b}(t) + \omega^2 \delta' e^{i\omega t} \dddot{b}(t) + (-\beta - \omega + \frac{\tilde{D}}{4}) \dddot{c}(t) \]
\[ i \dddot{d}(t) = \epsilon' e^{-i\omega t} \dddot{a}(t) - 2i \omega \epsilon' e^{-i\omega t} \dddot{a}(t) - \omega^2 \epsilon' e^{-i\omega t} \dddot{a}(t) \]
\[ - \delta' e^{-i\omega t} \dddot{b}(t) + 2i \omega \delta' e^{-i\omega t} \dddot{b}(t) + \omega^2 \delta' e^{-i\omega t} \dddot{b}(t) + (\beta + \omega + \frac{\tilde{D}}{4}) \dddot{d}(t) \]

Now at \( t = 0 \)
\[ i \dot{a}(0) = -\epsilon' \rightarrow \dot{a}(0) = i \epsilon' \quad (B.160) \]
\[ i \dot{b}(0) = -\delta' \rightarrow \dot{b}(0) = i \delta' \quad (B.161) \]
\[ i \dot{c}(0) = -\beta - \omega + \frac{\tilde{D}}{4} \rightarrow \dot{c}(0) = i(\beta + \omega - \frac{\tilde{D}}{4}) \quad (B.162) \]
\[ i \dot{d}(0) = 0 \rightarrow \dot{d}(0) = 0 \quad (B.163) \]
\[\dot{\ddot{a}}(0) = \left(-\frac{3\tilde{D}}{4}\right)(i\epsilon') + \alpha(i\delta') - \epsilon'i(\beta + \omega - \frac{\tilde{D}}{4}) + i\omega\epsilon'(\beta + \omega - \frac{\tilde{D}}{4})\]  
(B.164)

\[\dot{b}(0) = \alpha(i\epsilon') + \frac{\tilde{D}}{4}(i\delta') - \delta'i(\beta + \omega - \frac{\tilde{D}}{4})\]  
(B.165)

\[\ddot{c}(0) = -\epsilon'(i\epsilon') - \delta'(i\delta') + (-\beta - \omega + \frac{\tilde{D}}{4})i(\beta + \omega - \frac{\tilde{D}}{4})\]
(B.166)

\[\ddot{d}(0) = \epsilon'(i\epsilon') - \delta'(i\delta') \rightarrow \ddot{d}(0) = \epsilon'^2 - \delta'^2\]  
(B.167)

\[\dot{\ddot{a}}(0) = \left(-\frac{3\tilde{D}}{4}\right)(\frac{\tilde{D}}{2}\epsilon' + \alpha\delta' - \beta\epsilon') + \alpha\left(\frac{\tilde{D}}{2}\delta' + \alpha\epsilon' - \beta\delta'\right) + \epsilon'(\epsilon'^2 - \delta'^2)\]  
+ \[2i\omega\epsilon'i(\beta + \omega - \frac{\tilde{D}}{4}) + \omega^2\epsilon' - \epsilon'(-\epsilon'^2 - \delta'^2 - (\beta + \omega - \frac{\tilde{D}}{4})^2)\]  
(B.168)

\[\dot{\ddot{b}}(0) = \alpha\left(-\frac{\tilde{D}}{2}\epsilon' + \alpha\delta' - \beta\epsilon'\right) + \frac{\tilde{D}}{4}\left(\frac{\tilde{D}}{2}\delta' + \alpha\epsilon' - \beta\delta'\right)\]  

\(- \delta'(\epsilon'^2 - \delta'^2) + 2i\omega\delta'i(\beta + \omega - \frac{\tilde{D}}{4}) + \omega^2\delta'\]  
(B.169)

\[\dot{\ddot{c}}(0) = -\epsilon'\left(-\frac{\tilde{D}}{2}\epsilon' + \alpha\delta' - \beta\epsilon'\right) - 2i\omega\epsilon'(i\epsilon')\]  

\(- \delta'\left(\frac{\tilde{D}}{2}\delta' + \alpha\epsilon' - \beta\delta'\right) - 2i\omega\delta'(i\delta')\]  
(B.170)

\[\dot{\ddot{d}}(0) = \epsilon'\left(-\frac{\tilde{D}}{2}\epsilon' + \alpha\delta' - \beta\epsilon'\right) - 2i\omega\epsilon'(i\epsilon')\]  

\(- \delta'\left(\frac{\tilde{D}}{2}\delta' + \alpha\epsilon' - \beta\delta'\right) + 2i\omega\delta'(i\delta') + (\beta + \omega + \frac{\tilde{D}}{4})(\epsilon'^2 - \delta'^2)\]  
(B.171)
\begin{align*}
a(0) & = 0 \quad \text{(B.172)} \\
b(0) & = 0 \quad \text{(B.173)} \\
c(0) & = 1 \quad \text{(B.174)} \\
d(0) & = 0 \quad \text{(B.175)} \\
\dot{a}(0) & = i\epsilon' \\
\dot{b}(0) & = i\delta' \quad \text{(B.177)} \\
\dot{c}(0) & = i(\beta + \omega - \frac{\bar{D}}{4}) \quad \text{(B.178)} \\
\dot{d}(0) & = 0 \quad \text{(B.179)}
\end{align*}

\begin{align*}
\ddot{a}(0) & = -i\frac{7\bar{D}^2}{16}\epsilon' - \frac{\bar{D}}{4}\alpha\delta' + \frac{\bar{D}}{4}\beta\epsilon' + \alpha^2\epsilon' - \alpha\beta\delta' + \beta^2\epsilon' + 2\epsilon'^3 \quad \text{(B.184)} \\
\ddot{b}(0) & = -i[-\frac{\bar{D}}{4}\alpha\epsilon' + \alpha^2\delta' - \alpha\beta\epsilon' - \frac{3\bar{D}}{4}\beta\delta' + 2\delta'^3 + \delta'\beta^2 + \frac{3\bar{D}^2}{16}\delta'] \quad \text{(B.185)} \\
\ddot{c}(0) & = -i[-\frac{\bar{D}}{4}\epsilon'^2 - \frac{3\bar{D}}{4}\delta'^2 + 2\beta\epsilon'^2 + 2\beta\delta'^2 + 3\omega\epsilon'^2 + 3\omega\delta'^2 - 2\alpha\delta'\epsilon' + \beta^3 + 3\beta^2\omega + 3\beta\omega^2 \\
& \quad - \frac{3\bar{D}}{2}\omega\beta + \frac{3\bar{D}^2}{16}\beta + \omega^3 + \frac{3\bar{D}^2}{16}\omega - \frac{3\bar{D}}{4}\beta^2 - \frac{3\bar{D}}{4}\omega^2 - \frac{\bar{D}^3}{64}] \quad \text{(B.186)} \\
\ddot{d}(0) & = -i[-\frac{\bar{D}}{4}\epsilon'^2 - \frac{3\bar{D}}{4}\delta'^2 + 3\omega\epsilon'^2 - 3\omega\delta'^2] \quad \text{(B.187)}
\end{align*}
\[ \sum a_j = 0 \quad \text{(B.188)} \]
\[ \sum \tilde{a}_j a_j = \epsilon' \quad \text{(B.189)} \]
\[ \sum \tilde{a}_j^2 a_j = \frac{\tilde{D}}{2} \epsilon' - \alpha \delta' + \beta \epsilon' \quad \text{(B.190)} \]
\[ \sum \tilde{a}_j^3 a_j = \frac{7 \tilde{D}^2}{16} \epsilon' - \frac{\tilde{D}}{4} \alpha \delta' + \frac{\tilde{D}}{4} \beta \epsilon' + \alpha^2 \epsilon - \alpha \beta \delta' + \beta^2 \epsilon' + 2 \epsilon'^3 \quad \text{(B.191)} \]

\[ \sum b_j = 0 \quad \text{(B.192)} \]
\[ \sum \tilde{b}_j b_j = \delta' \quad \text{(B.193)} \]
\[ \sum \tilde{b}_j^2 b_j = -\frac{\tilde{D}}{2} \delta' - \alpha \epsilon' + \beta \delta' \quad \text{(B.194)} \]
\[ \sum \tilde{b}_j^3 b_j = -\frac{\tilde{D}}{4} \alpha \epsilon' + \alpha^2 \delta' - \alpha \beta \epsilon' - \frac{3 \tilde{D}}{4} \beta \delta' + 2 \delta'^3 + \delta' \beta^2 + \frac{3 \tilde{D}^2}{16} \delta' \quad \text{(B.195)} \]

\[ \sum c_j = 1 \quad \text{(B.196)} \]
\[ \sum \tilde{c}_j c_j = \beta + \omega - \frac{\tilde{D}}{4} \quad \text{(B.197)} \]
\[ \sum \tilde{c}_j^2 c_j = \epsilon'^2 + \delta'^2 + (\beta + \omega - \frac{\tilde{D}}{4})^2 \quad \text{(B.198)} \]
\[ \sum \tilde{c}_j^3 c_j = \frac{\tilde{D}}{4} \epsilon'^2 - \frac{3 \tilde{D}}{4} \delta'^2 + 2 \beta \epsilon'^2 + 2 \beta \delta'^2 + 3 \omega \epsilon'^2 + 3 \omega \delta'^2 - 2 \alpha \delta' \epsilon' + \beta^3 + 3 \beta^2 \omega + 3 \beta \omega^2 \]
\[ - \frac{3 \tilde{D}}{2} \omega \beta + \frac{3 \tilde{D}^2}{16} \beta + \omega^3 + \frac{3 \tilde{D}^2}{16} \omega - \frac{3 \tilde{D}}{4} \beta^2 - \frac{3 \tilde{D}}{4} \omega^2 - \frac{\tilde{D}^3}{64} \quad \text{(B.199)} \]
\[ \sum d_j = 0 \]  
\[ \sum \tilde{d}_j d_j = 0 \]  
\[ \sum d_j^2 d_j = -\epsilon'^2 + \delta'^2 \]  
\[ \sum \tilde{d}_j^3 d_j = -\frac{\tilde{D}}{4} \epsilon'^2 - \frac{3\tilde{D}}{4} \delta'^2 + 3\omega \epsilon'^2 - 3\omega \delta'^2 \]  
(B.200)  
(B.201)  
(B.202)  
(B.203)

Now I need \( e = \frac{a+b}{\sqrt{2}} \).

\[ \sum e_j = 0 \]  
\[ \sum \tilde{e}_j e_j = \frac{1}{\sqrt{2}} (\epsilon' + \delta') \]  
\[ \sum \tilde{e}_j^2 e_j = \frac{1}{\sqrt{2}} (\frac{\tilde{D}}{2} \epsilon' - \alpha \delta' + \beta \epsilon' - \frac{\tilde{D}}{2} \delta' - \alpha \epsilon' + \beta \delta') \]  
\[ \sum \tilde{e}_j^3 e_j = \frac{1}{\sqrt{2}} (\frac{7\tilde{D}^2}{16} \epsilon' - \frac{\tilde{D}}{4} \alpha \delta' + \frac{\tilde{D}}{4} \beta \epsilon' + \alpha^2 \epsilon' - \alpha \beta \delta' + \beta^2 \epsilon' + 2\epsilon'^3 \]  
\[ - \frac{\tilde{D}}{4} \alpha \epsilon' + \alpha^2 \delta' - \alpha \beta \epsilon' - \frac{3\tilde{D}}{4} \beta \delta' + 2\delta'^3 + \delta' \beta^2 + \frac{3\tilde{D}^2}{16} \delta' \) \]  
(B.204)  
(B.205)  
(B.206)  
(B.207)

