Non-Perturbative Bounds on Hyperfine-Induced Electron Spin Coherence Times

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We address the decoherence of a localized electron spin in an external magnetic field due to the hyperfine interaction with a lattice of nuclear spins. Using a completely non-perturbative method, rigorous bounds on the $T_1$ and $T_2$ coherence times for the electron spin are provided. It is shown that for magnetic fields $B$ greater than some critical field $B_c$ ($B_c \approx 0.001 - 2$ Tesla for the systems studied here), the $z$-polarization of the electron spin cannot relax, and hence $T_1$ is infinite. However, even at high fields dephasing can still occur. We provide a lower bound on the $T_2$ coherence time that explicitly takes into account the effects of a spin echo pulse sequence.

I. INTRODUCTION

Isolated electron spin systems are of great interest because of their potential application as coherent quantum memory in novel solid state devices[1, 2]. Several quantum computation proposals make use of electron spins, either as the qubits themselves or as intrinsic components of one- and two-qubit gates [3–6]. Furthermore, experimental efforts are just now succeeding at measuring single electron spin relaxation times [7–9]. Unfortunately, the exponential size of the Hilbert space of these spin systems and the variety of channels for decoherence makes an exact analysis of spin dynamics virtually impossible (without the use of a quantum computer)[10–13]. As a result, it is often necessary to resort to approximations or perturbative treatments to address the timescale of electron spin decoherence, even using a simplified Hamiltonian. Because approximations are inevitable, it is important to know which of these can be rigorously justified. In particular, it is necessary to know which terms in a very complex system Hamiltonian can be ignored because they do not contribute to decoherence on the experimentally relevant timescale. In this paper, we consider electron spin decoherence induced by the nuclear spins. In particular, we focus on the relevance of the $S_+ I_- + S_- I_+$ term in the Hamiltonian (see Eq. (1)), which comes from the contact hyperfine coupling of the electron and its surrounding nuclei.

Previously, the effect of the hyperfine interaction on decoherence has been studied in special cases or using various approximation methods. Recently, Erlingsson et. al used a semi-classical approximation to study electron spin correlation functions at low fields ($B \ll B_c$, where $B_c$ is on the order of the Overhauser field, see below) [14]. They found that the correlation functions were characterized by complex dynamics which did not decay, even at long times. Work by Khaetskii et al. has characterized the timescale of the longitudinal spin correlation for an electron coupled to surrounding nuclei via the hyperfine interaction [10, 15]. In refs. [10, 15], exact solutions for the case of completely polarized nuclei and for the case of homogeneous coupling, as well as perturbative solutions for the general, inhomogeneous situation were presented. From these solutions, the behavior of the general unpolarized, inhomogeneous case was inferred. Their results indicate that the longitudinal spin correlation time is on the order of $hN/A$ for a single quantum dot and on the order of $h\sqrt{N}/A$ for an ensemble of dots. Gate imperfections due to the hyperfine coupling were also treated using perturbation theory in [16]. However, it was noted in [15] that the perturbative solution would diverge to second-order and was therefore potentially unreliable. More recently, Coish et al. have treated the behavior of this system using a generalized master equation, again confirming a power-law decay for short timescales on the order of $hN/A$ [17]. In general, though, all treatments based on a time-dependent perturbation expansion will be valid only at times smaller than some maximum timescale specified by the order of the expansion[26]. Although such treatments are useful for examining short-timescale and small-magnitude fluctuations in electron polarization, in this paper we are interested in the long-timescale dynamics of the electron spin which leads to $1/e$ decay of the longitudinal or in-plane polarization of the electron. Note that we are not claiming any particular functional form for the decay of coherence, exponential or otherwise. Rather, we are evaluating the timescale on which the longitudinal and in-plane magnetization decays to a constant fraction (in this case $1/e$) of its initial value. Moreover, we focus on the high magnetic field regime which is relevant for the recent experimental developments in single spin read-out ($B = 8 – 14$ Tesla in ref. [9]).

In ref. [11] it was assumed that at sufficiently high magnetic fields the electron-nuclei flip-flop term ($S_+ I_- + S_- I_+$ in Eq. (1)) which is due to the hyperfine interaction can be neglected as a direct source for electron spin decoherence. In this paper, we would like to 1) justify this assumption and 2) determine how strong a magnetic field must be applied to safely neglect this term. Rather than resort to approximation to estimate the numerical value of $T_1$ and $T_2$, this paper will instead place rigorous, non-perturbative lower bounds on the spin relaxation time of the electron in the high-field regime ($B > B_c$). Furthermore, we will explicitly take into account the application of the spin echo
sequence which is often used experimentally to remove inhomogeneous broadening. To determine the importance of the electron-nuclear flip-flop term, we use the Hamiltonian presented in [10, 15, 17] which contains only the contact hyperfine term. In doing so, we can effectively isolate the decoherence contribution that derives exclusively from the hyperfine interaction. When the timescale of this hyperfine-induced decoherence is significantly greater than the timescale of decoherence due to other mechanisms (such as spectral diffusion induced by inter-nuclear dipolar coupling[11]) then it seems reasonable to ignore the direct effects of hyperfine coupling as a channel for electron spin decoherence.

It is important to define what we mean by “electron spin decoherence”. Typically, electron spins are assumed to undergo simple exponential decay governed by the Bloch equations. Thus the coherence times $T_1$ and $T_2$ can be defined as the timescales of this exponential decay. However, when decay is non-exponential, as in this system (see [10]), there is some flexibility in the definition of the coherence times. Electron spin dynamics includes both short-timescale fluctuations (which are generally small in magnitude [17]) and long-timescale (i.e., $1/e$) decay; in this paper, we take “electron spin decoherence” to mean the latter. In other words, the coherence time of an electron is defined as the time it takes for the longitudinal or transverse polarization of an initially polarized electron spin to decay to a specific fraction, e.g. $1/e$, of its initial value.

The decay of the in-plane magnetization is understood to occur in the context of a spin echo experiment. Spin echo experiments are designed to remove the effects of inhomogeneous broadening due to variations in the local field experienced by an ensemble of electron-nuclei systems (i.e. multiple independent systems). When applied to a single electron spin, a spin echo can be understood as removing the inhomogeneous broadening resulting from an ensemble of initial states of the single electron-nuclei system. The remaining decay of the in-plane magnetization comes from fluctuating Overhauser fields, which cannot in general be removed by the spin echo (but see [18] for numerical evidence that a substantial part of hyperfine-induced fluctuations may, in fact, be reversible). Because only bounds on relaxation behavior are desired, our analysis of this decay can be completely rigorous, despite the complexity of the problem and the size of the Hilbert space. Our results are applicable to localized electron spins in semiconductors such as quantum dots and donor impurities.

This paper is organized as follows: Sec. II outlines the general structure of the problem and presents the results of our analysis. The derivation of the theorems used in Sec. II can be found in the Appendix on EPAPS[27]. Applications to electron spins in semiconductors of relevance to solid state quantum information processing are made in Sec. III. Discussion and conclusions follow in Sec. IV.

II. RESULTS

Let $S$ be the electron spin precessing in a magnetic field $B$ applied in the $z$ direction. The electron spin is coupled to $N$ nuclear spins $I_1, I_2, \ldots, I_N$ via the hyperfine interaction, characterized by the coupling constants $A_1, A_2, \ldots, A_N$. Due to the hyperfine coupling, the nuclei induce an effective Overhauser field on the electron with magnitude $\sum_j A_j I_j$. Here we consider the Hamiltonian

$$H = \gamma_S B S_z + \gamma_I B \sum_j I_{jz} + \sum_j A_j \left( S_{z} I_{jz} + \frac{1}{2} (S_+ I_{j-} + S_- I_{j+}) \right),$$

where $\gamma_S$ and $\gamma_I$ are the gyromagnetic ratios of the electron and nuclei, respectively ($\gamma_S \approx 10^3 \gamma_I$). Eq. (1) can be written as

$$H = H_0 + V,$$

$$H_0 = \gamma_S B S_z + \gamma_I B \sum_j I_{jz} + \sum_j A_j S_z I_{jz},$$

$$V = \sum_j \frac{1}{2} A_j (S_+ I_{j-} + S_- I_{j+}).$$

This Hamiltonian conserves the total spin angular momentum $J_z = S_z + \sum_j I_{jz}$; hence, the total number of “down” spins is conserved. For convenience, we will label each of these blocks by the quantum number $L$, where $L$ is the total number of “down” spins (i.e. $J_z = N - 2L$, $L = (N - J)/2$). Then the Hamiltonian can be block diagonalized based on the $J_z$ operator [17]. There will be $N + 2$ blocks, one for each possible value of $L$, and the $L^{th}$ block will be an $\binom{N+1}{L} \times \binom{N+1}{L}$ matrix. For the remainder of the paper, it will be assumed that all analysis applies to some subspace labeled by a particular value of $L$.

The unperturbed Hamiltonian, $H_0$, commutes with the $S_z$ operator. Thus, the eigenstates of $H_0$ can be labeled by the $z$-polarization of the electron. On the other hand, the full Hamiltonian $H$ does not commute with $S_z$ because $V$
connects electron spin-up states to electron spin-down states, and vice-versa. At high fields greater than some critical field $B > B_c$, the Zeeman energy of the electron will dominate the energy of the eigenstates, and the electron spin-up and electron spin-down manifolds will be well separated in energy. Thus, if the perturbation is in some sense smaller in magnitude than this energy gap, the effects of the perturbation will be small and $S_z$ will still be an approximately good label for eigenstates of $H$ (see Figure 1).

Our analysis in Sec. II.A demonstrates that if the magnetic field $B$ is greater than the critical field $B_c$, then the electron spin magnetization $\langle S_z \rangle$ never changes appreciably. Because the eigenstates of $H$ are nearly eigenstates of $S_z$ at high fields, the $z$-polarization of the electron does not undergo substantial (i.e. $1/e$) decay (this result matches the perturbative result found in [15]). Furthermore, based on these arguments, a field-dependent bound is placed on the maximum fluctuation of $S_z$ from its initial value: the greater the field, the smaller the maximum possible fluctuation. Again, it should be emphasized that there may be small magnitude fluctuations in $\langle S_z(t) \rangle$, which occur on a finite timescale. However, because our definition of $T_1$ (the timescale for $1/e$ decay of $\langle S_z \rangle$) quantifies the large-scale coherence properties of the electron spin, the absence of large-scale decay renders $T_1$ effectively infinite. Although $T_1$ is infinite for large magnetic fields, dephasing processes can still occur provided that they conserve the $z$-polarization of the electron spin. Although this statement seems like a contradiction given that the perturbation, $V$, flips the electron spin, virtual transitions involving $V^2$ can nevertheless flip pairs of nuclear spins without affecting the electron spin. For this reason, it is also important to place a lower bound on $T_2$.

