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Non-Markovian Dynamics of a Localized Electron Spin Due to the Hyperfine Interaction

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Abstract. We review our theoretical work on the dynamics of a localized electron spin interacting with an environment of nuclear spins. Our perturbative calculation is valid for arbitrary polarization p of the nuclear spin system and arbitrary nuclear spin I in a sufficiently large magnetic field. In general, the electron spin shows rich dynamics, described by a sum of contributions with exponential decay, nonexponential decay, and undamped oscillations. We have found an abrupt crossover in the long-time spin dynamics at a critical shape and dimensionality of the electron envelope wave function. We conclude with a discussion of our proposed scheme to measure the relevant dynamics using a standard spin–echo technique.

Key Words: ESR, hyperfine interaction, nuclear spins, quantum computing, quantum dots, spin-echo, spintronics.

1. Introduction

Experiments with trapped ions [1] and nuclear magnetic resonance (NMR) [2] have proven that the basic elements of a quantum computer can be realized in practice. To progress beyond proof-of-principle experiments, the next generation of quantum information processors must overcome significant obstacles regarding scalability and decoherence. The scalability issue is largely solved by proposals for a solid-state implementation for quantum computing, where established fabrication techniques can be used to multiply the qubits and interface them with existing electronic devices. Due to their relative isolation from the surrounding environment, single electron spins in semiconductor quantum dots are expected to be exceptionally robust against decoherence [3]. Indeed, longitudinal relaxation times in these systems have been measured to be $T_1 = 0.85$ ms in a magnetic field of 8 T [4] and in GaAs quantum wells, the transverse dephasing time T_2^* for an ensemble of electron spins (which typically provides a lower bound for the intrinsic decoherence time T_2 of an isolated spin) has been measured to be in excess of 100 ns [5].

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Possible sources of decoherence for a single electron spin confined to a quantum dot are spin–orbit coupling and the contact hyperfine interaction with the surrounding nuclear spins [6]. The relaxation rate due to spin–orbit coupling $1/T_1$ is suppressed for localized electrons at low temperatures [7] and recent work has shown that T_2, due to spin–orbit coupling, can be as long as T_1 under realistic conditions [8]. However, since spin-carrying isotopes are common in the semiconductor industry, the contact hyperfine interaction (in contrast to the spin–orbit interaction) is likely an unavoidable source of decoherence, which does not vanish with decreasing temperature or carefully chosen quantum dot geometry [9].

In the last few years, a great deal of effort has been focused on a theoretical description of interesting effects arising from the contact hyperfine interaction for a localized electron. The predicted effects include a dramatic variation of T_1 with gate voltage in a quantum dot near the Coulomb blockade peaks or valleys [10], all-optical polarization of the nuclear spins [11], use of the nuclear spin system as a quantum memory [12, 13], and several studies of spin dynamics [14–21]. Here, our system of interest is an electron confined to a single GaAs quantum dot, but this work applies quite generally to other systems, such as electrons trapped at shallow donor impurities in Si:P [9].

An exact solution for the electron spin dynamics has been found in the special case of a fully polarized initial state of the nuclear spin system [14, 22]. This solution shows that the electron spin only decays by a fraction $\propto \frac{1}{N}$ of its initial value, where N is the number of nuclear spins within the extent of the electron wave function. The decaying fraction was shown to have a nonexponential tail for long times, which suggests non-Markovian (history dependent) behavior. For an initial nuclear spin configuration that is not fully polarized, no exact solution is available and standard time-dependent perturbation theory fails [14]. Subsequent exact diagonalization studies on small spin systems [23] have shown that the electron spin dynamics are highly dependent on the type of initial nuclear spin configuration. The unusual (nonexponential) form of decay, and the fraction of the electron spin that undergoes decay may be of interest in quantum error correction (QEC) since QEC schemes typically assume exponential decay to zero.

In the following section we describe a systematic perturbative theory of electron spin dynamics under the action of the Fermi contact hyperfine interaction. Further details of this work can be found in reference [24].