And I need \( f = \frac{b-a}{\sqrt{2}} \).

\[ \sum f_j = 0 \]  
\[ \sum \tilde{f}_j f_j = \frac{1}{\sqrt{2}} (-\epsilon' + \delta') \]  
\[ \sum \tilde{f}_j^2 f_j = \frac{1}{\sqrt{2}} (-\frac{\tilde{D}}{2} \epsilon' + \alpha \delta' - \beta \epsilon' - \frac{\tilde{D}}{2} \delta' - \alpha \epsilon' + \beta \delta') \]  
\[ \sum \tilde{f}_j^3 f_j = \frac{1}{\sqrt{2}} (-\frac{7\tilde{D}^2}{16} \epsilon' + \frac{\tilde{D}}{4} \alpha \delta' - \frac{\tilde{D}}{4} \beta \epsilon' - \alpha^2 \epsilon' + \alpha \beta \delta' - \beta^2 \epsilon' - 2\epsilon'^3 \]  
\[ - \frac{\tilde{D}}{4} \alpha \epsilon' + \alpha^2 \delta' - \alpha \beta \epsilon' - \frac{3\tilde{D}}{4} \beta \delta' + 2\delta'^3 + \delta' \beta^2 + \frac{3\tilde{D}^2}{16} \delta' \) \]  
(B.208)  
(B.209)  
(B.210)  
(B.211)
So finally I have

\[ \sum \bar{e}_j = 0 \]  \hspace{1cm} (B.212)

\[ \sum (\gamma_j - \frac{\bar{D}}{4})\bar{e}_j = \frac{1}{2}(\epsilon + \delta) \]  \hspace{1cm} (B.213)

\[ \sum (\gamma_j - \frac{\bar{D}}{4})^2\bar{e}_j = \frac{1}{2}\left(\frac{\bar{D}}{2}\epsilon - \alpha\delta + \beta\epsilon - \frac{\bar{D}}{2}\delta - \alpha\epsilon + \beta\delta\right) \]  \hspace{1cm} (B.214)

\[ \sum (\gamma_j - \frac{\bar{D}}{4})^3\bar{e}_j = \frac{1}{2}\left(\frac{7\bar{D}^2}{16}\epsilon - \frac{\bar{D}}{4}\alpha\delta + \frac{\bar{D}}{4}\beta\epsilon + \alpha^2\epsilon - \alpha\beta\delta + \beta^2\epsilon + \epsilon^3 - \frac{\bar{D}}{4}\alpha\epsilon + \alpha^2\delta - \alpha\beta\epsilon - \frac{3\bar{D}}{4}\beta\delta + \delta^3 + \delta\beta^2 + \frac{3\bar{D}^2}{16}\delta\right) \]  \hspace{1cm} (B.215)

\[ \sum \bar{f}_j = 0 \]  \hspace{1cm} (B.216)

\[ \sum (\gamma_j - \frac{\bar{D}}{4})\bar{f}_j = \frac{1}{2}(-\epsilon + \delta) \]  \hspace{1cm} (B.217)

\[ \sum (\gamma_j - \frac{\bar{D}}{4})^2\bar{f}_j = \frac{1}{2}\left(-\frac{\bar{D}}{2}\epsilon + \alpha\delta - \beta\epsilon - \frac{\bar{D}}{2}\delta - \alpha\epsilon + \beta\delta\right) \]  \hspace{1cm} (B.218)

\[ \sum (\gamma_j - \frac{\bar{D}}{4})^3\bar{f}_j = \frac{1}{2}\left(-\frac{7\bar{D}^2}{16}\epsilon + \frac{\bar{D}}{4}\alpha\delta - \frac{\bar{D}}{4}\beta\epsilon - \alpha^2\epsilon + \alpha\beta\delta - \beta^2\epsilon - \epsilon^3 - \frac{\bar{D}}{4}\alpha\epsilon + \alpha^2\delta - \alpha\beta\epsilon - \frac{3\bar{D}}{4}\beta\delta + \delta^3 + \delta\beta^2 + \frac{3\bar{D}^2}{16}\delta\right) \]  \hspace{1cm} (B.219)
APPENDIX C: CHECKING LIMITS
C.1 Nuclear Magnetic Interaction

The original quartic equation for $\gamma$ is

$$0 = \left[ \gamma^2 - (\Omega_N + \Omega_B)^2 - (\omega - \frac{1}{2}(\omega_N + \omega_B))^2 \right] \left[ \gamma^2 - \gamma \tilde{D} - (\Omega_B - \Omega_N)^2 - \left( \frac{1}{2}(\omega_N - \omega_B) \right)^2 \right]$$

$$- \left[ \left( \frac{1}{2}(\omega_N - \omega_B) \right)(\Omega_N + \Omega_B) - (\omega - \frac{1}{2}(\omega_N + \omega_B))(\Omega_B - \Omega_N) \right]^2$$

(C.1)

I let $\tilde{D} \to 0$, $\Omega_B \to 0$, and $\omega_B \to 0$. Then the equation for $\gamma$ becomes

$$0 = \left( \gamma^2 - \Omega_N^2 - \frac{\omega_N^2}{2} \right)^2 - \left( \omega - \frac{\omega_N}{2} \right)^2 - \left( \Omega_N \omega - \frac{\omega_N^2}{2} \right)^2$$

(C.2)

$$= (\gamma^2 - \Omega_N^2 - \frac{\omega_N^2}{2})^2 - \Omega_N^2 \omega^2$$

(C.3)

$$= \gamma^4 - 2\Omega_N^2 \gamma^2 - \Omega_N^4 + \Omega_N^2 \omega^2$$

(C.4)

$$= \gamma^4 + \gamma^2 [-2\Omega_N^2 - (\omega - \frac{\omega_N}{2})^2 - \left( \frac{\omega_N}{2} \right)^2] + \Omega_N^4 + \Omega_N^2 \omega^2$$

(C.5)

$$= \gamma^4 + \gamma^2 \left[ -2\Omega_N^2 - \omega^2 + \omega \omega_N - \frac{\omega_N^2}{2} \right] + \Omega_N^4 + \Omega_N^2 \omega^2$$

(C.6)

$$= \gamma^4 + \gamma^2 \left[ -2\Omega_N^2 + \omega^2 - \omega \omega_N + \frac{\omega_N^2}{2} \right] + \Omega_N^4 + \Omega_N^2 \omega^2$$

(C.7)

$$= \gamma^4 - \gamma^2 \left[ 2\Omega_N^2 + \omega^2 - \omega \omega_N + \frac{\omega_N^2}{2} \right] + \Omega_N^4 - \Omega_N^2 \omega \omega_N + \frac{1}{2} \Omega_N^2 \omega_N^2$$

(C.8)
Using the general quadratic formula,

\[ \gamma^2 = \frac{-b \pm \sqrt{b^2 - 4ac}}{2a} \]  
(C.9)

\[ b^2 = 4\Omega_N^4 + 2\Omega_N^2 \omega^2 - 2\Omega_N^4 \omega \omega_N + \Omega_N^2 \omega_N^2 \]

\[ + \omega^2 2\Omega_N^2 + \omega^4 - \omega^3 \omega_N + \frac{1}{2} \omega^2 \omega_N^2 \]

\[ - \omega \omega_N 2\Omega_N^2 - \omega_N \omega^3 + \omega^2 \omega_N^2 - \frac{1}{2} \omega \omega_N^3 \]

\[ + \omega_N^2 \Omega_N^2 + \frac{1}{2} \omega^2 \omega_N^2 - \frac{1}{2} \omega \omega_N^3 + \frac{1}{4} \omega_N^4 \]

\[ = 4\Omega_N^4 + 4\Omega_N^2 \omega^2 + 2\Omega_N^2 \omega_N^2 + 4\Omega_N^2 \omega \omega_N + \omega^4 - 2\omega^3 \omega_N + 2\omega^2 \omega_N^2 - \omega \omega_N^3 + \frac{1}{4} \omega_N^4 \]  
(C.10)

\[ b^2 - 4ac = 4\Omega_N^4 + 4\Omega_N^2 \omega^2 + 2\Omega_N^2 \omega_N^2 + 4\Omega_N^2 \omega \omega_N + \omega^4 - 2\omega^3 \omega_N + 2\omega^2 \omega_N^2 - \omega \omega_N^3 + \frac{1}{4} \omega_N^4 \]

\[ - 4\Omega_N^4 + 4\Omega_N^2 \omega \omega_N - 2\Omega_N^2 \omega_N^2 - \omega_N^2 \omega^2 + \omega^3 \omega_N - \frac{1}{4} \omega_N^4 \]

\[ = 4\Omega_N^2 \omega^2 + \omega^4 - 2\omega^3 \omega_N + \omega^2 \omega_N^2 \]

\[ = \omega^2 [4\Omega_N^2 + \omega^2 - 2\omega \omega_N + \omega_N^2] \]

\[ = \omega^2 [4\Omega_N^2 + (\omega - \omega_N)^2] \]  
(C.11)

\[ \gamma_{\pm}^2 = \frac{1}{2} [2\Omega_N^2 + \omega^2 - \omega \omega_N + \frac{1}{2} \omega_N^2 \pm \omega \sqrt{4\Omega_N^2 + (\omega - \omega_N)^2}] \]  
(C.12)
Now I let

\[ Q = 2\Omega_N^2 + \omega^2 - \omega\omega_N + \frac{1}{2}\omega_N^2 \]  
\[ R = \sqrt{4\Omega_N^2 + (\omega - \omega_N)^2} \]

So my four equations for \( \gamma \) are

\[ \gamma_1 = \sqrt{\frac{1}{2}(Q + \omega R)} \]  
\[ \gamma_2 = \sqrt{\frac{1}{2}(Q - \omega R)} \]  
\[ \gamma_3 = -\sqrt{\frac{1}{2}(Q + \omega R)} = -\gamma_1 \]  
\[ \gamma_4 = -\sqrt{\frac{1}{2}(Q - \omega R)} = -\gamma_2 \]

The probability for the nuclear spin to flip will be a combination of two possibilities: One that starts in state D (|\( \frac{1}{2}, \frac{1}{2} \rangle \)) and flips to state E (|\( \frac{1}{2}, -\frac{1}{2} \rangle \)) and one that starts in state C (|\( -\frac{1}{2}, -\frac{1}{2} \rangle \)) and flips to state F (|\( -\frac{1}{2}, \frac{1}{2} \rangle \)).