Derivation of the lower bound on $T_2$ in Sec. II.B follows a different methodology than the bound on $T_1$. Because $T_2$ is defined as the timescale of the decay of the spin echo envelope [19], a lower bound on the magnitude of $\langle S_z(2t) \rangle$ after a pulse echo sequence implies a lower bound on $T_2$. Specifically, the standard definition of $T_2$ is the minimal time, $t$ such that,

$$\langle S_z(2t) \rangle = \frac{1}{2e},$$

where a $\pi$ pulse is applied at time $t$ (we have dropped a factor of $\hbar$ for convenience). It is assumed that for an infinite system, there will be no large-scale recurrence in $S_z(2t)$, but only dissipative behavior. Small magnitude recurrence, due for instance to echo modulation [20], may still exist, but large-scale recurrence on the order of $1/e$ is assumed to be absent. Even if this large-scale recurrence does exist, the lower bound on $T_2$ is still valid. In particular, if it can be shown that for some value of $t'$, $\langle S_z(2t') \rangle \geq 1/2e$, it follows that $T_2 \geq t'$.

This bound on $\langle |S_z(2t')| \rangle$ can be proven using a variational argument. Every trial state $|\phi\rangle$ induces a probability distribution over the eigenenergies of $H$. The mean of this energy distribution is given by the expectation value

$$\langle E \rangle = \langle \phi | H | \phi \rangle.$$

Likewise, the variance of this energy distribution is

$$\langle \langle \Delta E \rangle^2 \rangle = \langle \phi | H^2 | \phi \rangle - \langle \phi | H | \phi \rangle^2.$$  

The variance of the energy will be equal to zero if and only if $|\phi\rangle$ is an eigenstate of $H$ (or a superposition of degenerate eigenstates). For all other states which are superpositions of non-degenerate eigenstates, the variance will be greater than zero. The essential feature of the theorems given in Sec. II.B is that the dynamics undergone by any state $|\phi\rangle$ are limited in timescale by the variance in energy of that state. Using this idea, it can be shown that the dephasing processes of the system have a timescale governed by the energy

$$W_{max} = \sum_j \frac{\hbar^4 A_j^2}{4(h\gamma S B)}.$$  

Conceptually, our bound on $T_2$ makes mathematically rigorous the idea that the coherence time of a state is inversely proportional to its linewidth.

Before proceeding to the proof of these results, two important points must be made. First, this method constructs only a rigorous lower bound on $T_2$; it does not predict the actual value of $T_2$ nor does it claim to be a tight lower bound on $T_2$. In fact, numerical work suggests that the spin echo pulse may actually remove nearly all hyperfine-induced decoherence, leading to only small magnitude decay of $S_z(t)$ under the spin echo sequence for all times, $t$ [18]. However, this evidence is numerical and has been confirmed only for small numbers of spins ($N \sim 10$). In this paper we are interested in what rigorous statements can be made regarding the $T_2$ time for a large system ($N \sim 10^4 - 10^6$).

Second, it is the special structure of this system at high $B$ fields that allows a useful bound to be constructed. Though this method is general and can be used to construct valid bounds on $T_2$ for other systems, these bounds may prove to be useless in practice (for instance, the rigorous bound that $T_2 > 10^{-30}$ s is valid, but is a practically useless bound). Fortunately, we find that our method provides practically useful lower bound values for real systems of relevance to quantum information processing (see Sec. III).
A. Bound on $T_1$

The proof of the bound on $T_1$ will be given first and will be organized as follows. First, we will show that the eigenstates of $H$ can be divided into two subspaces, the $+$ subspace and the $-$ subspace (Theorem 1). Eigenstates contained in the $+$ subspace contain large contributions from electron spin-up states, and eigenstates in the $-$ subspace contain large contributions from electron spin-down states. In other words, $S_z$ is an approximate quantum number for the true eigenstates of $H$. Second, we will show that this fact places bounds on the eigenvalues of $H$ (Theorem 2). Next, we will show that every eigenstate of $H_0$ is almost completely contained within either the $+$ or $-$ subspace of $H$ (Theorem 3). To complete the bound on $T_1$, we will show that these previous statements imply a lower bound on the decay of $S_z$ from its initial state (Theorem 4).

For future convenience, we define

$$A[k] = \sum_{j \in k \text{ largest}} A_j$$  \tag{9}$$

$$A_2[k] = \sum_{j \in k \text{ largest}} A_j^2$$  \tag{10}$$

$$A_{\text{max}} = \sum_j A_j$$  \tag{11}$$

$$A_{2,\text{max}} = \sum_j A_j^2$$  \tag{12}$$

In other words, $A[k]$ is the sum of the $k$ largest hyperfine coupling constants. $A_{\text{max}}$ is the sum of all $N$ hyperfine coupling constants and therefore represents the maximum magnitude of the Overhauser field. The matrix $H_0$ commutes with the $S_z$ and $I_{z\gamma}$ operators; hence eigenstates of $H_0$ can be labeled by the $z$-polarization of the electron spin and the $z$-polarization of the nuclei. Let $|\uparrow\rangle$ and $|\downarrow\rangle$ represent the $+z$ and $-z$ polarized states of the electron spin, respectively, and let $z$ be an $N$-bit string of $+1$s and $-1$s representing the $z$-polarization of the $N$ nuclei. The eigenstates of $H_0$ are of the form $|\uparrow; z\rangle$ or $|\downarrow; z\rangle$ (it should be remembered that $z$ will contain $L$ down spins and $z'$ will contain $L - 1$ down spins, since the total number of down spins, including the electron spin, is conserved). The eigenvalue $E_{0,z}^\uparrow$ corresponding to the eigenstate $|\uparrow; z\rangle$ is

$$E_{0,z}^\uparrow = \frac{1}{2} \hbar \gamma_S B + \frac{1}{2} (N - 2L) \hbar \gamma_I B + \frac{1}{4} \hbar^2 \sum_j A_j z_j.$$  \tag{13}$$

Similarly, the eigenvalue $E_{0,z'}^\downarrow$ corresponding to the eigenstate $|\downarrow; z\rangle$ is,

$$E_{0,z'}^\downarrow = -\frac{1}{2} \hbar \gamma_S B + \frac{1}{2} (N - 2L + 2) \hbar \gamma_I B - \frac{1}{4} \hbar^2 \sum_j A_j z'_j.$$  \tag{14}$$

From Eqs. (13) and (14) it is clear that the eigenvalues of $H_0$ fall within certain ranges. In particular, we can write

$$E_{0,\text{min}}^\uparrow \leq E_{0,z}^\uparrow \leq E_{0,\text{max}}^\uparrow$$  \tag{15}$$

where we have defined

$$E_{0,\text{min}}^\uparrow = \frac{1}{2} \hbar \gamma_S B + \frac{1}{2} (N - 2L) \hbar \gamma_I B - \frac{1}{4} \hbar^2 (2A[L] - A_{\text{max}})$$  \tag{16}$$

$$E_{0,\text{max}}^\uparrow = \frac{1}{2} \hbar \gamma_S B + \frac{1}{2} (N - 2L) \hbar \gamma_I B + \frac{1}{4} \hbar^2 (2A[N - L] - A_{\text{max}})$$  \tag{17}$$

$$E_{0,\text{min}}^\downarrow = -\frac{1}{2} \hbar \gamma_S B + \frac{1}{2} (N - 2L + 2) \hbar \gamma_I B - \frac{1}{4} \hbar^2 (2A[N - L + 1] - A_{\text{max}})$$  \tag{18}$$

$$E_{0,\text{max}}^\downarrow = -\frac{1}{2} \hbar \gamma_S B + \frac{1}{2} (N - 2L + 2) \hbar \gamma_I B + \frac{1}{4} \hbar^2 (2A[L - 1] - A_{\text{max}}).$$  \tag{19}$$

It is critical to all proofs that follow that there is some energy gap $\Delta E$ between the electron spin-up and spin-down subspaces (see Figure 1). In other words, the lowest energy electron spin-up state must be greater in energy than the
highest energy electron-spin down state. Using Eqs. (16) and (19), it can be demonstrated that an energy gap exists if
\[ \hbar (\gamma_S - \gamma_I) B \geq \frac{1}{2} \hbar^2 \left( A[N - L] + A[N - L + 1] - A_{\text{max}} \right). \] (20)

An additional condition that emerges from Theorem 2 sets the critical field slightly higher than Eq. (20) would imply. Furthermore, we would like the definition of \( B_c \) to be independent of \( L \). Therefore, for the remainder of the paper, we will assume that we are in the high-field regime defined by
\[ B > B_c, \] (21)
where
\[ B_c = \frac{\hbar A_{\text{max}}}{\gamma_S - \gamma_I}. \] (22)

Typical values of this critical field for various systems are given in Table 1.

Using these facts, we now show that the eigenstates of \( H \) are near-eigenstates of \( S_z \). We define the projection operators \( \Pi^\uparrow \) and \( \Pi^\downarrow \) such that
\[ \Pi^\uparrow = |\uparrow\rangle \langle \uparrow| \otimes I \] (23)
\[ \Pi^\downarrow = |\downarrow\rangle \langle \downarrow| \otimes I, \] (24)
where \( I \) is the identity operator on the nuclear spins. Then the following theorem can be proven:

**Theorem 1.** The eigenstates of \( H \) can be divided into two subspaces, which we will label + and −. An eigenstate \( |\psi^+\rangle \) in the + subspace obeys the conditions
\[ \langle \psi^+ | \Pi^\uparrow | \psi^+ \rangle \geq \xi_1 \] (25)
\[ \langle \psi^+ | \Pi^\downarrow | \psi^+ \rangle \leq \xi_2, \] (26)
and an eigenstate \( |\psi^-\rangle \) in the − subspace obeys the conditions
\[ \langle \psi^- | \Pi^\downarrow | \psi^- \rangle \geq \xi_1 \] (27)
\[ \langle \psi^- | \Pi^\uparrow | \psi^- \rangle \leq \xi_2, \] (28)
where
\[ \xi_{1,2} = \frac{1}{2} \pm \frac{1}{2} \sqrt{1 - \frac{4V^2_{\text{max}}}{(E^\uparrow_{0,\text{min}} - E^\downarrow_{0,\text{max}})^2}}, \] (29)
such that \( 1 \rightarrow +, 2 \rightarrow - \), and
\[ V_{\text{max}} = \frac{1}{2} \hbar^2 \sqrt{A[L]A[N + 1 - L]}. \] (30)

It is useful to note that
\[ V_{\text{max}} \leq \frac{1}{2} \hbar^2 A_{\text{max}} \] (31)
for all \( L \). Above the critical field given in Eq. (22), we find that \( \xi_1 > 1/2 \) and \( \xi_2 < 1/2 \). Hence, we can write the eigenstates of \( H \) as either \( |\psi^+_k\rangle \) or \( |\psi^-_k\rangle \), where the + eigenstates are those which contain contributions primarily from electron spin-up states (\(|\uparrow\rangle\)), and the − eigenstates contain contributions primarily from the electron spin down states (\(|\downarrow\rangle\)),
\[ \langle \psi^+_k | \Pi^\uparrow | \psi^+_k \rangle > 1/2 \] (32)
\[ \langle \psi^-_k | \Pi^\downarrow | \psi^-_k \rangle > 1/2. \] (33)

To summarize, we have shown that at high fields, states can still be approximately labeled according to their electron spin polarization.