2. Model and perturbative expansion

We investigate electron spin dynamics at times shorter than the nuclear dipole–dipole correlation time τ_{dd} ($\tau_{dd} \approx 10^{-4}$ s in GaAs is given directly by the inverse width of the nuclear magnetic resonance (NMR) line [25]). At these

time scales, the relevant Hamiltonian for a description of the electron and nuclear spin dynamics is that for the Fermi contact hyperfine interaction:

$$H = H_0 + V_{ff}, \tag{1}$$

$$H_0 = (b + h_z)S_z; \;\; V_{ff} = \frac{1}{2}(S_+h_- + S_-h_+), \tag{2}$$

where $\mathbf{S}$ is the electron spin operator, b gives the electron Zeeman splitting and $\mathbf{h} = \sum_k A_k \mathbf{I}_k$ is the quantum nuclear field. $S_\pm = S_x \pm iS_y$ and $h_\pm = h_x \pm ih_y$ are defined in the usual way. The hyperfine coupling constants are $A_k = Av_0 |\psi(\mathbf{r}_k)|^2$ where v_0 is the volume of a crystal unit cell containing one nuclear spin, $\psi(\mathbf{r})$ is the electron envelope wave function, and A is the strength of the hyperfine coupling. While the total number of nuclear spins in the system, N_{tot}, may be very large, there are fewer spins ($N < N_{\text{tot}}$) within the extent of the electron wave function. In GaAs, all naturally occurring isotopes carry spin $I = \frac{3}{2}$. In bulk GaAs, A has been estimated [25] to be $A = 90\ \mu eV$. This estimate is based on an average over the hyperfine coupling constants for the three nuclear isotopes ^{69}Ga, ^{71}Ga, and ^{75}As, weighted by their relative abundances. Natural silicon contains 4.7% ^{29}Si, which carries $I = \frac{1}{2}$, and 95% ^{28}Si, with $I = 0$. An electron bound to a phosphorus donor impurity in natural Si:P interacts with $N \approx 10^2$ surrounding ^{29}Si nuclear spins, in which case the hyperfine coupling constant is on the order of $A \approx 0.1\ \mu eV$ [9].

For large magnetic fields b, the flip–flop processes due to V_{ff} are suppressed by the electron Zeeman splitting, and it is reasonable to take $H \approx H_0$ (zeroth order in V_{ff}). The zeroth-order problem is algebraically simple, and leads to some interesting conclusions regarding the initial conditions. In this limit the longitudinal spin is time-independent, since $[S_z, H_0] = 0$, but the transverse spin can undergo nontrivial evolution. Assuming uniform hyperfine coupling constants ($A_k = A/N$) and nuclear spin $I = 1/2$ we evaluate the transverse electron spin dynamics for a nuclear spin bath of polarization p along the z-axis and two different nuclear spin initial states. When the A_k are uniform, the transverse spin exhibits periodic dynamics. However, at times much less than the period, given by the inverse level spacing ($t \ll N/A$, setting $\hbar = 1$), we find

$$\langle S_+ \rangle_t^{\text{unprep.}} = \langle S_+ \rangle_0 e^{i\omega_n t} e^{-t^2/2t_c^2}, \;\; t_c = \frac{2}{A}\sqrt{\frac{N}{1-p^2}}, \tag{3}$$

$$\langle S_+ \rangle_t^{\text{prep.}} = \langle S_+ \rangle_0 e^{i\omega_n t}, \;\; \omega_n = b + pA/2. \tag{4}$$

In Equation (3), the nuclear spin system is "unprepared." This state corresponds to either a translationally invariant direct-product state of the form $\Pi_k(\sqrt{f_\uparrow}\,|\uparrow\rangle_k + \sqrt{1-f_\uparrow}\,|\downarrow\rangle_k)$, where $p = 2f_\uparrow + 1$, or to a statistical mixture of

product states of the form $|\uparrow\downarrow\uparrow\uparrow\cdots\rangle$, with average polarization p. In contrast, Equation (4) corresponds to the case when the nuclear system has been "prepared" in an eigenstate $|n\rangle$ of the operator $h_z : h_z \mid n \rangle = [h_z]_{nn} \mid n \rangle$, ($[h_z]_{nn} = pA/2$). It is important to note that the decay of the 'unprepared' state is reversible. This decay can be recovered by performing a spin–echo measurement on the electron spin, since a π-rotation of the electron spin $S_z \to -S_z$ reverses the sign of H_0: $H_0 \to -H_0$, and results in time-reversed dynamics. When the hyperfine coupling constants are allowed to vary in space, higher-order corrections in V_{ff} will, however, result in irreversible decay. In the following, we generalize to nonuniform A_k and arbitrary nuclear spin I to evaluate this decay for an initial nuclear state that is an eigenstate of the h_z-operator.