The equations for my coefficients \( E_1, E_2, E_3, \) and \( E_4 \) needed in the first case are the following in this limit.

\[ E_1 + E_2 + E_3 + E_4 = 0 \]  
\[ \gamma_1 E_1 + \gamma_2 E_2 - \gamma_1 E_3 - \gamma_2 E_4 = \Omega_N \]  
\[ \gamma_1^2 E_1 + \gamma_2^2 E_2 + \gamma_1^2 E_3 + \gamma_2^2 E_4 = -\Omega_N\omega \]  
\[ \gamma_1^3 E_1 + \gamma_2^3 E_2 - \gamma_1^3 E_3 - \gamma_2^3 E_4 = \Omega_N[\Omega_N^2 + \omega^2 - \frac{1}{2}\omega\omega_N + \frac{1}{4}\omega_N^2] \]
And the equations for the second case are

\[ F_1 + F_2 + F_3 + F_4 = 0 \] (C.23)
\[ \gamma_1 F_1 + \gamma_2 F_2 - \gamma_1 F_3 - \gamma_2 F_4 = \Omega_N \] (C.24)
\[ \gamma_1^2 F_1 + \gamma_2^2 F_2 + \gamma_1^2 F_3 + \gamma_2^2 F_4 = \Omega_N \omega \] (C.25)
\[ \gamma_1^3 F_1 + \gamma_2^3 F_2 - \gamma_1^3 F_3 - \gamma_2^3 F_4 = \Omega_N [\Omega_N^2 + \omega^2 - \frac{1}{2} \omega \omega_N + \frac{1}{4} \omega_N^2] \] (C.26)

Now I let

\[ X = \Omega_N^2 + \omega^2 - \frac{1}{2} \omega \omega_N + \frac{1}{4} \omega_N^2 \] (C.27)

So my equations are

\[ E_1 + E_2 + E_3 + E_4 = 0 \] (C.28)
\[ \gamma_1 E_1 + \gamma_2 E_2 - \gamma_1 E_3 - \gamma_2 E_4 = \Omega_N \] (C.29)
\[ \gamma_1^2 E_1 + \gamma_2^2 E_2 + \gamma_1^2 E_3 + \gamma_2^2 E_4 = -\Omega_N \omega \] (C.30)
\[ \gamma_1^3 E_1 + \gamma_2^3 E_2 - \gamma_1^3 E_3 - \gamma_2^3 E_4 = \Omega_N X \] (C.31)

\[ F_1 + F_2 + F_3 + F_4 = 0 \] (C.32)
\[ \gamma_1 F_1 + \gamma_2 F_2 - \gamma_1 F_3 - \gamma_2 F_4 = \Omega_N \] (C.33)
\[ \gamma_1^2 F_1 + \gamma_2^2 F_2 + \gamma_1^2 F_3 + \gamma_2^2 F_4 = \Omega_N \omega \] (C.34)
\[ \gamma_1^3 F_1 + \gamma_2^3 F_2 - \gamma_1^3 F_3 - \gamma_2^3 F_4 = \Omega_N X \] (C.35)
Solving the system of equations I now obtain the following.

\[
E_1 = \frac{1 - \Omega_N \omega_1 - \Omega_N \gamma_2^2 + \Omega_N X}{2 (\gamma_1)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \quad (C.36)
\]

\[
E_2 = \frac{1 - \Omega_N \omega_2 - \Omega_N \gamma_1^2 + \Omega_N X}{2 (\gamma_2)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \quad (C.37)
\]

\[
E_3 = \frac{-1 \Omega_N \omega_1 - \Omega_N \gamma_2^2 + \Omega_N X}{2 (\gamma_1)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \quad (C.38)
\]

\[
E_4 = \frac{1 \Omega_N \omega_2 - \Omega_N \gamma_1^2 + \Omega_N X}{2 (\gamma_2)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \quad (C.39)
\]

\[
F_1 = \frac{1 - \Omega_N \omega_1 + \Omega_N \gamma_2^2 - \Omega_N X}{2 (\gamma_1)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \quad (C.40)
\]

\[
F_2 = \frac{-1 \Omega_N \omega_2 + \Omega_N \gamma_1^2 - \Omega_N X}{2 (\gamma_2)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \quad (C.41)
\]

\[
F_3 = \frac{-1 \Omega_N \omega_1 + \Omega_N \gamma_2^2 - \Omega_N X}{2 (\gamma_1)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \quad (C.42)
\]

\[
F_4 = \frac{-1 \Omega_N \omega_2 - \Omega_N \gamma_1^2 - \Omega_N X}{2 (\gamma_2)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \quad (C.43)
\]

Simplifying this a bit and knowing that \(\gamma_1^2 - \gamma_2^2 = \omega R\), I get

\[
E_1 = -\frac{\Omega_N \omega_1 + \gamma_2^2 - X}{2\omega R \gamma_1} \quad (C.44)
\]

\[
E_2 = \frac{\Omega_N \omega_2 + \gamma_1^2 - X}{2\omega R \gamma_2} \quad (C.45)
\]

\[
E_3 = \frac{-\omega_1 + \gamma_2^2 - X}{2\omega R \gamma_1} \quad (C.46)
\]

\[
E_4 = -\frac{-\omega_2 + \gamma_1^2 - X}{2\omega R \gamma_2} \quad (C.47)
\]
\[ F_1 = \frac{-\Omega_N - \omega \gamma_1 + \gamma_2^2 - X}{2\omega R \gamma_1} \quad (\text{C.48}) \]
\[ F_2 = \frac{\Omega_N - \omega \gamma_2 + \gamma_1^2 - X}{2\omega R \gamma_2} \quad (\text{C.49}) \]
\[ F_3 = \frac{\Omega_N \omega \gamma_1 + \gamma_2^2 - X}{2\omega R \gamma_1} \quad (\text{C.50}) \]
\[ F_4 = \frac{-\Omega_N \omega \gamma_2 + \gamma_1^2 - X}{2\omega R \gamma_2} \quad (\text{C.51}) \]

Now

\[ E_1^2 + E_3^2 = F_1^2 + F_3^2 = \frac{1}{2} \left( \frac{\Omega_N}{\omega R} \right)^2 \frac{1}{\gamma_1} \left[ \omega^2 \gamma_1^2 + (\gamma_2^2 - X)^2 \right] \quad (\text{C.52}) \]
\[ E_2^2 + E_4^2 = F_2^2 + F_4^2 = \frac{1}{2} \left( \frac{\Omega_N}{\omega R} \right)^2 \frac{1}{\gamma_2} \left[ \omega^2 \gamma_2^2 + (\gamma_1^2 - X)^2 \right] \quad (\text{C.53}) \]

So the probability is

\[ P_{\text{nuclear}} = E_1^2 + E_2^2 + E_3^2 + E_4^2 + F_1^2 + F_2^2 + F_3^2 + F_4^2 \quad (\text{C.54}) \]
\[ = \left( \frac{\Omega_N}{\omega R} \right)^2 \frac{1}{\gamma_1 \gamma_2} \left[ \omega^2 \gamma_1 \gamma_2 + \gamma_2^2 (\gamma_2 - X)^2 + \omega^2 \gamma_1 \gamma_2 + \gamma_1^2 (\gamma_2 - X)^2 \right] \quad (\text{C.55}) \]
\[ = \left( \frac{\Omega_N}{\omega R} \right)^2 \frac{1}{\gamma_1 \gamma_2} \left[ 2\omega^2 \gamma_1 \gamma_2 + \gamma_2^2 (\gamma_2 - X)^2 + \gamma_1^2 (\gamma_2 - X)^2 \right] \quad (\text{C.56}) \]
\[(\gamma_1^2 - X)^2 = \left(\frac{1}{2} Q + \frac{1}{2} \omega R - X\right)^2 \] (C.57)
\[
= \left(\Omega_N^2 + \frac{1}{2} \omega^2 - \frac{1}{2} \omega N + \frac{1}{4} \omega_N^2 + \frac{1}{2} \omega R - \Omega_N^2 - \omega^2 + \frac{1}{2} \omega \omega_N - \frac{1}{4} \omega_N^2\right)^2
\] (C.58)
\[
= \left(\frac{1}{2} \omega^2 + \frac{1}{2} \omega R\right)^2
\] (C.59)
\[
= \left(\frac{1}{2} \omega (\omega - R)\right)^2
\] (C.60)
\[
(\gamma_1^2 - X)^2 = \frac{1}{4} \omega^2 (\omega + R)^2
\] (C.61)

So now

\[
P_{\text{nuclear}} = \frac{\Omega_N^2}{\omega^2 R^2} \frac{1}{\gamma_1^2 \gamma_2^2} \left[2 \omega^2 \gamma_1^2 \gamma_2^2 + \frac{1}{4} \omega^2 \gamma_2^2 (\omega + R)^2 + \frac{1}{4} \omega^2 \gamma_1^2 (\omega - R)^2\right]
\] (C.63)
\[
= \frac{\Omega_N^2}{R^2} \frac{1}{\gamma_1^2 \gamma_2^2} \left[2 \gamma_1^2 \gamma_2^2 + \frac{1}{4} \gamma_2^2 (\omega + R)^2 + \frac{1}{4} \gamma_1^2 (\omega - R)^2\right]
\] (C.64)
\[
= \frac{\Omega_N^2}{R^2} \frac{1}{\gamma_1^2 \gamma_2^2} \left[2 \gamma_1^2 \gamma_2^2 + \frac{1}{4} (\gamma_1^2 + \gamma_2^2) (\omega^2 + R^2) + \frac{2}{4} (\gamma_2^2 - \gamma_1^2) (\omega R)\right]
\] (C.65)
\[
= \frac{\Omega_N^2}{R^2} \frac{1}{\gamma_1^2 \gamma_2^2} \left[2 \gamma_1^2 \gamma_2^2 + \frac{1}{4} (Q)(\omega^2 + R^2) + \frac{1}{2} (-\omega R)(\omega R)\right]
\] (C.66)
\[
= \frac{\Omega_N^2}{R^2} \frac{1}{\gamma_1^2 \gamma_2^2} \left[2 \gamma_1^2 \gamma_2^2 + \frac{1}{4} (\frac{1}{2} (\omega^2 + R^2))(\omega^2 + R^2) - \frac{1}{2} \omega^2 R^2\right]
\] (C.67)
\[
= \frac{\Omega_N^2}{R^2} \frac{1}{\gamma_1^2 \gamma_2^2} \left[2 \gamma_1^2 \gamma_2^2 + \frac{1}{8} (\omega^4 + R^4 + 2 \omega^2 R^2 - 4 \omega^2 R^2\right]
\] (C.68)
\[
= \frac{\Omega_N^2}{R^2} \frac{1}{\gamma_1^2 \gamma_2^2} \left[2 \gamma_1^2 \gamma_2^2 + \frac{1}{8} (\omega^2 - R^2)^2\right]
\] (C.69)
And we see that

\[
\gamma_1^2 \gamma_2^2 = \frac{1}{4}(Q^2 - \omega^2 R^2) \tag{C.70}
\]

\[
= \frac{1}{4}(\frac{1}{4}(R^4 + 2R^2\omega^2 + \omega^4) - \omega^2 R^2) \tag{C.71}
\]

\[
= \frac{1}{16}(R^4 + \omega^4 - 2R^2\omega^2) \tag{C.72}
\]

\[
= \frac{1}{16}(R^2 - \omega^2)^2 \tag{C.73}
\]

So then

\[
P_{\text{nuclear}} = \Omega_N^2 \frac{1}{R^2 \gamma_1^2 \gamma_2^2} [2\gamma_1^2 \gamma_2^2 + 2\gamma_1 \gamma_2] \tag{C.74}
\]

\[
= \Omega_N^2 \frac{1}{R^2} \tag{C.75}
\]

\[
= \frac{4\Omega_N^2}{4\Omega_N^2 + (\omega - \omega_N)^2} \tag{C.76}
\]