We now proceed with the second step of the proof.
Theorem 2. The eigenvalue $E^+_k$ corresponding to eigenstate $|\psi^+_k\rangle$ and the eigenvalue $E^-_k$ corresponding to eigenstate $|\psi^-_k\rangle$ obey the following conditions

$$E^-_{\min} \leq E^-_k \leq E^-_{\max}$$

(34)

and

$$E^+_{\min} \leq E^+_k \leq E^+_{\max}$$

(35)

where

$$E^+_{\min} = E^{\uparrow}_{0,\min} - V_{\max}$$

(36)

$$E^+_{\max} = E^{\uparrow}_{0,\max} + V_{\max}$$

(37)

$$E^-_{\min} = E^{\downarrow}_{0,\min} - V_{\max}$$

(38)

$$E^-_{\max} = E^{\downarrow}_{0,\max} + V_{\max}.$$

(39)

Theorem 2 demonstrates that the eigenvalues of the $+$ and $-$ eigenstates of $H$ fall in approximately the same ranges as the eigenvalues of the $\uparrow$ and $\downarrow$ eigenstates of $H_0$ (c.f. Eqs. (16–19)).

The third step in the proof is to show that any eigenstate of $S_z$, in other words, any state with a well-defined electron polarization, is contained almost completely in either the $+$ or $-$ subspace (note that this statement does not immediately follow from Theorem 1, in which we proved the converse; namely that $+$ or $-$ eigenstates of $H$ are almost completely contained within the $\uparrow$ or $\downarrow$ subspace.)

Theorem 3. Let $|\uparrow, \psi\rangle$ be an arbitrary state in which the electron is polarized in the $+z$ direction, i.e. $|\psi\rangle$ is some arbitrary state of the nuclear spins. Then,

$$\langle \uparrow, \psi | \Pi_- | \uparrow, \psi \rangle \leq \epsilon^2_\uparrow,$$

(40)

where $\Pi_-$ is the projector onto the $-$ eigenstates of $H$ and

$$\epsilon^2_\uparrow = \frac{(E^{\uparrow}_{0,\max} - E^{\uparrow}_{0,\min})^2 + V_{\max}^2}{(E^{\uparrow}_{\max} - E^{\uparrow}_{0,\min})^2}$$

(41)

Similarly,

$$\langle \downarrow, \psi | \Pi_+ | \downarrow, \psi \rangle \leq \epsilon^2_\downarrow,$$

(42)

where $\Pi_+$ is the projector onto the $+$ eigenstates of $H$ and

$$\epsilon^2_\downarrow = \frac{(E^{\downarrow}_{0,\max} - E^{\downarrow}_{0,\min})^2 + V_{\max}^2}{(E^{\downarrow}_{\max} - E^{\downarrow}_{0,\max})^2}$$

(43)

To simplify future analysis, we will use

$$\epsilon = \max \{ \epsilon_\uparrow, \epsilon_\downarrow \}$$

(44)

in place of $\epsilon_\uparrow$ and $\epsilon_\downarrow$. Then Theorem 3 demonstrates that an arbitrary state with a $\pm z$ polarized electron has at most an amplitude of $\epsilon$ on the $\mp$ subspace.

Finally, let us consider the time evolution of the state $|\uparrow, \psi\rangle$. We will show that at high fields, the $z$-polarization of a state does not vary substantially from its initial polarization. In other words, at fields $B > B_c$, $T_1$ is infinite because the $z$-polarization of the electron does not undergo substantial decay.

Theorem 4. Let $U$ be the time evolution operator given by,

$$U = \exp (iHt).$$

(45)
Given an arbitrary initial state $|\psi, \psi\rangle$ with the electron polarized in the $+z$ direction, the following inequality holds for all times $t$,

$$\langle \psi, \psi | U(t) S_z U(t) | \psi, \psi \rangle \geq \frac{\hbar}{2} (1 - 8\epsilon^2).$$

(46)

Similarly, given an arbitrary initial state $|\psi, \psi\rangle$ with the electron polarized in the $-z$ direction,

$$\langle \psi, \psi | U(t) S_z U(t) | \psi, \psi \rangle \leq -\frac{\hbar}{2} (1 - 8\epsilon^2).$$

(47)

Given that the expressions for $\epsilon^2$ and $\xi_2$ are complicated, it is useful to observe their general form. If we assume that $\hbar \gamma_S B \gg \hbar^2 A_{\text{max}}$, we can expand $\epsilon$ and $\xi_2$ to lowest order in $1/B$. To the lowest order in $1/B$ we find that $\epsilon^2 \approx 5\xi_2/4$ and

$$\xi_2 \approx \frac{\frac{1}{8} \hbar^4 A_{\text{max}}^2}{(\hbar(\gamma_S - \gamma_I)B)^2}$$

(48)

$\epsilon^2$ and $\xi_2$ are essentially the squares of the ratio of the Overhauser energy $\hbar^2 A_{\text{max}}$ to the electron Zeeman energy $\hbar \gamma_S B$. If this ratio is small, then the $z$ spin of the electron undergoes only small fluctuations on the order of $\epsilon^2$. Thus, for fields greater than $B_c$, decay is suppressed and $T_1$ is consequently infinite.

**B. Bound on $T_2$**

Let us now turn our attention to the second relaxation time $T_2$. $T_2$ describes the decay of the in-plane magnetization under a spin echo experiment, i.e. the decay of the expectation value $|\langle S_z(2t) \rangle|$, where a single spin echo $\pi$-pulse is applied at time $t$. The bound on $T_2$ is based on the variational principle mentioned above. It will be shown that the $+$ and $-$ eigenstates obey very similar evolution equations. Thus, the dephasing which occurs between $+$ and $-$ eigenstates is limited by the “magnitude” of this difference in dynamics.

We first begin with some general observations. Let $|\phi\rangle$ be a wavepacket formed from a linear combination of eigenstates of the Hamiltonian $H$. Then we can write,

$$\langle E \rangle = \langle \phi | H | \phi \rangle$$

(49)

$$= \sum_k |\langle \psi_k | \phi \rangle|^2 E_k$$

(50)

$$= \sum_k p_k E_k$$

(51)

$$\langle E^2 \rangle = \langle \phi | H^2 | \phi \rangle$$

(52)

$$= \sum_k |\langle \psi_k | \phi \rangle|^2 E_k^2$$

(53)

$$= \sum_k p_k E_k^2$$

(54)

where $|\psi_k\rangle$ is an eigenket of $H$ with eigenvalue $E_k$ and where we have defined $p_k = |\langle \psi_k | \phi \rangle|^2$. We can view $p_k$ as a probability distribution over the eigenstates of $H$ induced by the state $|\phi\rangle$. Then we can describe the root mean squared width $\Delta E$ of the distribution of energies as,

$$\Delta E^2 = \langle E^2 \rangle - \langle E \rangle^2$$

(55)

$$= \sum_k p_k (E_k - \langle E \rangle)^2.$$  

(56)

Theorems 5 and 6 will apply this idea to bounding the dynamics of $|\phi\rangle$.

**Theorem 5.** Let $|\phi\rangle$ be a trial state with energy width $\Delta E$, let $\Delta E_{\text{max}} = \alpha \Delta E$ be some energy width cutoff with $\alpha > 1$, and let $M$ be the set of eigenstates of $H$ states such that $|E_k - \langle E \rangle| > \Delta E_{\text{max}}$. Then,

$$\sum_{k \in M} p_k \leq \frac{1}{\alpha^2}$$

(57)
Theorem 6. Given a state $\phi$ with energy width $\Delta E$, then for times $t$ which satisfy,

$$t < \frac{\hbar \pi}{2\alpha \Delta E},$$

(58)

with $\alpha > 1$, the free evolution of $|\phi\rangle$ is given by,

$$U(t) |\phi\rangle = \exp (i\theta) \sqrt{1 - \lambda^2} |\phi\rangle + \lambda |r\rangle,$$

(59)

where $\theta$ is some arbitrary phase, $|r\rangle$ is a residual vector orthogonal to $|\phi\rangle$ and the parameter $\lambda$ satisfies the inequality,

$$\lambda^2 \leq c(\Delta E, t)^2.$$

(60)

The parameter $c(\Delta E, t)$ is a function of the root mean squared energy width $\Delta E$ and the time $t$ such that,

$$c(\Delta E, t)^2 = 1 - \left(1 - \frac{1}{\alpha^2}\right) \cos \left(\frac{\alpha \Delta E t}{\hbar}\right) + \frac{1}{\alpha^2},$$

(61)

and $\alpha$ is a parameter that can be chosen to optimize the resulting bound on $T_2$.

Using Theorem 6, it is possible to bound the dynamics of any initial state $|\phi\rangle$ by evaluating its energy width, $\Delta E$. We will come back to the choice of $\alpha$ later when we evaluate the $T_2$ bound for specific systems in Sec. III.