An exact generalized master equation (GME) can be derived for the electron spin operators beginning from the von Neumann equation for the full density operator ($\dot{\rho} = -i[H, \rho]$) [26]. The resulting GME is expanded in terms of V_{ff} through direct application of the Dyson identity. We find, quite remarkably, that the equations of motion for the longitudinal ($\langle S_z \rangle_t$) and transverse ($\langle S_+ \rangle_t = \langle S_x \rangle_t + i\langle S_y \rangle_t$) spin are decoupled to *all orders* in V_{ff} and take the form

$$\left\langle \dot{S}_z \right\rangle_t = N_z(t) - i \int_0^t dt' \sum\nolimits_{zz} (t - t') \langle S_z \rangle_{t'}, \tag{5}$$

$$\left\langle \dot{S}_+ \right\rangle_t = i\omega_n \langle S_+ \rangle_t - i \int_0^t dt' \sum\nolimits_{++} (t - t') \langle S_+ \rangle_{t'}. \tag{6}$$

These integro-differential equations can be converted to a pair of algebraic equations by Laplace transformation $f(s) = \int_0^\infty ds e^{-st} f(t)$, Re[s] > 0. The algebraic equations yield

$$S_z(s) = \frac{\langle S_z \rangle_0 + N_z(s)}{s + i\sum_{zz}(s)}; \quad S_+(s) = \frac{\langle S_+ \rangle_0}{s - i\omega_n + i\sum_{++}(s)}. \tag{7}$$

The numerator term $N_z(s)$ and self-energy $\sum_{zz}(s)$ are related to the self-energy matrix elements for spin-up and spin-down by $N_z(s) = -\frac{i}{2s}\left(\sum_{\uparrow\uparrow}(s) + \sum_{\uparrow\downarrow}(s)\right)$ and $\sum_{zz}(s) = \sum_{\uparrow\uparrow}(s) - \sum_{\uparrow\downarrow}(s)$. All self-energy matrix elements are written in terms of an expansion in powers of $V_{ff} : \sum_{\alpha\beta}(s) = \sum_{\alpha\beta}^{(2)}(s) + \sum_{\alpha\beta}^{(4)}(s) + \cdots, \alpha, \beta = (+, \uparrow, \downarrow)$. For a sufficiently large Zeeman splitting b we find that all higher-order self-energy matrix elements are suppressed by a smallness parameter Δ:

$$\sum\nolimits_{\alpha\beta}^{(2(k+1))}(s) \propto \Delta^k; \ \Delta = \frac{A}{2(b + pIA)}. \tag{8}$$

In Born approximation (second order in the flip–flop terms V_{ff}), and for an initial nuclear spin system with uniform polarization, we find

$$\sum\nolimits_{\uparrow\uparrow}(s) \approx \sum\nolimits^{(2)}_{\uparrow\uparrow}(s) = -iNc_{+}[I_{+}(s-i\omega_n)+I_{-}(s+i\omega_n)], \tag{9}$$

$$\sum\nolimits_{\uparrow\downarrow}(s) \approx \sum\nolimits^{(2)}_{\uparrow\downarrow}(s) = iNc_{-}[I_{-}(s-i\omega_n)+I_{+}(s+i\omega_n)], \tag{10}$$

$$\sum\nolimits_{++}(s) \approx \sum\nolimits^{(2)}_{++}(s) = -iN[c_{-}I_{+}(s)+c_{+}I_{-}(s)], \tag{11}$$

$$c_{\pm} = I(I+1) - \frac{1}{N_{\text{tot}}}\sum_k \left\langle I^z_k\left(I^z_k \pm 1\right)\right\rangle_0, \tag{12}$$

$$I_{\pm}(s) = \frac{1}{4N}\sum_k \frac{A_k^2}{s \mp i\frac{A_k}{2}} \approx \frac{d}{m}\int_0^1 dx \frac{x|\ln x|^v}{s \mp ix},\ v = \frac{d}{m} - 1. \tag{13}$$

In evaluating Equation (13), we have assumed the electron is in its orbital ground state, described by an isotropic envelope wave function of the form $\psi(r_k) = \psi(0)\exp\left[-\frac{1}{2}\left(\frac{r_k}{l_0}\right)^m\right]$. The index k gives the number of nuclear spins within radius r_k of the origin and N is the number of nuclear spins within radius l_0, so in d dimensions $\left(\frac{r_k}{l_0}\right)^d = \frac{k}{N}$. From the definition $A_k \propto \mid \psi(r_k) \mid^2$ this gives the coupling constants $A_k = 2\exp\left[-\left(\frac{k}{N}\right)^{\frac{m}{d}}\right]$ in units where $A_0/2 = 1$. For times $t \lesssim N^{3/2}/A$, it is strictly valid to take $N_{\text{tot}} \to \infty$ and change the sum to an integral in Equation (13), [24], which gives the final result for $I_{\pm}(s)$, above.