C.2 Electron Zeeman Interaction

The original quartic equation for \( \gamma \) is

\[
0 = [\gamma^2 - (\Omega_N + \Omega_B)^2 - (\omega - \frac{1}{2}(\omega_N + \omega_B))^2][\gamma^2 - \gamma \tilde{D} - (\Omega_B - \Omega_N)^2 - (\frac{1}{2}(\omega_N - \omega_B))^2]
\]

\[
- [(\frac{1}{2}(\omega_N - \omega_B))(\Omega_N + \Omega_B) - (\omega - \frac{1}{2}(\omega_N + \omega_B))(\Omega_B - \Omega_N)]^2 \tag{C.77}
\]
I let $\tilde{D} \to 0$, $\Omega_N \to 0$, and $\omega_N \to 0$. Then the equation for $\gamma$ becomes

\[
0 = (\gamma^2 - \Omega_B^2 - (\omega_B^2/2)\gamma^2 - (\omega - \omega_B^2/2)\gamma^2 - \Omega_B(\omega_B^2/2))^2
\] (C.78)

\[
= (\gamma^2 - \Omega_B^2 - (\omega_B^2/2)\gamma^2 - (\omega - \omega_B^2/2)\gamma^2 - \Omega_B(\omega_B^2/2))^2
\] (C.79)

\[
= (\gamma^2 - \Omega_B^2 - (\omega_B^2/2)\gamma^2 - (\omega - \omega_B^2/2)\gamma^2 - \Omega_B(\omega_B^2/2))^2
\] (C.80)

\[
= \gamma^4 - \gamma^2\Omega_B^2 - \gamma^2(\omega - \omega_B^2/2)^2 - \Omega_B^2\gamma^2 + \Omega_B^4 + \Omega_B^2(\omega - \omega_B^2/2)^2 - (\omega_B^2/2)^2
\] (C.81)

\[
+ (\omega_B^2/2)^2\Omega_B^2 + (\omega_B^2/2)^2(\omega - \omega_B^2/2)^2 - \Omega_B^2\omega^2
\]

\[
= \gamma^4 + \gamma^2[-2\Omega_B^2 - (\omega - \omega_B^2/2)^2 - (\omega_B^2/2)]
\] (C.82)

\[
+ [\Omega_B^4 + \Omega_B^2(\omega - \omega_B^2/2)^2 + (\omega_B^2/2)^2\Omega_B^2 + (\omega_B^2/2)(\omega - \omega_B^2/2)^2 - \Omega_B^2\omega^2]
\]

\[
= \gamma^4 + \gamma^2[-2\Omega_B^2 - \omega^2 + \omega_B - (\omega_B^2/2)]
\] (C.83)

\[
+ [\Omega_B^4 + \Omega_B^2\omega^2 - \Omega_B^2\omega_B + \Omega_B^2\omega_B + \frac{\Omega_B^2\omega_B}{4} + \frac{\Omega_B^2\omega_B}{4} + \frac{\omega_B^2\omega}{4} + \frac{\omega_B^2\omega}{4} + \frac{\omega_B^2\omega}{4} + \frac{\omega_B^2\omega}{4} - \Omega_B^2\omega^2]
\]

\[
= \gamma^4 - \gamma^2[2\Omega_B^2 + \omega^2 - \omega_B + (\omega_B^2/2)]
\] (C.84)

\[
+ [\Omega_B^4 - \Omega_B^2\omega_B + \frac{\Omega_B^2\omega_B}{2} + \frac{\Omega_B^2\omega_B}{4} + \frac{1}{4}\omega_B^2\omega^2 - \frac{1}{4}\omega_B^3\omega + \frac{1}{16}\omega_B^4]
\]

Using the general quadratic formula,

\[
\gamma^2 = \frac{-b \pm \sqrt{b^2 - 4ac}}{2a}
\] (C.85)
\[ b^2 = 4\Omega_B^4 + 2\Omega_B^2\omega^2 - 2\Omega_B^2\omega\omega_B + \Omega_B^2\omega_B^2 \] (C.86)

\[ + \omega^2\Omega_B^2 + \omega^4 - \omega^3\omega_B + \frac{1}{2}\omega^2\omega_B^2 \] (C.87)

\[ - \omega\omega_B\Omega_B^2 - \omega_B\omega^3 + \omega^2\omega_B^2 - \frac{1}{2}\omega\omega_B^3 \] (C.88)

\[ + \omega_B^2\Omega_B^2 + \frac{1}{2}\omega^2\omega_B^2 - \frac{1}{2}\omega\omega_B^3 + \frac{1}{4}\omega^4 \] (C.89)

\[ = 4\Omega_B^4 + 4\Omega_B^2\omega^2 + 2\Omega_B^2\omega_B^2 + 4\Omega_B^2\omega\omega_B + \omega^4 - 2\omega^3\omega_B + 2\omega^2\omega_B^2 - \omega\omega_B^3 + \frac{1}{4}\omega^4 \] (C.90)

\[ b^2 - 4ac = 4\Omega_B^4 + 4\Omega_B^2\omega^2 + 2\Omega_B^2\omega_B^2 + 4\Omega_B^2\omega\omega_B + \omega^4 - 2\omega^3\omega_B + 2\omega^2\omega_B^2 - \omega\omega_B^3 + \frac{1}{4}\omega^4 \] (C.91)

\[ - 4\Omega_B^4 + 4\Omega_B^2\omega\omega_B - 2\Omega_B^2\omega_B^2 - \omega^3\omega_B - \omega_B^3\omega - \frac{1}{4}\omega^4 \] (C.92)

\[ = 4\Omega_B^2\omega^2 + \omega^4 - 2\omega^3\omega_B + \omega^2\omega_B^2 \] (C.93)

\[ = \omega^2[4\Omega_B^2 + \omega^2 - 2\omega\omega_B + \omega_B^2] \] (C.94)

\[ = \omega^2[4\Omega_B^2 + (\omega - \omega_B)^2] \] (C.95)

\[ \gamma_{\pm}^2 = \frac{1}{2}[2\Omega_B^2 + \omega^2 - \omega\omega_B + \frac{1}{2}\omega_B^2 \pm \omega\sqrt{4\Omega_B^2 + (\omega - \omega_B)^2}] \] (C.96)

Now I let

\[ Q = 2\Omega_B^2 + \omega^2 - \omega\omega_B + \frac{1}{2}\omega_B^2 \] (C.97)

\[ R = \sqrt{4\Omega_B^2 + (\omega - \omega_B)^2} \] (C.98)
So my four equations for $\gamma$ are

$$\gamma_1 = \sqrt{\frac{1}{2}(Q + \omega R)}$$  \hspace{1cm} (C.99)

$$\gamma_2 = \sqrt{\frac{1}{2}(Q - \omega R)}$$  \hspace{1cm} (C.100)

$$\gamma_3 = -\sqrt{\frac{1}{2}(Q + \omega R)} = -\gamma_1$$  \hspace{1cm} (C.101)

$$\gamma_4 = -\sqrt{\frac{1}{2}(Q - \omega R)} = -\gamma_2$$  \hspace{1cm} (C.102)

The probability for the electron spin to flip will be a combination of two possibilities: One that starts in state D ($|\frac{1}{2}, \frac{1}{2}\rangle$) and flips to state F ($|\frac{1}{2}, -\frac{1}{2}\rangle$) and one that starts in state C ($|\frac{1}{2}, -\frac{1}{2}\rangle$) and flips to state E ($|\frac{1}{2}, -\frac{1}{2}\rangle$).

The equations for my coefficients $F_1, F_2, F_3,$ and $F_4$ needed in the first case are the following in this limit.

$$F_1 + F_2 + F_3 + F_4 = 0$$  \hspace{1cm} (C.103)

$$\gamma_1 F_1 + \gamma_2 F_2 - \gamma_1 F_3 - \gamma_2 F_4 = \Omega_B$$  \hspace{1cm} (C.104)

$$\gamma_1^2 F_1 + \gamma_2^2 F_2 + \gamma_1^2 F_3 + \gamma_2^2 F_4 = -\Omega_B \omega$$  \hspace{1cm} (C.105)

$$\gamma_1^3 F_1 + \gamma_2^3 F_2 - \gamma_1^3 F_3 - \gamma_2^3 F_4 = \Omega_B [\Omega_B^2 + \omega^2 - \frac{1}{2} \omega \omega_B + \frac{1}{4} \omega_B^2]$$  \hspace{1cm} (C.106)

And the equations for the second case are

$$E_1 + E_2 + E_3 + E_4 = 0$$  \hspace{1cm} (C.107)

$$\gamma_1 E_1 + \gamma_2 E_2 - \gamma_1 E_3 - \gamma_2 E_4 = \Omega_B$$  \hspace{1cm} (C.108)

$$\gamma_1^2 E_1 + \gamma_2^2 E_2 + \gamma_1^2 E_3 + \gamma_2^2 E_4 = \Omega_B \omega$$  \hspace{1cm} (C.109)

$$\gamma_1^3 E_1 + \gamma_2^3 E_2 - \gamma_1^3 E_3 - \gamma_2^3 E_4 = \Omega_B [\Omega_B^2 + \omega^2 - \frac{1}{2} \omega \omega_B + \frac{1}{4} \omega_B^2]$$  \hspace{1cm} (C.110)
Now I let

\[ X = \Omega_B^2 + \omega^2 - \frac{1}{2} \omega \Omega_B + \frac{1}{4} \omega_B^2 \]  

(C.111)

So my equations are

\[ F_1 + F_2 + F_3 + F_4 = 0 \]  

(C.112)

\[ \gamma_1 F_1 + \gamma_2 F_2 - \gamma_1 F_3 - \gamma_2 F_4 = \Omega_B \]  

(C.113)

\[ \gamma_2^2 F_1 + \gamma_2^2 F_2 + \gamma_1^2 F_3 + \gamma_2^2 F_4 = -\Omega_B \omega \]  

(C.114)

\[ \gamma_3^2 F_1 + \gamma_2^2 F_2 - \gamma_1^2 F_3 - \gamma_2^2 F_4 = \Omega_B X \]  

(C.115)

\[ E_1 + E_2 + E_3 + E_4 = 0 \]  

(C.116)

\[ \gamma_1 E_1 + \gamma_2 E_2 - \gamma_1 E_3 - \gamma_2 E_4 = \Omega_B \]  

(C.117)

\[ \gamma_2^2 E_1 + \gamma_1^2 E_2 + \gamma_1^2 E_3 + \gamma_2^2 E_4 = \Omega_B \omega \]  

(C.118)

\[ \gamma_3^2 E_1 + \gamma_2^2 E_2 - \gamma_1^2 E_3 - \gamma_2^2 E_4 = \Omega_B X \]  

(C.119)

Solving the system of equations I now obtain the following.

\[ E_1 = \frac{1}{2} \frac{-\Omega_B \omega \gamma_1 - \Omega_B \gamma_1^2 + \Omega_B X}{(\gamma_1)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \]  

(C.120)

\[ E_2 = \frac{-1}{2} \frac{-\Omega_B \omega \gamma_2 - \Omega_B \gamma_2^2 + \Omega_B X}{(\gamma_2)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \]  

(C.121)

\[ E_3 = \frac{-1}{2} \frac{\Omega_B \omega \gamma_1 - \Omega_B \gamma_1^2 + \Omega_B X}{(\gamma_1)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \]  

(C.122)

\[ E_4 = \frac{1}{2} \frac{\Omega_B \omega \gamma_2 - \Omega_B \gamma_2^2 + \Omega_B X}{(\gamma_2)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \]  