We now turn to the problem of using the results of Theorems 5 and 6 to place bounds on $T_2$ for our system of an electron spin coupled to a lattice of nuclear spins. We assume that we start with a thermally equilibrated ensemble of nuclear spins. A spin echo experiment begins with an initial $\pi/2$ pulse denoted by the operator,

$$R_{\pi/2} = \left(|\Rightarrow\rangle \langle \Uparrow| + |\Leftarrow\rangle \langle \Downarrow|\right) \otimes I_I$$

(62)

where

$$|\Rightarrow\rangle = \frac{1}{\sqrt{2}} (|\Uparrow\rangle - |\Downarrow\rangle)$$

(63)

$$|\Leftarrow\rangle = \frac{1}{\sqrt{2}} (|\Uparrow\rangle + |\Downarrow\rangle).$$

(64)

The system then undergoes free evolution for a time $t$. This free evolution is followed by a $\pi$ pulse, denoted by the operator

$$R_{\pi} = \left(|\Downarrow\rangle \langle \Uparrow| + |\Uparrow\rangle \langle \Downarrow|\right) \otimes I_I,$$

(65)

which flips the electron spin. Finally, there is a second period of free evolution for a time $t$. This sequence is described by the unitary spin echo evolution operator, $\tilde{U}$,

$$\tilde{U} = U(t) R_{\pi} U(t).$$

(66)

The decay function $v(2t)$ is given by the magnitude of the in-plane magnetization after this sequence.

At high fields, it is possible to cool the system well below the Zeeman energy of the electron, thereby approximately polarizing the electron in the $-z$ direction. However, because the Zeeman energy of a nucleus is $10^3$ smaller than that of the electron, the nuclei will, in general, be highly thermalized [11]. For the purposes of our analysis, we can then assume that in the initial state of the system, the electron is polarized in the $-z$ direction (i.e. aligned with the field) and is uncoupled from the nuclei, which are in a completely mixed state. The density matrix for such a system is given by

$$\rho_0 = k |\Downarrow\rangle \langle \Downarrow| \otimes I_I$$

(67)

where $k$ is some normalization constant which we will drop for convenience. Thus, we need to calculate the quantity

$$v(2t) = \left| \text{Tr} \left[ R_{\pi/2} \tilde{U} R_{\pi/2} \rho_0 \right] \right|.$$

(68)
Using the cyclic property of the trace and Eq. (67), we can simplify this expression to

\[
v(2t) = \frac{1}{2} \left| \text{Tr} \left[ \hat{U}^\dagger S_+ \hat{U} R_\pi / 2 \rho_0 R^2_\pi / 2 \right] \right|
\]

\[
= \frac{1}{2} \left| \text{Tr} \left[ \hat{U}^\dagger S_+ \hat{U} (\mathcal{I}_S + R_\pi) \otimes \mathcal{I}_I \right] \right|
\]

\[
= \frac{1}{2} \left| \text{Tr} \left[ \hat{U}^\dagger S_+ \hat{U} \right] + \frac{1}{2} \text{Tr} \left[ \hat{U}^\dagger S_+ \hat{U} R_\pi \right] \right|
\]

\[
= \frac{1}{2} \left| \text{Tr} \left[ \hat{U}^\dagger S_+ \hat{U} R_\pi \right] \right|
\]

where \( \mathcal{I}_S \) is the identity operator on the electron spin.

If we evaluate the trace in Eq. (72) using the eigenbasis of \( H \), we can separate the contributions of the \( + \) and \( - \) eigenstates,

\[
v(2t) = \frac{1}{2} \text{Tr} \left[ \hat{U}^\dagger S_+ \hat{U} R_\pi \right]
\]

\[
= \frac{1}{2} \sum_i \langle \psi^-_i | \hat{U}^\dagger S_+ \hat{U} R_\pi | \psi^+_i \rangle + \sum_i \langle \psi^+_i | \hat{U}^\dagger S_+ \hat{U} R_\pi | \psi^-_i \rangle
\]

\[
\geq \frac{1}{2} \sum_i \langle \psi^-_i | \hat{U}^\dagger S_+ \hat{U} R_\pi | \psi^+_i \rangle - \frac{1}{2} \sum_i \langle \psi^+_i | \hat{U}^\dagger S_+ \hat{U} R_\pi | \psi^-_i \rangle
\]

Using Theorem 1 and Theorem 4, we find, after some tedious algebra, that the contribution of the \( + \) eigenstates is negligible,

\[
\sum_i \langle \psi^+_i | \hat{U}^\dagger S_+ \hat{U} R_\pi | \psi^-_i \rangle \leq 2 \epsilon \sqrt{\xi_2} + 4 \epsilon^2 + \xi_2.
\]

If we let \( | \psi^-_i \rangle = | \uparrow, \psi^-_{\psi,i} \rangle + | \downarrow, \psi^-_{\psi,i} \rangle \) be an eigenstate in the \( - \) subspace, we can also simplify the contribution of the \( - \) states,

\[
\sum_i \langle \psi^-_i | \hat{U}^\dagger S_+ \hat{U} R_\pi | \psi^-_i \rangle \geq \sum_i \langle \uparrow, \psi^-_{\psi,i} | \hat{U}^\dagger S_+ \hat{U} R_\pi | \downarrow, \psi^-_{\psi,i} \rangle - 2 \epsilon \sqrt{\xi_2} - 4 \epsilon^2 \xi_2.
\]

Combining Eqs. (75–76) we obtain

\[
v(2t) \geq \frac{1}{2} \sum_i \langle \uparrow, \psi^-_{\psi,i} | \hat{U}^\dagger S_+ \hat{U} R_\pi | \downarrow, \psi^-_{\psi,i} \rangle - 2 \epsilon \sqrt{\xi_2} - 2 \epsilon^2 - \frac{1}{2} \xi_2 - 2 \epsilon^2 \xi_2
\]

The problem of evaluating \( v(2t) \) has been reduced to determining the time evolution of the states \( | \downarrow, \psi^-_{\psi,i} \rangle \) and \( | \uparrow, \psi^-_{\psi,i} \rangle \). The physical interpretation of the derivation of Eq. (78) is that we have separated out terms which primarily contribute to dephasing processes (i.e., the terms in the sum over \( i \)) from terms due to the longitudinal relaxation of the electron (i.e., the remaining terms). These longitudinal terms are on the order of \( \xi_2 \) or \( \epsilon^2 \) since this is the magnitude of longitudinal relaxation demonstrated in Theorem 4.

We must next evaluate the sum in Eq. (78). It is clear that under free evolution, \( | \downarrow, \psi^-_{\psi,i} \rangle \) simply acquires an overall phase (minus some small residual), since it is the projection of an eigenstate of \( H \) onto the \( \downarrow \) subspace. This statement is formalized in Theorem 7. A similar idea can be used to characterize the evolution of \( | \uparrow, \psi^-_{\psi,i} \rangle \). Because \( | \uparrow, \psi^-_{\psi,i} \rangle \) is not necessarily close to an eigenstate of \( H \), its evolution can be complicated and may not in general be characterized simply by an overall phase. If we could calculate the energy width of \( | \uparrow, \psi^-_{\psi,i} \rangle \), then we could use Theorem 6 to bound its dynamics. However, because the explicit form of the nuclear state \( | \psi^-_{\psi,i} \rangle \) is not known, we must use a more subtle method of obtaining the energy width. Theorem 8 shows that a trial state \( | \phi \rangle \) can be constructed with energy width given by a quantity \( ||W||_2 \) that is determined by \( A_{2,\text{max}}, E_{\text{max}}^- \), and \( E_{0,\text{max}}^- \). It is also shown that \( | \uparrow, \psi^-_{\psi,i} \rangle \) is the projection of \( | \phi \rangle \) onto the \( \uparrow \) subspace. Using this fact, Theorem 9 then bounds the evolution of \( | \uparrow, \psi^-_{\psi,i} \rangle \). We now present these three results.
Theorem 7. Let \(|\psi^-_t\rangle = |\uparrow, \psi^-_{\hat{g}, i}\rangle + |\downarrow, \psi^-_{\hat{q}, i}\rangle\) be an eigenstate of \(H\) in the \(-\) subspace. Then
\[
U |\downarrow, \psi^-_{\hat{q}, i}\rangle = e^{i\omega \hat{q}, i t} |\downarrow, \psi^-_{\hat{q}, i}\rangle + |r^-_{\hat{q}, i}\rangle
\]  
(79)
where
\[
|r^-_{\hat{q}, i}\rangle = \left( e^{i\omega \hat{q}, i t} - U \right) |\uparrow, \psi^-_{\hat{g}, i}\rangle.
\]  
(80)

Again, because \(|\downarrow, \psi^-_{\hat{q}, i}\rangle\) is close to an eigenstate of \(H\), its evolution will essentially be characterized by an overall phase plus a small residual. Because \(|\uparrow, \psi^-_{\hat{q}, i}\rangle\) is not necessarily close to an eigenstate of \(H\), we cannot use the same reasoning to characterize its evolution. Instead, we show that \(|\uparrow, \psi^-_{\hat{q}, i}\rangle\) is close to a trial state \(|\phi_i\rangle\) whose evolution we can characterize.

Theorem 8. There exists a trial state \(|\phi_i\rangle\) such that
\[
\Pi_{\hat{g}} |\phi_i\rangle = |\uparrow, \psi^-_{\hat{q}, i}\rangle
\]  
(81)
and
\[
\langle \phi_i | H^2 | \phi_i \rangle - \langle \phi_i | H | \phi_i \rangle^2 \leq \|W\|^2
\]  
(82)
where
\[
\|W\|^2 \leq \frac{\hbar^4}{4 \left( E_{\text{max}} - E_{\text{0, max}} \right)} A_{\text{2, max}} (1 + \epsilon)
\]  
(83)

According to Theorem 8, the trial state \(|\phi_i\rangle\) has an energy width of \(\|W\|_2\). Using Theorem 6, we know that for \(t < \hbar \pi/2 \|W\|_2\) the evolution of \(|\phi_i\rangle\) will then be well-described by phase evolution with some arbitrary phase denoted by \(\omega \hat{q}, i t\) (corresponding to \(\theta\) in Theorem 4) plus some small residual. Because \(|\phi_i\rangle\) is close to the state \(|\uparrow, \psi^-_{\hat{q}, i}\rangle\), Theorem 9 now uses the same reasoning as in Theorem 7 to characterize the evolution of \(|\uparrow, \psi^-_{\hat{q}, i}\rangle\).