The time-dependent spin expectation values can now be recovered from the Laplace transform expressions by evaluating the Bromwich inversion integral:

$$\langle S_z\rangle_t = \frac{1}{2\pi i}\int_{C_B} ds e^{st} S_z(s);\ \langle S_+\rangle_t = \frac{1}{2\pi i}\int_{C_B} ds e^{st} S_+(s). \tag{14}$$

To evaluate these integrals we close the contour in the negative real half-plane, avoiding all branch cuts of the functions $S_z(s)$; $S_+(s)$, and apply the residue theorem. The relevant contour is shown in Figure 1 for $S_+(s)$ or $S_z(s-i\omega_n)$ within Born approximation when $d = m = 2$ (this applies to a two-dimensional quantum dot with parabolic confinement). The spin expectation values that result from this procedure are the sum of contributions with undamped oscillations (from poles on the imaginary axis), exponential decay (from poles with finite negative real part) and nonexponential long-time tails (from branch cut integrals). Since the contributions from poles on the imaginary axis do not decay, the spin expectation

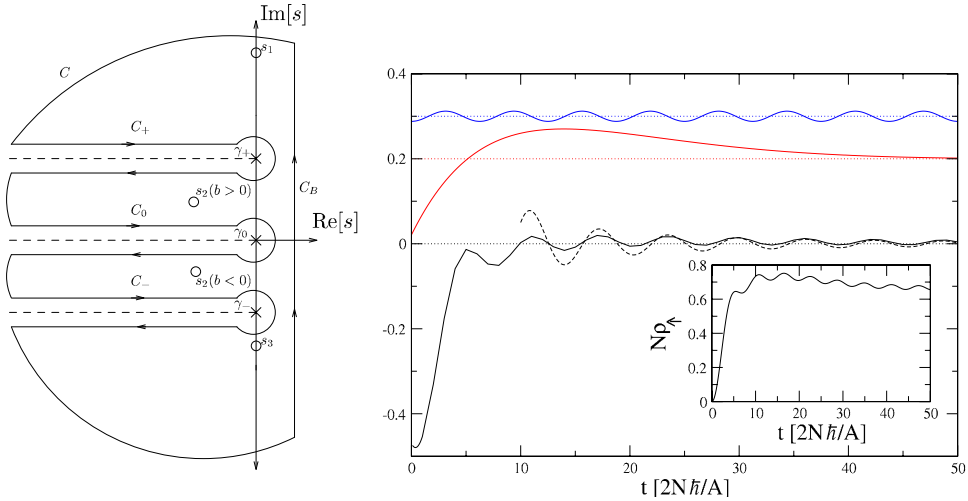

Figure 1. Contour integral (*left*) that determines all contributions (*right*) to the spin expectation values, including undamped oscillations (*right*, *top*), exponential decay (*right*, *middle*) and nonexponential decay (*right*, *bottom*; *solid line*: numerical integration, *dashed line*: analytic asymptotic expression). All three contributions are added to obtain $\rho_\uparrow(t) = 1/2 + \langle S_z \rangle_t$ (*right*, inset). For these calculations we have chosen $d = m = 2$, $I = 1/2$, the initial condition $\rho_\uparrow(0) = 0$, and values of b and p that correspond to $\Delta = 10/11$.

values (in a suitable co-rotating frame) are given generically in terms of a constant piece, and a remainder with nontrivial dynamics:

$$\langle S_X \rangle_t = \langle S_X \rangle_0 + R_X(t), \quad X = +, z. \tag{15}$$

We show the different contributions to $R_z(t)$ in Figure 1 for $d = m = 2$ in the weakly perturbative regime (where $\Delta \lesssim 1$).