(C.123)
\[ F_1 = \frac{1}{2} - \Omega_B \gamma_2 - \Omega_B X \frac{\Omega_B \gamma_1 + \gamma_2^2}{(\gamma_1)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \]  
(C.124)

\[ F_2 = \frac{1}{2} - \Omega_B \gamma_2 + \Omega_B \gamma_2^2 - \Omega_B X \frac{\Omega_B \gamma_1 + \gamma_2^2}{(\gamma_2)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \]  
(C.125)

\[ F_3 = \frac{1}{2} \Omega_B \omega \gamma_1 + \Omega_B \gamma_2^2 - \Omega_B X \frac{\Omega_B \omega \gamma_1 + \gamma_2^2}{(\gamma_1)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \]  
(C.126)

\[ F_4 = -\frac{1}{2} \Omega_B \omega \gamma_2 + \Omega_B \gamma_2^2 - \Omega_B X \frac{\Omega_B \omega \gamma_2 + \gamma_2^2}{(\gamma_2)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \]  
(C.127)

Simplifying this a bit and knowing that \( \gamma_1^2 - \gamma_2^2 = \omega R \), I get

\[ E_1 = -\frac{\Omega_B}{2 \omega R} \frac{\omega \gamma_1 + \gamma_2^2 - X}{\gamma_1} \]  
(C.128)

\[ E_2 = \frac{\Omega_B}{2 \omega R} \frac{\omega \gamma_2 + \gamma_1^2 - X}{\gamma_2} \]  
(C.129)

\[ E_3 = \frac{\Omega_B}{2 \omega R} \frac{-\omega \gamma_1 + \gamma_2^2 - X}{\gamma_1} \]  
(C.130)

\[ E_4 = -\frac{\Omega_B}{2 \omega R} \frac{-\omega \gamma_2 + \gamma_1^2 - X}{\gamma_2} \]  
(C.131)

\[ F_1 = -\frac{\Omega_B}{2 \omega R} \frac{-\omega \gamma_1 + \gamma_2^2 - X}{\gamma_1} \]  
(C.132)

\[ F_2 = \frac{\Omega_B}{2 \omega R} \frac{-\omega \gamma_2 + \gamma_1^2 - X}{\gamma_2} \]  
(C.133)

\[ F_3 = \frac{\Omega_B}{2 \omega R} \frac{\omega \gamma_1 + \gamma_2^2 - X}{\gamma_1} \]  
(C.134)

\[ F_4 = -\frac{\Omega_B}{2 \omega R} \frac{\omega \gamma_2 + \gamma_1^2 - X}{\gamma_2} \]  
(C.135)

Now

\[ E_1^2 + E_3^2 = F_1^2 + F_3^2 = \frac{1}{2} \left( \frac{\Omega_B}{\omega R} \right)^2 \frac{1}{\gamma_1^2} \left[ \omega^2 \gamma_1^2 + (\gamma_2^2 - X)^2 \right] \]  
(C.136)

\[ E_2^2 + E_4^2 = F_2^2 + F_4^2 = \frac{1}{2} \left( \frac{\Omega_B}{\omega R} \right)^2 \frac{1}{\gamma_2^2} \left[ \omega^2 \gamma_2^2 + (\gamma_1^2 - X)^2 \right] \]  
(C.137)
So the probability is

\[ P_{\text{electron}} = E_1^2 + E_2^2 + E_3^2 + E_4^2 + F_1^2 + F_2^2 + F_3^2 + F_4^2 \]
\[ = \left( \frac{\Omega_B}{\omega R} \right)^2 \frac{1}{\gamma_1^2 \gamma_2^2} \left[ \omega^2 \gamma_1^2 \gamma_2^2 + \gamma_2^2 (\gamma_2 - X)^2 + \omega^2 \gamma_1^2 \gamma_2^2 + \gamma_1^2 (\gamma_1 - X)^2 \right] \]
\[ = \left( \frac{\Omega_B}{\omega R} \right)^2 \frac{1}{\gamma_1^2 \gamma_2^2} \left[ 2 \omega^2 \gamma_1^2 \gamma_2^2 + \gamma_2^2 (\gamma_2 - X)^2 + \gamma_1^2 (\gamma_1 - X)^2 \right] \] (C.138) (C.139) (C.140)

Now

\( (\gamma_1^2 - X)^2 = \left( \frac{1}{2} Q + \frac{1}{2} \omega R - X \right)^2 \) (C.141)
\[ = (\Omega_B^2 + \frac{1}{2} \omega^2 - \frac{1}{2} \omega \omega_B + \frac{1}{4} \omega^2 B - \frac{1}{2} \omega R - \Omega_B^2 - \omega^2 + \frac{1}{2} \omega \omega_B - \frac{1}{4} \omega_B^2)^2 \] (C.142)
\[ = (-\frac{1}{2} \omega^2 + \frac{1}{2} \omega R)^2 \] (C.143)
\[ = (-\frac{1}{2} \omega (\omega - R))^2 \] (C.144)
\[ = \frac{1}{4} \omega^2 (\omega - R)^2 \] (C.145)
\[ (\gamma_2^2 - X)^2 = \frac{1}{4} \omega^2 (\omega + R)^2 \] (C.146)
So now

\[ P_{\text{electron}} = \frac{\Omega_B^2}{\omega^2 R^2} \frac{1}{\gamma_1^2 \gamma_2^2} [2\omega^2 \gamma_1^2 \gamma_2^2 + \frac{1}{4} \omega^2 \gamma_2^2 (\omega + R)^2 + \frac{1}{4} \omega^2 \gamma_1^2 (\omega - R)^2] \]  

(C.147)

\[ = \frac{\Omega_B^2}{R^2} \frac{1}{\gamma_1^2 \gamma_2^2} [2\gamma_1^2 \gamma_2^2 + \frac{1}{4} \gamma_1^2 (\omega + R)^2 + \frac{1}{4} \gamma_2^2 (\omega - R)^2] \]  

(C.148)

\[ = \frac{\Omega_B^2}{R^2} \frac{1}{\gamma_1^2 \gamma_2^2} [2\gamma_1^2 \gamma_2^2 + \frac{1}{4} (\gamma_1^2 + \gamma_2^2)(\omega^2 + R^2) + \frac{1}{2} (-\omega R)(\omega R)] \]  

(C.149)

\[ = \frac{\Omega_B^2}{R^2} \frac{1}{\gamma_1^2 \gamma_2^2} [2\gamma_1^2 \gamma_2^2 + \frac{1}{4} (\frac{1}{2}(\omega^2 + R^2))(\omega^2 + R^2) - \frac{1}{2} \omega^2 R^2] \]  

(C.150)

\[ = \frac{\Omega_B^2}{R^2} \frac{1}{\gamma_1^2 \gamma_2^2} [2\gamma_1^2 \gamma_2^2 + \frac{1}{8} (\omega^4 + R^4 + 2\omega^2 R^2 - 4\omega^2 R^2)] \]  

(C.151)

\[ = \frac{\Omega_B^2}{R^2} \frac{1}{\gamma_1^2 \gamma_2^2} [2\gamma_1^2 \gamma_2^2 + \frac{1}{8} (\omega^2 - R^2)^2] \]  

(C.152)

And we see that

\[ \gamma_1^2 \gamma_2^2 = \frac{1}{4} (Q^2 - \omega^2 R^2) \]  

(C.153)

\[ = \frac{1}{4} \left( \frac{1}{4} (R^4 + 2R^2 \omega^2 + \omega^4) - \omega^2 R^2 \right) \]  

(C.154)

\[ = \frac{1}{16} (R^4 + \omega^4 - 2R^2 \omega^2) \]  

(C.155)

\[ = \frac{1}{16} (R^2 - \omega^2)^2 \]  

(C.156)

So then

\[ P_{\text{electron}} = \frac{\Omega_B^2}{R^2} \frac{1}{\gamma_1^2 \gamma_2^2} [2\gamma_1^2 \gamma_2^2 + 2\gamma_1^2 \gamma_2^2] \]  

(C.157)

\[ = \frac{\Omega_B^2}{R^2} [4] \]  

(C.158)

\[ = \frac{4\Omega_B^2}{R^2 + (\omega - \omega_B)^2} \]  

(C.159)
C.3 Uncoupled Electron Zeeman Interaction and Nuclear Magnetic Interaction

The original quartic equation for $\gamma$ is

$$0 = [\gamma^2 - (\Omega_N + \Omega_B)^2 - (\omega - \frac{1}{2}(\omega_N + \omega_B))^2]\left[\gamma^2 - \gamma \tilde{D} - (\Omega_B - \Omega_N)^2 - \left(\frac{1}{2}(\omega_N - \omega_B)\right)^2\right]$$

$$- \left[(\frac{1}{2}(\omega_N - \omega_B))(\Omega_N + \Omega_B) - (\omega - \frac{1}{2}(\omega_N + \omega_B))(\Omega_B - \Omega_N)\right]^2$$

(C.161)

Letting $\tilde{D} = 0$

$$0 = [\gamma^2 - (\Omega_N + \Omega_B)^2 - (\omega - \frac{1}{2}(\omega_N + \omega_B))^2]\left[\gamma^2 - (\Omega_B - \Omega_N)^2 - \left(\frac{1}{2}(\omega_N - \omega_B)\right)^2\right]$$

$$- \left[(\frac{1}{2}(\omega_N - \omega_B))(\Omega_N + \Omega_B) - (\omega - \frac{1}{2}(\omega_N + \omega_B))(\Omega_B - \Omega_N)\right]^2$$

(C.162)

To simplify let,

$$X = (\Omega_N + \Omega_B)^2 + (\omega - \frac{1}{2}(\omega_N + \omega_B))^2$$

(C.163)

$$Y = (\Omega_B - \Omega_N)^2 + \left(\frac{1}{2}(\omega_N - \omega_B)\right)^2$$

(C.164)

$$Z = [(\frac{1}{2}(\omega_N - \omega_B))(\Omega_N + \Omega_B) - (\omega - \frac{1}{2}(\omega_N + \omega_B))(\Omega_B - \Omega_N)]^2$$

(C.165)

Then

$$0 = [\gamma^2 - X][\gamma^2 - Y] - Z$$

(C.166)

$$0 = \gamma^4 - \gamma(X + Y) + XY - Z$$

(C.167)
\[ \gamma_{\pm} = \frac{-b \pm \sqrt{b^2 - 4ac}}{2a} \]  
\[ = \frac{1}{2} (X + Y \pm \sqrt{X^2 + 2XY + Y^2 - 4XY + 4Z}) \]  
\[ = \frac{1}{2} (X + Y \pm \sqrt{(X - Y)^2 + 4Z}) \]

Let

\[ Q = X + Y \]  
\[ R = \sqrt{(X - Y)^2 + 4Z} \]

Then

\[ \gamma_{\pm} = \frac{1}{2} (Q \pm R) \]

So my four equations for \( \gamma \) are

\[ \gamma_1 = \sqrt{\frac{1}{2} (Q + R)} \]  
\[ \gamma_2 = \sqrt{\frac{1}{2} (Q - R)} \]  
\[ \gamma_3 = -\sqrt{\frac{1}{2} (Q + R)} = -\gamma_1 \]  
\[ \gamma_4 = -\sqrt{\frac{1}{2} (Q - R)} = -\gamma_2 \]

The probability for the nuclear spin to flip will be a combination of two possibilities: One that starts in state D (\(|\frac{1}{2}, \frac{1}{2}\rangle\)) and flips to state E (\(|\frac{1}{2}, -\frac{1}{2}\rangle\)) and one that starts in state C (\(|-\frac{1}{2}, -\frac{1}{2}\rangle\)) and flips to state F (\(|-\frac{1}{2}, \frac{1}{2}\rangle\)).