Theorem 9. Let \(|\psi^-_t\rangle = |\uparrow, \psi^-_{\hat{g}, i}\rangle + |\downarrow, \psi^-_{\hat{q}, i}\rangle\) be an eigenstate of \(H\) in the \(-\) subspace. Then for \(t < \hbar \pi/2 \|W\|_2\),
\[
U |\uparrow, \psi^-_{\hat{g}, i}\rangle = e^{i\omega \hat{g}, i t} \sqrt{1 - \lambda_i^2} |\uparrow, \psi^-_{\hat{g}, i}\rangle + \lambda_i |r^-_{\hat{g}, i}\rangle + \left( e^{i\omega \hat{q}, i t} - U \right) |\downarrow, \phi_{\hat{q}, i}\rangle
\]  
(84)
where the following inequalities are satisfied,
\[
\lambda_i^2 \leq c(\|W\|_2, t)^2,
\]  
(85)
\[
\langle \downarrow, \phi_{\hat{q}, i} | \downarrow, \phi_{\hat{q}, i} \rangle \leq c^2,
\]  
(86)
\[
\left| \langle r^-_{\hat{g}, i} | \uparrow, \psi^-_{\hat{g}, i} \rangle \right| \leq \epsilon.
\]  
(87)

Using Theorems 7 and 9, we obtain the following results for the evolution of \(|\downarrow, \psi^-_{\hat{q}, i}\rangle\) and \(|\uparrow, \psi^-_{\hat{q}, i}\rangle\) under the spin echo experiment:
\[
\hat{U} |\downarrow, \psi^-_{\hat{g}, i}\rangle = e^{i(\omega \hat{q}, i + \omega \hat{g}, i) t} \sqrt{1 - \lambda_i^2} |\uparrow, \psi^-_{\hat{g}, i}\rangle + e^{i\omega \hat{q}, i t} \lambda_i |r^-_{\hat{g}, i}\rangle
\]  
(88)
\[
+ e^{i\omega \hat{g}, i t} \left( e^{i\omega \hat{g}, i t} - U \right) |\downarrow, \phi_{\hat{q}, i}\rangle + U R_{\pi} |r^-_{\hat{g}, i}\rangle
\]
\[
\hat{U} \left| \uparrow, \psi_{\theta,i}^- \right\rangle = e^{i(\omega_{\theta,i} + \omega_{\theta,i})t} \sqrt{1 - \lambda^2} \left| \downarrow, \psi_{\theta,i}^- \right\rangle + \lambda_i U R_{\pi} \left| r_{\theta,i}^- \right\rangle 
+ \left( e^{i\omega_{\theta,i} t} U R_{\pi} - U R_{\pi} U \right) \left| \downarrow, \phi_{\theta,i}^- \right\rangle + e^{i\omega_{\theta,i} t} \sqrt{1 - \lambda^2} \left| r_{\theta,i}^- \right\rangle.
\]

One effect of the spin echo evolution is especially important to note. If we compare the free evolution of \( \left| \downarrow, \psi_{\theta,i}^- \right\rangle \) and \( \left| \uparrow, \psi_{\theta,i}^- \right\rangle \), we find that they acquire phases of \( \omega_{\theta,i} t \) and \( \omega_{\theta,i} t \), respectively. Thus, there is a relative phase difference between the two evolutions, which would lead to a fast dephasing time \( T_2 \) when we sum over \( i \) in Eq. (78) to account for our initial, incoherent thermal distribution. However, when the spin echo experiment is performed, Eqs. (88) and (89) show that both states evolve with the phase factor of \( (\omega_{\theta,i} + \omega_{\theta,i})t \), leading to no relative phase difference. As expected, the inhomogeneous dephasing due to an incoherent distribution over initial states is removed by the spin echo experiment.

Finally, we can use Eqs. (88) and (89) to obtain,

\[
\left\langle \downarrow, \psi_{\theta,i}^- \right| \hat{U}^\dagger S_+ \hat{U} \left| \uparrow, \psi_{\theta,i}^- \right\rangle \geq 1 - \xi_2 - \epsilon \left( \| W \|_2, t \right) - 2c(\| W \|_2, t)^2.
\]

Combining Eq. (78) and Eq. (90), we obtain as a final result

\[
v(2t) \geq \frac{1}{2} \left( 1 - 2\xi_2 - 4\epsilon^2 - 4\epsilon \sqrt{\xi_2} - 4\epsilon^2 \xi_2 - \epsilon c(\| W \|_2, t) - 2c(\| W \|_2, t)^2 \right)
\]

for \( t < \hbar \pi / 2 \alpha \| W \|_2 \).

Though this bound is rigorous, it is somewhat complicated. In practice, numerical evaluation of the expressions for \( \epsilon, \xi_2 \), and \( \| W \|_2 \) can be greatly simplified if we assume that \( B \gg B_c \), as in Eq. (48). In this case, we can also use the approximation that

\[
\| W \|_2 \approx W_{\text{max}},
\]

where

\[
W_{\text{max}} = \frac{\hbar^4 A_{2,\text{max}}}{4(\hbar \gamma_S B)}.
\]

It should be emphasized that this approximation \( (B \gg B_c) \) is not intrinsic to our proof and is used only to make our results more compact and easier to present. If a strict bound were desired, we could drop this approximation and keep only the weaker condition that \( B > B_c \).

To obtain a bound on \( T_2 \), we set \( v(2t) = 1/2e \),

\[
c(\| W \|_2, t)^2 + \epsilon c(\| W \|_2, t) + \xi_2 + 2\epsilon^2 + 2\epsilon \sqrt{\xi_2} + 2\epsilon^2 \xi_2 + \frac{1}{2e} - \frac{1}{2} = 0
\]

Let \( c_{\text{max}} \) be the positive root of this equation. Then we solve for \( t_b \) in terms of \( \alpha \) such that \( c(\| W \|_2, t_b) = c_{\text{max}} \),

\[
t_b = \max_\alpha \left\{ \frac{\hbar}{\Delta \gamma_\alpha} \cos^{-1} \left( \frac{1 - c_{\text{max}} + \frac{1}{\alpha}}{1 - \frac{1}{\alpha}} \right) \right\}.
\]

This value of \( t_b \) will be a lower bound on \( T_2 \).

III. APPLICATION TO PHYSICAL SYSTEMS

To demonstrate how our bounding method can be used in practice, we apply it here to three systems of relevance to quantum information processing[3, 21]: (1) a donor impurity in Si, (2) a donor impurity in GaAs, and (3) a GaAs quantum dot. Table 1 shows the important physical parameters for these systems. To obtain a lower bound on \( T_2 \) for each system, we must evaluate Eq. (95) for the given system’s parameters. Because we are free to choose the parameter \( \alpha \) in Theorem 6 with only the restriction that \( \alpha > 1 \), we can select \( \alpha \) to maximize \( t_b \). In each of the following examples, this optimization procedure was carried out to obtain the largest possible \( t_b \).
Consider first the case of a phosphorous impurity atom in a lattice of natural silicon (which contains $4.67\%$ of the spin-1/2 $^{28}\text{Si}$ isotope). Spin coherence is affected by hyperfine coupling between the electron and nuclei and dipolar coupling between nuclei. The hyperfine coupling constants are given by the equation [11, 22],

$$A_j = \frac{8\pi}{3} \gamma_j \gamma_{\text{H}} |\Psi(x_j)|^2,$$

(96)

where $x_j$ is the position of nuclear spin $j$ and $\Psi$ is a hydrogenic orbital for the impurity electron. Using our bounding technique, we can determine when the $S_+I_- + S_-I_+$ terms in the Hamiltonian can be safely neglected as the primary source for electron spin decoherence. The coherence time due to dipole-dipole coupling of nuclei for a sample of natural Si in the presence of an external field aligned with the [111] direction has been estimated to be $T_2 \sim 0.65 \text{msec}$ [11].

This value is essentially independent of the magnetic field strength. In contrast, Figure 2 shows that our lower bound is directly proportional to $B$. Using the data from Figure 2, we find that at approximately $B = 3.0 \text{ Tesla}$, decoherence due to the dipolar nuclear coupling dominates that due to the hyperfine interaction. Thus, for this system, at fields above 3.0 Tesla it is safe to neglect the $S_+I_- + S_-I_+$ terms in the Hamiltonian as sources for decoherence.

Next, we investigate the case of a donor impurity in GaAs and a GaAs quantum dot (20 nm radius, 10 nm thickness). Ga and As nuclei are both spin $3/2$. We can easily extend our spin-1/2 formalism to include higher spin nuclei if we note that a spin-3/2 particle can be treated as a composite system of three spin-1/2 particles. However, we also note that the Ga and As nuclei will have different gyromagnetic ratios $\gamma_{\text{Ga}} = 8.16 \times 10^7$ (s/Tesla), $\gamma_{\text{As}} = 6.42 \times 10^7$ (s/Tesla), and $\gamma_{\text{Ga}} = 4.58 \times 10^7$ (s/Tesla). As a result the Zeeman energy of the electron can be exchanged with the nuclear Zeeman energy, providing a new channel for $T_1$ relaxation. In order to apply our bound to the GaAs systems, we therefore must make the assumption that at high fields nuclear polarization is not transferred between nuclei of different types. We then let $\gamma_j = \gamma_{\text{Ga}}$, and apply our proof as before. Again, the important parameters are shown in Table 1. Figure 3 shows the lower bound on $T_2$ as a function of $B$ for the donor impurity in a GaAs lattice (solid line) and a GaAs quantum dot (dot-dashed line). A lower bound for $T_2$ is provided for fields greater than $\sim 7.5 \text{ Tesla}$. For both systems, our method also shows that $T_2$ is infinite above a critical field of $\sim 2.25 \text{ Tesla}$. Previous studies have shown that nuclear spectral diffusion causes decoherence on a timescale of $\sim 10 \mu\text{sec}$ for both these GaAs systems [11, 23]. Hence, Figure 3 shows that for magnetic fields greater than approximately 12.5 Tesla, nuclear spectral diffusion will be the dominant decoherence mechanism and the $S_+I_- + S_-I_+$ terms in the Hamiltonian can be neglected.

IV. DISCUSSION AND CONCLUSIONS

It is useful to assign a physical significance to the quantities that appear in our bounds. For instance, the relevant quantity in the $T_1$ bound is given alternatively by $\xi^2$ or $\xi_2$, both of which quantify the mixing of the spin-up and spin-down eigenstates of $H_0$. These quantities scale as the square of the ratio of the Overhauser energy $\hbar^2 A_{\text{max}}$ and the electron Zeeman splitting, $\hbar \gamma_S B$. In the language of perturbation theory, the large energy denominator suppresses mixing of spin-up and spin-down states via the perturbation $V$.