In the strongly perturbative regime (when $\Delta \ll 1$), the time dependence of $R_X(t)$ is given exclusively in terms of the functions $I_\pm(t) = \frac{1}{2\pi i}\int_{C_B} ds e^{st} I_\pm(s)$ that appear in Equation (13), above. There is an abrupt crossover in the long-time behavior of these functions at a critical value of d/m. For $d/m < 2$, the major contributions to $I_\pm(t \gg N/A)$ come from the upper limit of the integral in Equation (13), corresponding to nuclear spins near the center of the quantum dot. This leads to a modulation of the spin dynamics at a frequency $A/2N$. When $d/m \geq 2$, the major contributions come from the lower limit, corresponding to nuclear spins far from the dot center. In this case, the long-time dynamics are smoothly varying, with no modulation:

$$R_X(t) \sim I_\pm(t \gg N/A) \propto \begin{cases} \left(\frac{1}{t}\right)^{d/m} e^{\pm i\frac{A}{2N}t}, \nu = \frac{d}{m} - 1 < 1 \\ \frac{\ln^\nu t}{t^2}, \nu = \frac{d}{m} - 1 \geq 1 \end{cases}. \tag{16}$$

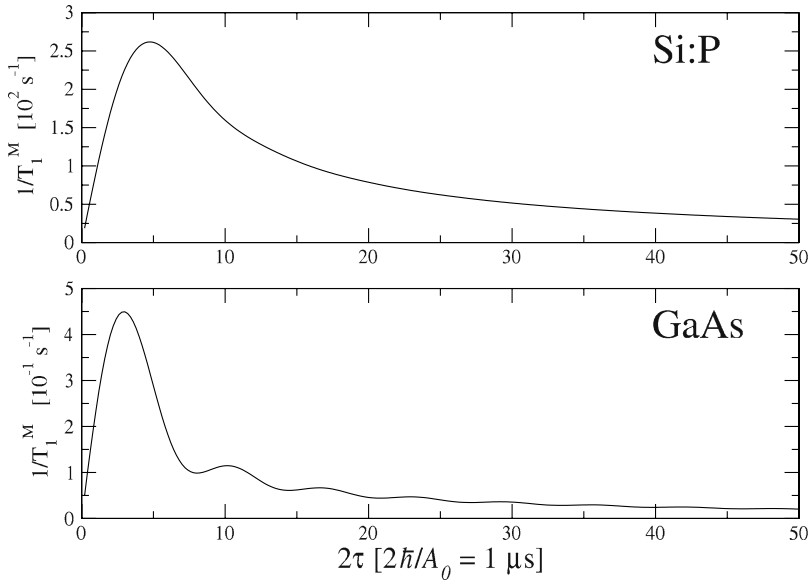

Figure 2. Longitudinal-spin decay rate of the CPMG echo envelope ($1/T_1^{\mathrm{M}} \propto R_z(2\tau)/2\tau$) as a function of the free evolution time 2τ between π-pulses. We plot the results for an electron bound to a phosphorus donor, where $N = 10^2$ (*top*) and a two-dimensional GaAs quantum dot with $N = 10^5$ (*bottom*).

The difference in these two cases should be visible in a spin–echo experiment that uses a Carr–Purcell–Meiboom–Gill (CPMG) spin–echo sequence: $\frac{\pi}{2} - (\tau - \pi_x - \tau - \mathrm{ECHO} - \tau - \pi_{-x} - \tau - \mathrm{ECHO})_{\mathrm{repeat}}$. We consider the strongly perturbative limit ($\Delta \ll 1$), and to resolve the relevant dynamics, the time between π-pulses must satisfy $\tau \ll \sqrt{\delta}\tau_{dd}$, where $\delta = \Delta^2/N$, and τ_{dd} is the nuclear spin dipolar correlation time [24]. Under these conditions the CPMG echo envelope decay rate as a function of τ is determined exclusively by the remainder term according to $1/T_1^{\mathrm{M}} \propto R_z(2\tau)/2\tau$ for the longitudinal component and $1/T_2^M \propto Re[R_+(2\tau)]/2\tau$ for the transverse components. We plot the CPMG decay rate as a function of 2τ in Figure 2 for two systems of interest. For an electron trapped at a donor impurity in bulk silicon, $d = 3$ and the orbital wave function is exponential ($m = 1$). This corresponds to $\nu = 2$ in Equation (16). In a two-dimensional GaAs quantum dot ($d = 2$) with parabolic confinement, the ground-state orbital electron wave function is a Gaussian ($m = 2$), which corresponds to $\nu = 0$ in Equation (16).

3. Conclusions

We have reviewed our theoretical description for the dynamics of a localized electron spin interacting with a nuclear spin environment. We have predicted a

sharp crossover in the relevant dynamics at a critical value of the dimensionality and form of the electron envelope wave function, and have described a standard method that could be used to reveal the relevant dynamics. We stress that the electron spin dynamics are in general very rich, described by contributions with exponential decay, nonexponential decay and undamped oscillations. Furthermore, this work may have profound implications for the future of spin-based solid-state quantum information processing and quantum error correction, where previous studies have assumed exponential decay to zero.

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