The equations for my coefficients \( E_1, E_2, E_3, \) and \( E_4 \) needed in the first case are the following
in this limit.

\[ E_1 + E_2 + E_3 + E_4 = 0 \]  
(C.178)

\[ \gamma_1 E_1 + \gamma_2 E_2 - \gamma_1 E_3 - \gamma_2 E_4 = \Omega_N \]  
(C.179)

\[ \gamma_1^2 E_1 + \gamma_2^2 E_2 + \gamma_1^2 E_3 + \gamma_2^2 E_4 = \Omega_N (-\omega + \omega_B) \]  
(C.180)

\[ \gamma_1^3 E_1 + \gamma_2^3 E_2 - \gamma_1^3 E_3 - \gamma_2^3 E_4 = \Omega_N [\Omega_N^2 + 3\Omega_B^2 + \omega^2 - \frac{3}{2}\omega\omega_N - \frac{3}{2}\omega\omega_B \]  
\[ + \frac{1}{4}\omega_N^2 + \frac{3}{2}\omega_B^2] \]  
(C.181)

And the equations for the second case are

\[ F_1 + F_2 + F_3 + F_4 = 0 \]  
(C.182)

\[ \gamma_1 F_1 + \gamma_2 F_2 - \gamma_1 F_3 - \gamma_2 F_4 = \Omega_N \]  
(C.183)

\[ \gamma_1^2 F_1 + \gamma_2^2 F_2 + \gamma_1^2 F_3 + \gamma_2^2 F_4 = -\Omega_N (-\omega + \omega_B) \]  
(C.184)

\[ \gamma_1^3 F_1 + \gamma_2^3 F_2 - \gamma_1^3 F_3 - \gamma_2^3 F_4 = \Omega_N [\Omega_N^2 + 3\Omega_B^2 + \omega^2 - \frac{1}{2}\omega\omega_N \]  
\[ - \frac{3}{2}\omega\omega_B + \frac{1}{4}\omega_N^2 + \frac{3}{4}\omega_B^2] \]  
(C.185)

Now I let

\[ T = \Omega_N^2 + 3\Omega_B^2 + \omega^2 - \frac{1}{2}\omega\omega_N - \frac{3}{2}\omega\omega_B + \frac{1}{4}\omega_N^2 + \frac{3}{4}\omega_B^2 \]  
(C.186)
So my equations are

\[ E_1 + E_2 + E_3 + E_4 = 0 \]  
(C.187)

\[ \gamma_1 E_1 + \gamma_2 E_2 - \gamma_1 E_3 - \gamma_2 E_4 = \Omega_N \]  
(C.188)

\[ \gamma_1^2 E_1 + \gamma_2^2 E_2 + \gamma_1^2 E_3 + \gamma_2^2 E_4 = \Omega_N (-\omega + \omega_B) \]  
(C.189)

\[ \gamma_1^3 E_1 + \gamma_2^3 E_2 - \gamma_1^3 E_3 - \gamma_2^3 E_4 = \Omega_N T \]  
(C.190)

\[ F_1 + F_2 + F_3 + F_4 = 0 \]  
(C.191)

\[ \gamma_1 F_1 + \gamma_2 F_2 - \gamma_1 F_3 - \gamma_2 F_4 = \Omega_N \]  
(C.192)

\[ \gamma_1^2 F_1 + \gamma_2^2 F_2 + \gamma_1^2 F_3 + \gamma_2^2 F_4 = -\Omega_N (-\omega + \omega_B) \]  
(C.193)

\[ \gamma_1^3 F_1 + \gamma_2^3 F_2 - \gamma_1^3 F_3 - \gamma_2^3 F_4 = \Omega_N T \]  
(C.194)

Solving the system of equations I now obtain the following.

\[ E_1 = -\frac{1}{2} \frac{-\Omega_N (-\omega + \omega_B) \gamma_1 + \Omega_N \gamma_2^2 - \Omega_N T}{(\gamma_1)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \]  
(C.195)

\[ E_2 = \frac{1}{2} \frac{-\Omega_N (-\omega + \omega_B) \gamma_2 + \Omega_N \gamma_1^2 - \Omega_N T}{(\gamma_2)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \]  
(C.196)

\[ E_3 = \frac{1}{2} \frac{\Omega_N (-\omega + \omega_B) \gamma_1 + \Omega_N \gamma_2^2 - \Omega_N T}{(\gamma_1)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \]  
(C.197)

\[ E_4 = -\frac{1}{2} \frac{\Omega_N (-\omega + \omega_B) \gamma_2 + \Omega_N \gamma_1^2 - \Omega_N T}{(\gamma_2)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \]  
(C.198)
\[ F_1 = \frac{1}{2} \frac{-\Omega_N(-\omega + \omega_B)\gamma_1 - \Omega_N\gamma_2^2 + \Omega_N T}{(\gamma_1)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \quad (C.199) \]

\[ F_2 = -\frac{1}{2} \frac{-\Omega_N(-\omega + \omega_B)\gamma_2 - \Omega_N\gamma_1^2 + \Omega_N T}{(\gamma_2)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \quad (C.200) \]

\[ F_3 = -\frac{1}{2} \frac{\Omega_N(-\omega + \omega_B)\gamma_1 - \Omega_N\gamma_2^2 + \Omega_N T}{(\gamma_1)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \quad (C.201) \]

\[ F_4 = \frac{1}{2} \frac{\Omega_N(-\omega + \omega_B)\gamma_2 - \Omega_N\gamma_1^2 + \Omega_N T}{(\gamma_2)(\gamma_1 - \gamma_2)(\gamma_1 + \gamma_2)} \quad (C.202) \]

Simplifying this a bit and knowing that \( \gamma_1^2 - \gamma_2^2 = R \)

\[ E_1 = -\frac{\Omega_N}{2R} \frac{-(-\omega + \omega_B)\gamma_1 + \gamma_2^2 - T}{\gamma_1} \quad (C.204) \]

\[ E_2 = \frac{\Omega_N}{2R} \frac{-(-\omega + \omega_B)\gamma_2 + \gamma_1^2 - T}{\gamma_2} \quad (C.205) \]

\[ E_3 = \frac{\Omega_N}{2R} \frac{(-\omega + \omega_B)\gamma_1 + \gamma_2^2 - T}{\gamma_1} \quad (C.206) \]

\[ E_4 = -\frac{\Omega_N}{2R} \frac{(-\omega + \omega_B)\gamma_2 + \gamma_1^2 - T}{\gamma_2} \quad (C.207) \]

\[ F_1 = -\frac{\Omega_N}{2R} \frac{(-\omega + \omega_B)\gamma_1 + \gamma_2^2 - T}{\gamma_1} \quad (C.208) \]

\[ F_2 = \frac{\Omega_N}{2R} \frac{(-\omega + \omega_B)\gamma_2 + \gamma_1^2 - T}{\gamma_2} \quad (C.209) \]

\[ F_3 = \frac{\Omega_N}{2R} \frac{(-\omega + \omega_B)\gamma_1 + \gamma_2^2 - T}{\gamma_1} \quad (C.210) \]

\[ F_4 = -\frac{\Omega_N}{2R} \frac{(-\omega + \omega_B)\gamma_2 + \gamma_1^2 - T}{\gamma_2} \quad (C.211) \]
Now

\[ E_1^2 + E_3^2 = F_1^2 + F_3^2 = \frac{1}{2}(\frac{\Omega_N}{R})^2 1_{\gamma_1^2} [(\omega - \omega_B)^2 \gamma_1^2 + (\gamma_2^2 - T)^2] \] (C.212)

\[ E_2^2 + E_4^2 = F_2^2 + F_4^2 = \frac{1}{2}(\frac{\Omega_N}{R})^2 1_{\gamma_2^2} [(\omega - \omega_B)^2 \gamma_2^2 + (\gamma_1^2 - T)^2] \] (C.213)

\[ P_{B,N} = E_1^2 + E_2^2 + E_3^2 + E_4^2 = F_1^2 + F_2^2 + F_3^2 + F_4^2 \]

\[ = \left(\frac{\Omega_N}{R}\right)^2 \frac{1}{\gamma_1^2 \gamma_2^2} [(\omega - \omega_B)^2 \gamma_1^2 \gamma_2^2 + \gamma_1^2 (\gamma_2^2 - T)^2 + (\omega - \omega_B)^2 \gamma_1^2 \gamma_2^2 + \gamma_1^2 (\gamma_1^2 - T)^2] \] (C.214)

\[ = \left(\frac{\Omega_N}{R}\right)^2 \frac{1}{\gamma_1^2 \gamma_2^2} [2(\omega - \omega_B)^2 \gamma_1^2 \gamma_2^2 + \gamma_1^2 \gamma_2^2 + \gamma_1^2 (\gamma_2^2 - T)^2] \] (C.215)

\[ = \left(\frac{\Omega_N}{R}\right)^2 \frac{1}{\gamma_1^2 \gamma_2^2} [2(\omega - \omega_B)^2 \gamma_1^2 \gamma_2^2 + \gamma_1^2 (\gamma_1^2 - T)^2 - \frac{3}{4} \omega_B^2] \] (C.216)

Now

\[ \gamma_1^2 - T = \frac{1}{2} Q + \frac{1}{2} R - T \] (C.217)

\[ = \frac{1}{2} R + \frac{1}{2} (X + Y) - T \] (C.218)

\[ = \frac{1}{2} R + \Omega_N^2 + \Omega_B^2 + \frac{1}{2} \omega_N^2 - \frac{1}{2} \omega \omega_N - \frac{1}{2} \omega \omega_B + \frac{1}{4} \omega_N^2 + \frac{1}{4} \omega_B^2 \]

\[ - \Omega_N^2 - 3 \Omega_B^2 - \omega_N^2 + \frac{1}{2} \omega \omega_N + \frac{3}{2} \omega \omega_B - \frac{1}{4} \omega_N^2 - \frac{3}{4} \omega_B^2 \] (C.219)

\[ = \frac{1}{2} R - 2 \Omega_B^2 - \frac{1}{2} \omega^2 + \omega \omega_B - \frac{1}{2} \omega_B^2 \] (C.220)

\[ = \frac{1}{2} (R - 4 \Omega_B^2 - \omega^2 + 2 \omega \omega_B - \omega_B^2) \] (C.221)

\[ = \frac{1}{2} (R - W) \] (C.222)

\[ \gamma_2^2 - T = \frac{1}{2} Q - \frac{1}{2} R - T \] (C.223)

\[ = \frac{1}{2} (-R - W) \] (C.224)
Where

\[ W = 4\Omega^2_B + \omega^2 - 2\omega\omega_B + \omega^2_B \]  
\[ = 4\Omega^2_B + (\omega - \omega_B)^2 \]  
(C.225)  
(C.226)