On the other hand, the operator which sets the scale for $T_2$ is $W$, which can be written as,

$$W = \Pi_f \left( R_2 V \frac{1}{E_i - H_0} V R_2 - V \frac{1}{E_i^* + H_0} V \right) \Pi_s.$$

(97)

This expression motivates us to identify the operator $W$ with difference in effective Hamiltonians felt by electron spin-up and electron spin-down states. For instance, the second term in this equation describes the Hamiltonian experienced by an electron spin-up state scattered via two applications of $V$ into a second electron spin-up state. On the other hand, the first term applies the same scattering process to an electron spin-down state. Taking the difference of these two terms allows cancellation to occur, reducing the magnitude of $W$, and corresponding to the refocusing of spin states accomplished by the spin echo experiment. If no spin echo were applied, then the first of these terms would be absent, and the magnitude of $W$ would be substantially larger, leading to a faster decay of coherence.

In summary, we have used a completely non-perturbative approach to place a rigorous lower bounds on the $T_1$ and $T_2$ coherence times of an electron spin coupled via the contact hyperfine interaction to a lattice of nuclear spins. For $T_1$, we have shown that for $B > B_c$, the $z$-polarization of the electron is nearly conserved by time evolution; hence $T_1$ is infinite. For $T_2$, we have obtained an analytic expression for a lower bound $t_0$, having explicitly taken into account the effects of a spin echo experiment, which would be required to remove any additional inhomogeneous broadening. This analysis holds above some critical field satisfying the high field condition, $B > B_c$. We have also assumed that at the start of an experiment, the electron is polarized and uncoupled from the completely mixed nuclear initial state; similar results can be derived for any highly mixed nuclear initial state. In general, this method
TABLE I: This table lists the relevant parameters for our calculations of the lower bounds on $T_1$ and $T_2$ for the electron spin. $B_c$ is given by Eq. (22) and $W_{max}$ by Eq. (93). The parameters for the Si:donor and GaAs:donor systems were calculated using simple hydrogenic orbitals as described in [11, 22]. The GaAs quantum dot considered had a radius of 20 nm and a thickness of 10 nm. The parameters for the quantum dot were calculated as described in [11]. Here $\hbar B/W_{max}$ provides an approximate bound for $T_2$ for any field strength $B$.

can be used to determine when the contact hyperfine interaction can be safely neglected as a direct mechanism for electron spin decoherence. We have demonstrated the utility of the bounds for a donor impurity in Si, and -within the approximation that spontaneous nuclear polarization transfer does not occur- for a donor impurity in GaAs, and a GaAs quantum dot.

Future directions for this investigation are two-fold. First, numerical simulations indicate that the coherence time $T_2$ due to hyperfine coupling only might, in fact, be substantially higher than the lower bound given here, possibly even infinite [18]. The primary contribution to $T_2$ comes from the diagonal matrix elements of $W$, which corresponds to the self-energy of the perturbation. Removing this term would greatly improve the lower bound on $T_2$; unfortunately, it is unclear how this term can be suppressed. Also, we have not taken advantage of the substantial symmetries present in the system [18, 24]. Taking advantage of these symmetries in our analysis could dramatically increase our lower bound on $T_2$. Second, this method might be extended to yield bounds on the fidelity of single qubit operations on the electron spin, which would be important for any solid state quantum computing applications. Finally, it is likely that our method could be extended to any quasi-two level system with a significant energy gap that is coupled to a bath via an off-diagonal perturbation. It would be useful to generalize the method so that it could be applied to other systems of interest to quantum computation.
Eigenenergies of $H_0$  

$V_{\text{max}} \uparrow \{ \} \uparrow$  

$\hbar B(\gamma_S - \gamma_I)$  

$\Uparrow \{ \} \Downarrow$  

FIG. 1: A schematic representation of the eigenspectra of $H_0$ and $H$. The eigenstates of the unperturbed Hamiltonian can be grouped into two subspaces, $\uparrow$ and $\downarrow$, based on the $z$-polarization of the electron. In Theorems 1 and 2, we demonstrate that the eigenstates of the full Hamiltonian $H$ can be grouped into two subspaces, $+$ and $-$, based on the approximate $z$-polarization of the electron. Theorem 3 further states that the $+$ and $-$ eigenstates fall within the same range as the $\uparrow$ and $\downarrow$ eigenstates, respectively.

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FIG. 2: Lower bound on $T_2$ versus magnetic field for a donor impurity in a sample of natural Si. Note: this plot shows the dependence of the lower bound on the magnetic field, not the dependence of $T_2$ itself. At high magnetic fields $B \gg B_c$ the lower bound scales approximately as $B$. The dashed line shows the predicted timescale for decoherence due to nuclear spectral diffusion arising from internuclear dipolar coupling [11]. Thus, at fields greater than 3.0 Tesla, this nuclear spectral diffusion will dominate hyperfine coupling as a source for decoherence.

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FIG. 3: Lower bound on $T_2$ versus magnetic field $B$ for a donor impurity in GaAs and a GaAs quantum dot with radius $l_0 = 20$ nm and thickness $z_0 = 10$ nm. The dashed line shows the approximate decoherence time of $\sim 10 \mu$sec for both systems due to dipolar-induced nuclear spectral diffusion [11, 23]. At fields above $\sim 12.5$ Tesla, spectral diffusion will dominate hyperfine coupling as a source for decoherence.

[26] For instance, in [17], the continuum hypothesis is only strictly valid for timescales $t \ll \hbar N^{3/2}/A$ [W.A. Coish, Private communication]

[27] See EPAPS Document No. [number will be inserted by publisher] for the derivations of the theorems presented in Sec. II. A direct link to this document may be found in the online article’s HTML reference section. The document may also be reached via the EPAPS homepage (http://www.aip.org/pubservs/epaps.html) or from ftp.aip.org in the directory /epaps/. See the EPAPS homepage for more information.
Proof of Theorem 1. Let \( |\psi\rangle \) be an eigenstate of \( H \) such that,
\[
H |\psi\rangle = E |\psi\rangle.
\]
(98)
Without loss of generality, we can write \( |\psi\rangle \) as,
\[
|\psi\rangle = |⇑, \psi⇑\rangle + |⇓, \psi⇓\rangle.
\]
(99)
where
\[
|⇑, \psi⇑\rangle = \Pi⇑ |\psi\rangle,
\]
(100)
\[
|⇓, \psi⇓\rangle = \Pi⇓ |\psi\rangle,
\]
(101)
Recall that \( \Pi⇑ \), \( \Pi⇓ \) are the electron spin projection operators defined in Eqs. (23) and (24). Multiplying Eq. (98) on the left by either \( \langle ⇑, \psi⇑| \) or \( \langle ⇓, \psi⇓| \), we obtain the two equations,
\[
\langle ⇑, \psi⇑| H |⇑, \psi⇑\rangle + \langle ⇑, \psi⇑| V |⇓, \psi⇓\rangle = E \langle ⇑, \psi⇑| ⇑, \psi⇑\rangle
\]
(102)
\[
\langle ⇓, \psi⇓| H |⇓, \psi⇓\rangle + \langle ⇓, \psi⇓| V |⇑, \psi⇑\rangle = E \langle ⇓, \psi⇓| ⇓, \psi⇓\rangle.
\]
(103)
It is straightforward to set a bound on the quantities \( \langle ⇑, \psi⇑| H |⇑, \psi⇑\rangle \) and \( \langle ⇓, \psi⇓| H |⇓, \psi⇓\rangle \) since the eigenspectrum of \( H_0 \) is known (see Eqs. (13) and (14)). Specifically, it can be shown that,
\[
\langle ⇑, \psi⇑| H_0 |⇑, \psi⇑\rangle \geq E_{0,\text{min}}^{\uparrow} c_{\uparrow}^2
\]
(104)
and
\[
\langle ⇓, \psi⇓| H_0 |⇓, \psi⇓\rangle \leq E_{0,\text{max}}^{\downarrow} c_{\downarrow}^2,
\]
(105)
where
\[
c_{\uparrow} = \sqrt{\langle ⇑, \psi⇑| ⇑, \psi⇑\rangle}
\]
(106)
\[
c_{\downarrow} = \sqrt{\langle ⇓, \psi⇓| ⇓, \psi⇓\rangle}.
\]
(107)
Here, we pause to introduce several relations involving matrix norms which we will make use of in what follows. Let \( X \) be an arbitrary Hermitian matrix. Then following the standard definitions, we let \( \|X\|_1 \) represent the maximum absolute column sum norm
\[
\|X\|_1 = \max_j \sum_i |X_{ij}|.
\]
(108)
The maximum absolute row sum norm is given by
\[
\|X\|_\infty = \max_i \sum_j |X_{ij}|.
\]
(109)
Finally, the spectral norm, can be defined as
\[
\|X\|_2 = \max_{|\phi\rangle} |\langle \phi| X |\phi\rangle|.
\]
(110)
When applied to a vector \( \|\psi\|_2 \) is simply the \( L_2 \) norm of \( |\psi\rangle \). We will also use two useful relations regarding these norms[25]
\[
\|X |\psi\rangle\|_2 \leq \|X\|_2 \|\psi\|_2
\]
(111)
\[
\|X\|_2 \leq \|X\|_1 \|X\|_\infty.
\]
(112)
We can use these relations applied to the matrix \( V \) to bound the remaining quantities in Eqs. (106) and (107). Using Eq. (111) we obtain
\[
\|\langle ⇑, \psi⇑| V |⇓, \psi⇓\rangle\|_2 \leq c_{\uparrow} c_{\downarrow} \|\Pi⇑ V \Pi⇓\|_2,
\]
(113)
Because the absolute column sum and absolute row sum norms of $V$ are easy to calculate, we can use them to bound the spectral norm of $V$ within a given $L$ subspace using Eq. (112)

$$\|\Pi_{\uparrow}V\Pi_{\uparrow}\|_2 \leq \frac{1}{2} h^2 \sqrt{A[L]A[N-L+1]}$$

$$\equiv \ V_{\text{max}}$$

Similarly,

$$\|\Pi_{\downarrow}V\Pi_{\downarrow}\|_2 \leq \ V_{\text{max}}.$$  

Then we obtain the two bounding inequalities,

$$E_{0,\text{min}}^\phi c_\uparrow^2 + c_\uparrow c_\downarrow V_{\text{max}} \geq E c_\uparrow^2$$  

$$E_{0,\text{max}}^\phi c_\downarrow^2 + c_\uparrow c_\downarrow V_{\text{max}} \geq E c_\downarrow^2$$

Combining Eqs. (117) and (118), we obtain the inequality

$$c_\uparrow^2 + c_\downarrow^2 + c_\uparrow c_\downarrow \frac{E_{0,\text{min}}^\phi - E_{0,\text{max}}^\phi}{V_{\text{max}}} \geq 0.$$  

Since the normalization of $|\psi\rangle$ implies that $c_\uparrow^2 + c_\downarrow^2 = 1$, some brief algebra then yields

$$c_\uparrow^4 - c_\downarrow^2 + \frac{V_{\text{max}}^2}{(E_{0,\text{min}}^\phi - E_{0,\text{max}}^\phi)^2} \geq 0.$$  

Solving the equality for $c_\uparrow^2$, we obtain the two solutions $c_\uparrow = \xi_1, \xi_2$ where $\xi_1$ and $\xi_2$ are given in Eq. (29). Thus, using Eqs. (100) and (106), we have shown that every eigenstate $|\psi_k\rangle$ of $H$ satisfies one of the two inequalities,

$$\langle \psi_k| \Pi_{\uparrow} |\psi_k\rangle \geq \xi_1$$

or

$$\langle \psi_k| \Pi_{\downarrow} |\psi_k\rangle \leq \xi_2$$

QED

Proof of Theorem 2. We will show only one of these bounds as the rest can be demonstrated by identical procedures. For a $+ \psi$ eigenstate, we know from the proof of Theorem 1 that

$$c_\psi \leq \sqrt{\xi_2}$$

$$c_\phi \geq \sqrt{\xi_1}.$$  

Then Eq. (117) gives us,

$$E_{k,\text{min}}^\uparrow \geq E_{0,\text{min}}^\phi - \frac{\xi_2}{\xi_1} \sqrt{V_{\text{max}}}$$

$$\geq E_{0,\text{min}}^\phi - V_{\text{max}}$$

$$= E_{\text{min}}^\uparrow.$$  

Where we have recalled that our high field condition ensures that $\xi_1 > \xi_2$ (see Eq. (29)). The other bounds are derived in exactly the same way. QED

Proof of Theorem 3. We will demonstrate this theorem for the spin-up case; the spin-down case follows the same argument. Let $|\uparrow,\psi\rangle$ be an arbitrary state with the electron polarized in the $+z$ direction. Since $|\uparrow,\psi\rangle$ has no spin-down component, its expectation value over $V$ is 0. Given the eigenspectrum of $H_0$, it is straightforward to show that,

$$E_{k,\text{max}}^\uparrow \geq \langle \uparrow,\psi| H |\uparrow,\psi\rangle \geq E_{0,\text{min}}^\phi$$

(129)
Given the matrix norm of the perturbation $V$ in Eq. (116), it is equally straightforward to show that,

$$
\langle \uparrow, \psi | (H - \langle H \rangle)^2 | \uparrow, \psi \rangle \leq \frac{1}{4} \left( E_{0, \text{max}}^\uparrow - E_{0, \text{min}}^\uparrow \right)^2 + V_{\text{max}}^2.
$$  

(130)

However, we can also write the left hand side of Eq. (130) as,

$$
\langle \uparrow, \psi | (H - \langle H \rangle)^2 | \uparrow, \psi \rangle = \sum_k \left( | \langle \uparrow, \psi | \psi_k^+ \rangle |^2 (E_k^+ - \langle H \rangle)^2 + | \langle \uparrow, \psi | \psi_k^- \rangle |^2 (E_k^- - \langle H \rangle)^2 \right)
$$

(131)

$$
= \sum_k \left( p_k^+ (E_k^+ - \langle H \rangle)^2 + p_k^- (E_k^- - \langle H \rangle)^2 \right),
$$

(132)

where

$$
p_k^\pm = | \langle \uparrow, \psi | \psi_k^\pm \rangle |^2.
$$

(133)

Then using Eqs. (34) and (35), we can bound this quantity as,

$$
\sum_k p_k^+ (E_k^+ - \langle H \rangle)^2 + p_k^- (E_k^- - \langle H \rangle)^2 \geq (0)^2 \sum_k p_k^+ + \left( E_{\text{max}}^\uparrow - E_{\text{min}}^\uparrow \right)^2 \sum_k p_k^-
$$

(134)

$$
= \left( E_{\text{max}}^\uparrow - E_{\text{min}}^\uparrow \right)^2 \langle \uparrow, \psi | \Pi_- | \uparrow, \psi \rangle.
$$

(135)

Combining Eqs. (130) and (135), we obtain,

$$
\langle \uparrow, \psi | \Pi_- | \uparrow, \psi \rangle \leq \epsilon_{\uparrow}^2
$$

(136)

where

$$
\epsilon_{\uparrow}^2 = \frac{1}{4} \left( E_{0, \text{max}}^\uparrow - E_{0, \text{min}}^\uparrow \right)^2 + V_{\text{max}}^2.
$$

(137)

QED

**Proof of Theorem 4.** We will demonstrate the inequality for the initial state $| \uparrow, \psi \rangle$; the spin-down inequality can be proved in a similar manner. Given the initial state $| \uparrow, \psi \rangle$, the state at time $t$ will be given by,

$$
| \psi(t) \rangle = U | \uparrow, \psi \rangle.
$$

(138)

The amplitude in the electron spin-down subspace after time $t$ is given by,

$$
\| \Pi_\uparrow U | \uparrow, \psi \rangle \|_2 = \max_{| \phi \rangle} | \langle \downarrow, \phi | U | \uparrow, \psi \rangle |
$$

(139)

$$
= \max_{| \phi \rangle} | \langle \downarrow, \phi | (\Pi_+ + \Pi_-) U (\Pi_+ + \Pi_-) | \uparrow, \psi \rangle |
$$

(140)

$$
\leq \max_{| \phi \rangle} | \langle \downarrow, \phi | \Pi_+ U \Pi_+ | \uparrow, \psi \rangle | + \max_{| \phi \rangle} | \langle \downarrow, \phi | \Pi_- U \Pi_- | \uparrow, \psi \rangle |
$$

(141)

$$
\leq \max_{| \phi \rangle} \| \Pi_+ | \downarrow, \phi \rangle \|_2 \cdot \| \Pi_+ | \uparrow, \psi \rangle \|_2 + \max_{| \phi \rangle} \| \Pi_- | \downarrow, \phi \rangle \|_2 \cdot \| \Pi_- | \uparrow, \psi \rangle \|_2
$$

(142)

$$
\leq 2 \epsilon
$$

(143)

where the maximization is taken over all normalized nuclear states $| \phi \rangle$. We have also used Eq. (136) and the fact that $U$ and $\Pi_{\pm}$ commute. Similarly, we can obtain,

$$
\| \Pi_\downarrow U | \uparrow, \psi \rangle \|_2 \geq \sqrt{1 - 4 \epsilon^2}.
$$

(144)

Using Eqs. (143) and (144), we can now bound the z-polarization of the electron starting from an initial state which is polarized. First, we note that the operator $S_z$ can be written in terms of the projection operators $\Pi_\uparrow$ and $\Pi_\downarrow$,

$$
S_z = \frac{\hbar}{2} (\Pi_\uparrow - \Pi_\downarrow) \otimes \mathbb{I}_I.
$$

(145)
Using Eq. (144), we find that if the initial state of the system is given by $|\uparrow, \psi\rangle$, then the z-polarization at an arbitrary time $t$ is bounded by,

$$\langle \uparrow, \psi | U^\dagger S_z U | \uparrow, \psi \rangle = \frac{\hbar}{2} \left( \langle \uparrow, \psi | U^\dagger \Pi_\uparrow U | \uparrow, \psi \rangle - \langle \uparrow, \psi | U^\dagger \Pi_\downarrow U | \uparrow, \psi \rangle \right)$$

$$\geq \frac{\hbar}{2} (1 - 8\epsilon^2). \quad \text{(146)}$$

QED

Proof of Theorem 5. From the statement of Theorem 5, we have defined $\Delta E_{max} = \alpha \Delta E, \alpha > 1$. We have also defined the set of eigenstates $M$ such that for every eigenstate $k$ in $M$, we have $|E_k - \langle E\rangle| > \Delta E_{max}$. Then we can write

$$\langle (\Delta E)^2 \rangle = \sum_k p_k (E_k - \langle E\rangle)^2$$

$$= \sum_{k \in M} p_k (E_k - \langle E\rangle)^2 + \sum_{k \notin M} p_k (E_k - \langle E\rangle)^2$$

$$\geq \sum_{k \in M} p_k (E_k - \langle E\rangle)^2$$

$$\geq \sum_{k \in M} p_k (\Delta E_{max})^2$$

$$= (\Delta E_{max})^2 \sum_{k \in M} p_k$$

$$\geq \sum_{k \in M} p_k \leq \frac{1}{\alpha^2} \quad \text{(154)}$$

QED

Proof of Theorem 6.

$$|\langle \phi | U | \phi \rangle| = \left| \sum_k p_k \exp \frac{iE_k t}{\hbar} \right|$$

$$= \left| \sum_{k \notin M} p_k \exp \frac{iE_k t}{\hbar} \right| + \left| \sum_{k \in M} p_k \exp \frac{iE_k t}{\hbar} \right|$$

$$\geq \left| \sum_{k \notin M} p_k \exp \frac{iE_k t}{\hbar} \right| - \left| \sum_{k \in M} p_k \exp \frac{iE_k t}{\hbar} \right|$$

$$\geq \exp \frac{i \langle E \rangle t}{\hbar} \sum_{k \notin M} p_k \exp \frac{i(E_k - \langle E\rangle)t}{\hbar} - \frac{1}{\alpha^2}$$

$$\geq \left| \sum_{k \notin M} p_k \exp \frac{i\Delta E_k t}{\hbar} \right| - \frac{1}{\alpha^2}$$

$$\geq \left( \sum_{k \notin M} p_k \cos \frac{\Delta E_k t}{\hbar} \right)^2 + \left( \sum_{k \notin M} p_k \sin \frac{\Delta E_k t}{\hbar} \right)^2 - \frac{1}{\alpha^2}$$

$$\frac{\Delta E_k t}{\hbar}$$

$$\geq \left( \frac{1}{\alpha^2} \right) \cos \frac{\Delta E_k t}{\hbar} - \frac{1}{\alpha^2}$$

$$\geq \sum_{k \notin M} p_k \cos \frac{\Delta E_k t}{\hbar} - \frac{1}{\alpha^2}$$
where the last inequality holds provided that,

\[ t < \frac{\hbar \pi}{2\alpha \Delta E}. \]  