So now

\[ P_{B,N} = \frac{\Omega^2_N}{R^2} \frac{1}{\gamma_1 \gamma_2} \left[ 2(\omega - \omega_B)^2 \gamma_1^2 \gamma_2^2 + \frac{1}{4} \gamma_2^2 (R + W)^2 + \frac{1}{4} \gamma_1^2 (R - W)^2 \right] \] 
(C.227)  
\[ = \frac{\Omega^2_N}{R^2} \frac{1}{\gamma_1 \gamma_2} \left[ 2(\omega - \omega_B)^2 \gamma_1^2 \gamma_2^2 + \frac{1}{4} (\gamma_1^2 + \gamma_2^2)(R^2 + W^2) + \frac{2}{4} (\gamma_2^2 - \gamma_1^2)(RW) \right] \] 
(C.228)  
\[ = \frac{\Omega^2_N}{R^2} \frac{1}{\gamma_1 \gamma_2} \left[ 2(\omega - \omega_B)^2 \gamma_1^2 \gamma_2^2 + \frac{1}{4} (Q)(R^2 + W^2) + \frac{1}{2} (-R)(RW) \right] \] 
(C.229)  
\[ = \frac{\Omega^2_N}{R^2} \frac{4}{(Q^2 - R^2)} \left[ 2(\omega - \omega_B)^2 \frac{1}{4}(Q^2 - R^2) + \frac{1}{4} QR^2 + \frac{1}{4} QW^2 - \frac{1}{2} R^2 W \right] \] 
(C.230)  
\[ = \frac{\Omega^2_N}{R^2} \frac{2}{(Q^2 - R^2)} \left[ (\omega - \omega_B)^2 (Q^2 - R^2) + \frac{1}{2} QR^2 + \frac{1}{2} QW^2 - R^2 W \right] \] 
(C.231)

And we see that

\[ Q = X + Y \] 
(C.232)  
\[ = (\Omega_N + \Omega_B)^2 + \left( \omega - \frac{1}{2}(\omega_N + \omega_B) \right)^2 + (\Omega_B - \Omega_N)^2 + \left( \frac{1}{2}(\omega_N - \omega_B) \right)^2 \] 
(C.233)  
\[ = 2\Omega^2_B + 2\Omega^2_N + \frac{1}{2}(\omega - \omega_N)^2 + \frac{1}{2}(\omega - \omega_B)^2 \] 
(C.234)  
\[ = \frac{1}{2} \left[ (4\Omega^2_B + (\omega - \omega_B)^2) + (4\Omega^2_N + (\omega - \omega_N)^2) \right] \] 
(C.235)
\[ R^2 = (X - Y)^2 + 4Z \]  
\[ = [(4\Omega_B\Omega_N + (\omega - \omega_N)(\omega - \omega_B)]^2 + 4[\Omega_N(\omega - \omega_B) - \Omega_B(\omega - \omega_N)]^2 \]  
\[ = 16\Omega_B^2\Omega_N^2 + (\omega - \omega_N)^2(\omega - \omega_B)^2 + 4\Omega_N^2(\omega - \omega_B)^2 + 4\Omega_B^2(\omega - \omega_N)^2 \]  
\[ = (\omega - \omega_N)^2(4\Omega_B^2 + \frac{1}{2}(\omega - \omega_B)^2) + (\omega - \omega_B)^2(4\Omega_N^2 + \frac{1}{2}(\omega - \omega_N)^2) + 16\Omega_B^2\Omega_N^2 \]  
\[ + 16\Omega_B^2\Omega_N^2 - (\omega - \omega_N)^2(\omega - \omega_B)^2 \]  

Now let

\[ B = 4\Omega_B^2 + (\omega - \omega_B)^2 \]  
\[ N = 4\Omega_N^2 + (\omega - \omega_N)^2 \]  
\[ b = (\omega - \omega_B)^2 \]  
\[ n = (\omega - \omega_N)^2 \]  

Then

\[ Q = \frac{1}{2}(B + N) \]  
\[ R^2 = nB + bN + 16\Omega_B^2\Omega_N^2 - nb \]  
\[ W = B = 2Q - N \]
Then

\[ \frac{1}{2}QR^2 + \frac{1}{2}QW^2 - R^2W = \frac{1}{2}QR^2 + \frac{1}{2}Q(4Q^2 - 4QN + N^2) - R^2(2Q - N) \]  
\[ = \frac{1}{2}QR^2 + 2Q^3 - 2Q^2N + \frac{1}{2}QN^2 - 2R^2Q + R^2N \]  
\[ = 2Q(Q^2 - R^2) + \frac{1}{2}QR^2 - N(Q^2 - R^2) - NQ^2 - \frac{1}{2}QN^2 \]  
\[ = (2Q - N)(Q^2 - R^2) + \frac{1}{2}Q(R^2 - NQ + N^2) \]  
\[ = (B)(Q^2 - R^2) + \frac{1}{4}(B + N)(R^2 - 2N\left(\frac{1}{2}\right)(B + N) + N^2) \]  
\[ = B(Q^2 - R^2) + \frac{1}{4}(B + N)(R^2 - NB) \]  

\[ NB = (4\Omega_N + (\omega - \omega_N)^2)(4\Omega_B + (\omega - \omega_B)^2) \]  
\[ = (4\Omega_N + n)(4\Omega_B + b) \]  
\[ = 16\Omega_B^2\Omega_N^2 + 4\Omega_B^2n + 4\Omega_N^2b + nb \]  
\[ R^2 - NB = nB + bN - nb + 16\Omega_B^2\Omega_N^2 - 16\Omega_B^2\Omega_N^2 - 4\Omega_B^2n - 4\Omega_N^2b - nb \]  
\[ = n(4\Omega_B^2 + b) + b(\Omega_N^2 + n) - 2nb - 4\Omega_B^2n - 4\Omega_N^2b \]  
\[ = 0 \]  

Then

\[ \frac{1}{2}QR^2 + \frac{1}{2}QW^2 - R^2W = B(Q^2 - R^2) + 0 \]
And

\[ P_{B,N} = \frac{\Omega_N^2}{R^2} \frac{2}{(Q^2 - R^2)} [(\omega - \omega_B)^2(Q^2 - R^2) + \frac{1}{2} QR^2 + \frac{1}{2} QW^2 - R^2W] \]  (C.261)

\[ = \frac{\Omega_N^2}{R^2} \frac{2}{(Q^2 - R^2)} [(\omega - \omega_B)^2(Q^2 - R^2) + B(Q^2 - R^2)] \]  (C.262)

\[ = \frac{2\Omega_N^2}{R^2} [(\omega - \omega_B)^2 + B] \]  (C.263)

\[ = \frac{2\Omega_N^2}{R^2} [(\omega - \omega_B)^2 + 4\Omega_B^2 + (\omega - \omega_B)^2] \]  (C.264)

\[ = \frac{4\Omega_N^2}{R^2} [2\Omega_B^2 + (\omega - \omega_B)^2] \]  (C.265)

So finally

\[ P_{B,N} = \frac{4\Omega_N^2 [2\Omega_B + (\omega - \omega_B)^2]}{(\omega - \omega_N)^2(4\Omega_B^2 + \frac{1}{2}(\omega - \omega_B)^2) + (\omega - \omega_B)^2(4\Omega_N^2 + \frac{1}{2}(\omega - \omega_N)^2) + 16\Omega_B^2 \Omega_N^2} \]  (C.266)
APPENDIX D: Second Quantization
It is useful to write the nuclear Green function in matrix form,

\[
\hat{G}^n_{i,i}(\omega) \equiv \begin{pmatrix}
\tilde{G}^n_{i,i;\uparrow\uparrow}(\omega) & \tilde{G}^n_{i,i;\uparrow\downarrow}(\omega) \\
\tilde{G}^n_{i,i;\downarrow\uparrow}(\omega) & \tilde{G}^n_{i,i;\downarrow\downarrow}(\omega)
\end{pmatrix},
\] (D.1)

Then, the bare \(\hat{G}^{n,0}_{i,i}(\omega)\) may be written as

\[
\hat{G}^{n,0}_{i,i}(\omega) = \frac{1}{\omega^2 - R_n^2} \begin{pmatrix}
\omega + (\omega_0 - \omega_n)/2 & -\Omega_n/2 \\
-\Omega_n/2 & \omega - (\omega_0 - \omega_n)/2
\end{pmatrix},
\] (D.2)

which is immediately seen to satisfy

\[
\hat{G}^{n,0}_{i,i}(\omega) = \frac{1}{\omega^2 - R_n^2} \left[\omega\sigma_0 + \sigma_3(\omega_0 - \omega_n)/2 - \sigma_1\Omega_n/2\right].
\] (D.3)

where \(\sigma_0\) is the rank-2 identity matrix and \(\sigma_1\) and \(\sigma_3\) are standard Pauli matrices. Then, it is easy to write the Dyson equation in this matrix form,

\[
[\hat{G}^n_{i,i}(\omega)]^{-1} = [\hat{G}^{n,0}_{i,i}(\omega)]^{-1} - \hat{\Sigma}_{i,i}(\omega),
\] (D.4)

so we need to find the form for the inverse of the bare nuclear Green function. Using the standard techniques common in superconductivity theory, we may write

\[
[\hat{G}^{n,0}_{i,i}(\omega)]^{-1} = \omega\sigma_0 - \sigma_3(\omega_0 - \omega_n)/2 + \sigma_1\Omega_n/2,
\] (D.5)
which leads to

\[
\hat{g}^{m,0}_{i,i}(\omega) = \frac{1}{\omega \sigma_0 - \sigma_3(\omega_0 - \omega_n)/2 + \sigma_1 \Omega_n/2} \left( \frac{\omega \sigma_0 + \sigma_3(\omega_0 - \omega_n)/2 - \sigma_1 \Omega_n/2}{\omega \sigma_0 + \sigma_3(\omega_0 - \omega_n)/2 - \sigma_1 \Omega_n/2} \right)
\]

\[
= \frac{\omega \sigma_0 + \sigma_3(\omega_0 - \omega_n)/2 - \sigma_1 \Omega_n/2}{(\omega^2 - R^2_n)\sigma_0} \hat{g}^{m,0}_{i,i}(\omega),
\]

since the anticommutator of the Pauli matrices \(\sigma_i\) satisfies \(\{\sigma_i, \sigma_j\} = 2\delta_{i,j}\).

The sums over \(\sigma''\) corresponding to the five Feynman diagrams pictured in Figs. 1 and 2 can
then be readily evaluated. From Eq. (17), one obtains

\[
\sum_{\sigma''} \tilde{G}_{i,i',\sigma''}(\omega) \tilde{G}_{i,i',-\sigma''}(\omega) = \frac{1}{(\omega^2 - R_n^2)^2} \\
\times \left[ -\delta_{\sigma,\sigma'} \sigma_n [\omega + \sigma(\omega_0 - \omega_n)/2] \\
+ \delta_{\sigma',-\sigma} [\omega^2 - R_n^2 + 2(\Omega_n/2)^2] \right], \quad (D.9)
\]

\[
= 2\delta_{\sigma,\sigma'} \tilde{G}_{i,i',\sigma,-\sigma}(\omega) \tilde{G}_{i,i',\sigma,\sigma}(\omega) \\
+ \delta_{\sigma,-\sigma'} \left[ \tilde{G}_{i,i',\sigma}(\omega) \tilde{G}_{i,i',-\sigma}(\omega) + \left( \tilde{G}_{i,i',\sigma,-\sigma}(\omega) \right)^2 \right], \quad (D.10)
\]

\[
= \tilde{G}_{i,i}(\omega) \sigma_{1} \tilde{G}_{i,i}(\omega), \quad (D.11)
\]

\[
\sum_{\sigma''} \sigma'' \tilde{G}_{i,i,\sigma''}(\omega) \tilde{G}_{i,i',-\sigma''}(\omega) = \frac{1}{(\omega^2 - R_n^2)^2} \\
\times \left[ \delta_{\sigma,\sigma'} \left( \sigma_3 + \sigma_2 \left( \tilde{G}_{i,i,\sigma}(\omega)^2 - \tilde{G}_{i,i,\sigma,\sigma}(\omega)^2 \right) \right) \\
- \delta_{\sigma,-\sigma'} (\omega_0 - \omega_n) \Omega_n/2 \right], \quad (D.12)
\]

\[
= \delta_{\sigma,\sigma'} \left[ \left( \tilde{G}_{i,i,\sigma}(\omega)^2 - \tilde{G}_{i,i,\sigma,\sigma}(\omega)^2 \right) \right] \\
+ \delta_{\sigma,-\sigma'} \sigma_3 \tilde{G}_{i,i,\sigma}(\omega) \left( \tilde{G}_{i,i,\sigma,\sigma}(\omega) - \tilde{G}_{i,i,\sigma,\sigma}(\omega) \right) \quad (D.13)
\]

\[
= \tilde{G}_{i,i}(\omega) \sigma_{3} \tilde{G}_{i,i}(\omega). \quad (D.14)
\]

The Klemm-Clem transformations also must be applied for each conduction band to \(\omega_{j,e}'\) and \(\Omega_{j,e}'\). For a diagonal \(g_j\) tensor with elements \(g_{j,xx}, g_{j,yy},\) and \(g_{j,zz}\), for example,

\[
\omega_{j,e}' = B_0 [g_{j,xx}^2 \sin^2 \theta \cos^2 \phi + g_{j,yy}^2 \sin^2 \theta \sin^2 \phi + g_{j,zz}^2 \cos^2 \phi]^{1/2} \quad (D.15)
\]
transforms to

\[ \tilde{\omega}'_{j,e} = B_0 \beta_j(\theta, \phi) \]

\[ = B_0 \left[ g_{j,xx}^2 m_{j,1} \sin^2 \theta \cos^2 \phi + g_{j,yy}^2 m_{j,2} \sin^2 \theta \sin^2 \phi + g_{j,zz}^2 m_{j,3} \cos^2 \theta \right]^{1/2}. \]  

(D.16)

As for the oscillatory field term \( \Omega'_j,e \), we note that for \( B_0 = B_0 \hat{r} \) as assumed, \( B_1 \) is in the plane normal to \( B_0 \), and oscillates in time. Even for the simplest case \( B_0 = B_0 \hat{z} \), \( B_1 \) has components in the \( \hat{x} \) and \( \hat{y} \) directions. For a general \( B_0 = B_0 \hat{r} \) direction, in our model, we have \( B_1(t) = B_1[\dot{\phi} \cos(\omega_0 t) - \dot{\theta} \sin(\omega_0 t)] \). Therefore, our best “time-independent” estimate of \( \Omega'_j,e \) is to take the root-mean-square (RMS) time average over an integral number of periods of \( g_j \cdot B_1(t) \),

\[ \Omega'_j,e \approx \sqrt{\left[ g_j \cdot B_1(t) \right]^2} \]

\[ = B_1 \left[ g_{j,xx}^2 \cos^2 \theta \cos^2 \phi + \sin^2 \phi + g_{j,yy}^2 \cos^2 \theta \sin^2 \phi + \cos^2 \phi \right]^{1/2}, \]  

(D.18)

leading to the RMS value of the transformed \( \tilde{\Omega}'_{j,e} \),

\[ \tilde{\Omega}'_{j,e} = B_1 \gamma_j(\theta, \phi) \]

\[ = B_1 \left[ g_{j,xx}^2 (m_{j,2} \sin^2 \phi + m_{j,3} \cos^2 \theta) + g_{j,yy}^2 (m_{j,1} \sin^2 \phi + m_{j,3} \cos^2 \theta) \right]^{1/2}. \]  

(D.20)

The integrals over \( \tilde{k}_{j,||} \) and sums over the Landau level numbers \( n_j \) need to be examined.
more carefully. We define

\[ I_{\sigma''', \pm \sigma''}^{ce} = \frac{eB_0 \alpha_j}{2\pi} \sum_{n_j=0}^{\infty} \alpha_j \int_{-\infty}^{\infty} \frac{dk_j}{2\pi} G_{ce,0}(k_j, n_j, \omega') \]

\[ = \frac{eB_0 \alpha_j}{2\pi} \sum_{n_j=0}^{\infty} \alpha_j \int_{-\infty}^{\infty} \frac{dk_j}{2\pi} \sum_{\sigma=\pm} \omega' - \varepsilon_j(k_j, n_j) + \mu + \tilde{\sigma} R_{j,e} + i\delta_{\tilde{\sigma}}, \]

\[ \delta_{\tilde{\sigma}} = \delta \text{ sgn}[\varepsilon_j(k_j, n_j) - \mu - \tilde{\sigma} R_{j,e}], \]

\[ A_{-\tilde{\sigma}, \sigma'''}' = \frac{\tilde{\sigma} \Omega_j}{4 R_{j,e}}, \]

\[ A_{+\tilde{\sigma}, \sigma''} = \frac{2 R_{j,e} - \tilde{\sigma} \sigma'''(\omega_0 - \tilde{\omega}_{j,e})}{4 R_{j,e}}. \]

We first evaluate the \( k_j \) integral. Using the Cauchy integral theorem, the integral contains a pole in the lower half plane only when \( \delta_{\tilde{\sigma}} > 0 \). Thus, we have

\[ \int_{-\infty}^{\infty} \frac{dk_j}{2\pi} \frac{1}{\omega' - \varepsilon_j(k_j, n_j) + \mu + \tilde{\sigma} R_{j,e} + i\delta_{\tilde{\sigma}}} = -i N_{j,\tilde{\sigma}} \]

\[ \times \Theta \left[ \mu + \tilde{\sigma} R_{j,e} - eB_0 \alpha_j(n_j + 1/2)/m_j \right], \]

\[ N_{j,\tilde{\sigma}} = \sqrt{\frac{m_j}{2[\mu + \tilde{\sigma} R_{j,e} - eB_0 \alpha_j(n_j + 1/2)/m_j]}} \]

(D.27)
is the one-dimensional density of states for spin $\tilde{\sigma}$. Then we have

$$I_{\sigma''',\pm,\sigma'''}^{ce} = -i\frac{eB_0}{2\pi}\alpha_j^2 \sum_{\tilde{\sigma}} \sum_{n_j=0}^{n_{j,\max_{\tilde{\sigma}}}} N_{j,\tilde{\sigma}} A_{\tilde{\sigma},\sigma'''}^\pm,$$

$$n_{j,\max_{\tilde{\sigma}}} = \frac{m_j(\mu + \tilde{\sigma} R'_{j,e})}{eB_0\alpha_j} - \frac{1}{2},$$

where the $A_{\tilde{\sigma},\sigma'''}^\pm$ are given in Eqs. (83) and (84), respectively.

The integrals over $\omega'$ in diagrams 1b, 2a, and 2b can be evaluated exactly. We let

$$a = U_q \langle \hat{n}_{i,q,\sigma'''} \rangle,$$

$$c = \sigma''\left(\omega_0 - \omega_e\right)/2,$$

$$r = R_e.$$

We let

$$I_{1b}^{le}(\omega''') = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \tilde{G}_{i,i,q;\sigma''',\sigma'''}^{de,0}(\omega') \tilde{G}_{i,i,q,\sigma'''}^{de,0}(\omega'),$$

$$I_{2a}^{le}(\omega''') = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} [\tilde{G}_{i,i,q,\sigma''',\sigma'''}^{de,0}(\omega')]^2,$$

$$I_{2b}^{le}(\omega''') = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \tilde{G}_{i,i,q,\sigma''',\sigma'''}^{de,0}(\omega') \tilde{G}_{i,i,q,\sigma''',\sigma'''}^{de,0}(\omega'),$$

$$I_{2c}^{le}(\omega''') = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \tilde{G}_{i,i,q,\sigma''',\sigma'''}^{de,0}(\omega') \tilde{G}_{i,i,q,\sigma''',\sigma'''}^{de,0}(\omega').$$
With the aid of symbolic programming software, we then find that

\[
I_{1b}^{le}(\sigma''') = \frac{i \Omega_e}{4} \frac{c + a - r}{r(c + 2a - r)},
\]

\[
I_{2a}^{le}(\sigma''') = \frac{-i (c + r)(3a - c + r)}{4r^3(c - 2a - r)^3} \times \left[ 6a^2c - 5ac^2 + c^3 - 6a^2r + 12acr - 3c^2r - 3ar^2 + 3cr^2 - r^3 \right],
\]

\[
I_{2b}^{le}(\sigma''') = \frac{-i \Omega_e(3a - c + r)}{8r^3(r - c + 2a)^3} \times \left[ 2ar^2 + 6a^2c + 7arc - 5ac^2 + c^3 - 2c^2r + cr^2 \right],
\]

\[
I_{2c}^{le}(\sigma''') = \frac{i}{4(b_1 - r)^2(b_2 - r)^2r^3} \times \left\{ \left( -(b_1 - r)^2[(b_2 - r)^2(c^2 + r^2) - a_2(-2c^2r + b_2(c^2 + r^2))] \right) \\
+ a_1 \left[ (b_2 - r)^2[-2c^2r + b_1(c^2 + r^2)] \right] \\
+ a_2 \left[ -(b_1[-2c^2r + b_2(c^2 + r^2)] + r[2b_2c^2 + r(-3c^2 + r^2)]) \right] \right\},
\]

where

\[
a_{1,2} = U_q \langle \hat{n}_{i,q,\pm\sigma'''} \rangle,
\]

\[
b_{1,2} = (\pm) \sigma'''(\omega_0 - \omega_e)/2 + 2a_{1,2}.
\]
In the limits that $U_q \langle \tilde{n}_{i,q,-\sigma} \rangle \to 0, \infty$, we have

\begin{align*}
I_{1b}^{le}(\sigma''') & \to iK_{1b} \frac{\Omega_e}{R_e}, \\
I_{2a}^{le}(\sigma''') & \to -iK_{2a} \frac{\Omega_e^2}{R_e^2}, \\
I_{2b}^{le}(\sigma''') & \to -iK_{2b} \frac{\sigma'''(\omega_0 - \omega_e)\Omega_e}{R_e^2}, \\
I_{2c}^{le}(\sigma''') & \to -iK_{2c} \left[ \frac{R_e^2 + (\omega_0 - \omega_e)^2/4}{R_e^3} \right],
\end{align*}

(D.44) - (D.47)

where $K_{1b} = \frac{1}{4}$, $K_{2a} = K_{2b} = \frac{1}{16}$ and $K_{2c} = \frac{1}{4}$ in the $U_q \to 0$ limit, and $K_{1b} = \frac{1}{8}$, $K_{2b} = K_{2a} = \frac{9}{64}$, and $K_{2c} = \frac{1}{16}$ as $U_q \to \infty$. After the overall $\sigma'''$ summations, $I_{1b}^{le}$, $I_{2a}^{le}$, $I_{2b}^{le}$, and $I_{2c}^{le}$ are all finite. Moreover, since both the $\tilde{k}_{j,||}$ and $\omega'$ integrals give rise to pure imaginary factors, the overall contributions to the Knight shift and to its associated linewidth changes are real.
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