QED

**Proof of Theorem 7.** Since \( |\psi_i^-\rangle \) is an eigenstate of \( H \), its evolution will correspond to multiplication by some overall phase \( \exp i\omega_{\psi,i} t \). Then the remainder of the proof is trivial

\[
U \left| \downarrow, \psi^-_{\psi,i} \right\rangle = U \left| \psi^-_{\psi,i} \right\rangle - \left| \uparrow, \psi^-_{\psi,i} \right\rangle
\]

\[ = e^{i\omega_{\psi,i} t} \left| \downarrow, \psi^-_{\psi,i} \right\rangle + \left( e^{i\omega_{\psi,i} t} - U \right) \left| \uparrow, \psi^-_{\psi,i} \right\rangle. \]

QED

**Proof of Theorem 8.** Let \( E_i^- \) be the eigenvalue corresponding to \( |\psi_i^-\rangle \). Let us consider the normalized trial state,

\[ |\phi_i\rangle = \left| \uparrow, \psi^-_{\psi,i} \right\rangle + \left| \downarrow, \phi_{\psi,i} \right\rangle \]  

where

\[ |\phi_{\psi,i}\rangle = \frac{1}{-E_i^- - H_0} V \left| \uparrow, \psi^-_{\psi,i} \right\rangle. \]

We select this trial state because it has the desirable property that \( \Pi_\theta |\phi_i\rangle = \left| \uparrow, \psi^-_{\psi,i} \right\rangle \). Our selection of the spin-down component of \( |\phi\rangle \) is designed to minimize the overall energy width of \( |\phi\rangle \). We then obtain,

\[
H |\phi_i\rangle = (H_0 + V) \left( \left| \uparrow, \psi^-_{\psi,i} \right\rangle + \frac{1}{-E_i^- - H_0} V \left| \uparrow, \psi^-_{\psi,i} \right\rangle \right)
\]

\[ = H_0 \left| \uparrow, \psi^-_{\psi,i} \right\rangle - E_i^- \frac{1}{-E_i^- - H_0} V \left| \uparrow, \psi^-_{\psi,i} \right\rangle + V \frac{1}{-E_i^- - H_0} \left| \uparrow, \psi^-_{\psi,i} \right\rangle \]

\[ = -R_\pi H_0 \left| \downarrow, \psi^-_{\psi,i} \right\rangle - E_i^- \frac{1}{-E_i^- - H_0} V \left| \uparrow, \psi^-_{\psi,i} \right\rangle + V \frac{1}{-E_i^- - H_0} \left| \uparrow, \psi^-_{\psi,i} \right\rangle \]

\[ = -E_i^- |\phi_i\rangle + R_\pi V \frac{1}{-E_i^- - H_0} VR_\pi \left| \uparrow, \psi^-_{\psi,i} \right\rangle + V \frac{1}{-E_i^- - H_0} \left| \uparrow, \psi^-_{\psi,i} \right\rangle \]

where we have used the fact that \( R_\pi H_0 R_\pi = -H_0 \). If we define the operator

\[ W = \Pi_\theta \left( R_\pi V \frac{1}{-E_i^- - H_0} VR_\pi + V \frac{1}{-E_i^- - H_0} \right) \]

then we can evaluate the expectation value,

\[ \langle \phi_i | H | \phi_i \rangle = -E_i^- + \langle \phi_i | W | \phi_i \rangle \]

\[ \langle \phi_i | H^2 | \phi_i \rangle = (E_i^-)^2 - 2E_i^- \langle \phi_i | W | \phi_i \rangle + \langle \phi_i | W^2 | \phi_i \rangle \]

\[ \langle (\Delta H)^2 \rangle = \langle \phi_i | W^2 | \phi_i \rangle - \langle \phi_i | W | \phi_i \rangle^2 \]

These expectation values can be bounded using the L2 norm of \( W \) (see Eq. (110) for a definition of the L2 matrix norm). To obtain this norm, we write \( W \) in the \( |z\) basis,

\[ W = W_{diag} + W_{off-diag}. \]

Here we define \( W_{diag} \) as the diagonal portion of \( W \) in the \( \{|z\}\) basis,

\[ W_{diag} = \sum_z \sum_{i,k} \left( \frac{\hbar^4 A_{ij}^2}{4 (E_i^- + E_{\phi,i} + h^2 A_{jk})} - \frac{\hbar^4 A_{ij}^2}{4 (E_i^- + E_{\phi,i} + h^2 A_{jk}/2)} \right) |z\rangle \langle z| \]
where \( j \) sums over nuclei for which \( z_j = -1 \) and \( k \) sums over nuclei such that \( z_k = +1 \). \( W_{\text{off-diag}} \) is the off-diagonal portion of \( W \),

\[
W_{\text{off-diag}} = \sum_{\mathbf{z}, \mathbf{z}'} \left( \frac{\hbar^4 A_j A_k}{4 \left( E_i^- + E_{0,x}^\parallel + \hbar^2 A_i/2 \right)} - \frac{\hbar^4 A_j A_k}{4 \left( E_i^- + E_{0,z}^\parallel + \hbar^2 A_j/2 \right)} \right) |\mathbf{z}'\rangle \langle \mathbf{z}| \tag{179}
\]

\[
= \sum_{\mathbf{z}, \mathbf{z}'} \left( \frac{\hbar^4 A_j A_k (A_j - A_k)}{8 \left( E_i^- + E_{0,x}^\parallel - \hbar^2 A_i/2 \right)} \left( E_i^- + E_{0,z}^\parallel + \hbar^2 A_j/2 \right) \right) |\mathbf{z}'\rangle \langle \mathbf{z}| \tag{180}
\]

where \( \mathbf{z} \) and \( \mathbf{z}' \) are nuclear spin configurations which differ only at nuclear positions \( j \) and \( k \) such that that \( z_j = -1 \), \( z_k = +1 \) and \( z_j' = +1 \), \( z_k' = -1 \). We note that the two terms in Eq. (179) are nearly equal, leading to a significant cancellation in Eq. (180). This cancellation is a result of the spin echo experiment, and will be discussed in Sec. IV.

Because \( \hbar^2 A_j, \hbar^2 A_k \) are negligible relative to \( E_i^- + E_{0,x}^\parallel \), we can neglect these terms in the denominator. We also note that \( |E_i^- + E_{0,z}^\parallel| \geq |E_{\text{max}}^- + E_{0,z}^\parallel| \). Then using the inequality in Eq. (112), we can bound \( \|W\|_2 \) as

\[
\|W\|_2 \leq \frac{\hbar^4}{4 \left( E_{\text{max}}^- + E_{0,z}^\parallel \right)} A_{2,\text{max}} + \frac{\hbar^6}{4 \left( E_{\text{max}}^- + E_{0,z}^\parallel \right)} A_{2,\text{max}} A_{\text{max}} \tag{181}
\]

The second term in this equality is strictly less than the first term by a factor of \( \epsilon \) (see Eqs. (43) and (44)). So we can write the strict inequality,

\[
\|W\|_2 \leq \frac{\hbar^4}{4 \left( E_{\text{max}}^- + E_{0,z}^\parallel \right)} A_{2,\text{max}} (1 + \epsilon) \tag{182}
\]

Hence it follows from Eqs. (174) and (175) and Eq. (182) that

\[
\langle \phi | H^2 | \phi \rangle - \langle \phi | H | \phi \rangle^2 \leq \frac{\hbar^4}{4 \left( E_{\text{max}}^- + E_{0,z}^\parallel \right)} A_{2,\text{max}} (1 + \epsilon) \tag{183}
\]

QED

Proof of Theorem 9. Since we know the energy width of the trial state \( |\phi_i\rangle \), we can apply Theorem 6 to characterize its dynamics

\[
U |\phi_i\rangle = e^{i \omega_{\hat{\phi},i} t} \sqrt{1 - \lambda_i^2} |\phi_i\rangle + \lambda_i |r_{\hat{\phi},i}^-\rangle. \tag{184}
\]

In Eq. (184), \( \omega_{\hat{\phi},i} \) is an overall phase corresponding to \( \theta \) in Theorem 6. \( \lambda_i \) is the magnitude of the residual evolution, and is bounded by Eq. (60). In order to characterize the evolution of \( |\uparrow, \psi_{\phi,i}^-\rangle \), we then note that

\[
|\uparrow, \psi_{\phi,i}^-\rangle = |\phi_i\rangle - |\downarrow, \phi_{\phi,i}\rangle. \tag{185}
\]

Using similar reasoning as in the proof for Theorem 7, we obtain

\[
U \left| \uparrow, \psi_{\phi,i}^- \right\rangle = U \left( |\phi_i\rangle - |\downarrow, \phi_{\phi,i}\rangle \right) \tag{186}
\]

\[
= e^{i \omega_{\hat{\phi},i} t} \sqrt{1 - \lambda_i^2} \left| \uparrow, \psi_{\phi,i}^- \right\rangle + \lambda_i \left| r_{\hat{\phi},i}^- \right\rangle + \left( \sqrt{1 - \lambda_i^2} e^{i \omega_{\hat{\phi},i} t} - U \right) |\downarrow, \phi_{\phi,i}\rangle \tag{187}
\]

The final element in our proof is to demonstrate the three inequalities in Eqs. (85–87). Eq. (85) follows directly from Theorem 6. Eq. (86) follows from the fact that we can bound the magnitude of \( |\uparrow, \phi_{\phi,i}\rangle \),

\[
\langle \downarrow, \phi_{\phi,i} | \downarrow, \phi_{\phi,i} \rangle = \left| \langle \uparrow, \psi_{\phi,i}^- | V \frac{1}{(-E_i^- - H_0)^2} V |\uparrow, \psi_{\phi,i}^- \rangle \right|^2 \leq \epsilon^2. \tag{188}
\]
Finally, Eq. (87) follows from the consideration that (from Theorem 6), \( \langle r^-_{\hat{\Phi}, i} | \phi_i \rangle = 0 \). Then,

\[
\left| \langle r^-_{\hat{\Phi}, i} | \psi^-_{\psi, i} \rangle \right| = \left| \langle r^-_{\hat{\Phi}, i} | \phi_i \rangle - \langle r^-_{\hat{\Phi}, i} | \psi, \phi^-_{\psi, i} \rangle \right| \leq \| r^-_{\hat{\Phi}, i} \| \| \psi, \phi^-_{\psi, i} \| \leq \epsilon.
\]

QED