

Electron spin dynamics due to hyperfine coupling in quantum dots

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The dynamics of spins in semiconductor quantum dots often is controlled by their hyperfine coupling to nuclear spins. We develop a straightforward and efficient approach to describe the dynamics and the effective decoherence of the electron spins due to hyperfine coupling in realistic quantum dots. Systems with a large number of nuclei and an arbitrary initial nuclear polarization for which the number of nuclei initially flipped over is much less than the total number of nuclei are treated. This treatment employs a pole approximation within a Schrödinger equation of motion for the state of the coupled electron and nuclear spin system, and it allows us to treat systems with arbitrary initial conditions. We find that typical time scales for the effective spin decoherence are on the order of tens of microseconds.

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The spin degree of freedom of electrons is of considerable interest in contexts ranging from “spin electronic” devices to implementations for quantum bits (qubits) in quantum computing. A major obstacle in these applications is decoherence due to coupling of the spin to its environment. For example, a spin in a quantum dot (QD) is an attractive candidate for a qubit, but the spin decoherence time must be longer than the times needed to manipulate the spins in gate operations.

Sources of spin decoherence in quantum dots include phonon induced modulations of spin-orbit coupling^{1,2} and hyperfine coupling to nuclei.^{3,4} Recent work on spin relaxation due to phonon scattering gives relatively long spin relaxation times at low temperatures.^{1,2} For sufficiently high magnetic fields, electron spin lifetimes on the order of milliseconds have been measured.⁵ At low magnetic fields, however, the hyperfine interaction between the electron spin and the unpaired nuclear spins typically present in III-V semiconductor quantum dots provides an unavoidable source of losses and dominates the spin decoherence in dots. The effective decoherence in these systems arises from variations of hyperfine coupling across the dot. Although strictly speaking the coupled electron-nuclear spin system evolves coherently in time, coupling to a large number of nuclei gives an effective decoherence in realistic laboratory times. QDs have large numbers of nuclei (10^4 – 10^6) with varying initial nuclear polarizations. Each of these two features presents formidable difficulties in calculating the spin dynamics of these systems.

An approach for the electron spin dynamics for the case of a fully polarized nuclear system has been given,³ and numerical simulations for models of QDs with very small numbers of nuclei ($N \leq 20$) have been made.^{4,6,7} In the case where the external magnetic field is much larger than the hyperfine coupling, the relaxation times can be described by a standard perturbative approach.⁸ A master equation approach also has been developed in which the hyperfine interaction is switched on adiabatically.⁹ This method is applicable to starting conditions for which the electron and spin systems are decoupled, which is generally not the case for an electron spin interacting with nuclei in solids. Recently, an

analytical model based on the Heisenberg equation of motion for high magnetic fields and for the nuclear interaction switched on adiabatically has also been given.¹⁰

Here, we present a straightforward method to describe the spin dynamics in realistic QDs with large numbers of nuclei and with varying initial polarizations. It is based on a Schrödinger equation of motion approach for the wave function describing the system of coupled electron and nuclear spins, and it uses a pole approximation that is applicable for systems with large numbers of nuclei. It provides an efficient method for calculations, and it allows us to treat systems with initially coupled electronic and nuclear systems.

We consider the spin s of an electron in the conduction band in a QD coupled to the nuclear spins $\mathbf{I}_i$ via the hyperfine Fermi contact interaction. For simplicity we take the nuclear spins to have magnitude of $1/2$. An applied magnetic field $\mathbf{B}$ along the z direction gives a Zeeman splitting, and the total Hamiltonian is

$$H = (\epsilon + \gamma_z)s_z + \gamma_-s_+ + \gamma_+s_-, \quad (1)$$

$$\gamma_z = \sum_i A_i I_{i,z}, \quad \gamma_{-(+)} = \sum_i \frac{A_i}{2} I_{i,-(+)} \quad (2)$$

Here, $\epsilon = \mu_B g B$ where μ_B is the Bohr magneton and g is the Zeeman g factor. A_i is the hyperfine coupling of the electron spin in the QD to a nucleus where $A_i = A v_0 |\psi(\mathbf{r}_i)|^2$ and A is the total strength of the hyperfine coupling in a unit cell. $v_0 = 1/n_0$ is the inverse density of nuclear spins, and $\psi(\mathbf{r}_i)$ is the electron wave function at the nuclear position $\mathbf{r}_i$. Thus, A_i varies over the dot, and for a dot with N nuclear spins, $A_i \sim A/N$. Dipole-dipole coupling becomes important only at times longer than 10^{-4} s and is not included here.⁸ The nuclear Zeeman interaction is much smaller than that for the electron and is not included.

The time evolution of the electron spin is given by the state vector $|\psi(t)\rangle$ of the coupled electron-nuclear system from $i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = H |\psi(t)\rangle$, where $\hbar$ is taken to be 1. The z component of the total spin of the coupled system, $M_z = s_z$

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$+\sum_{i=1}^N I_{i,z}$, is a conserved quantity, and the total number of eigenstates is 2^{N+1} . For a given direction of the electron spin and for m of the N nuclear spins flipped down, there are

$$N_m = \binom{N}{m}$$

arrangements of the nuclear spins in the QD. The total number of states for a given M_z is $N_{M_z} = N_m + N_{m+1}$, where we see that states with both m and $m+1$ nuclear spins flipped down contribute to the states of a single M_z . Details of the classification and enumeration of the states of this system as well as their use in finding the formal exact solution of the time-dependent Schrödinger equation are given in Ref. 11. An arbitrary state vector of the system can be written as a linear superposition of orthonormal states with different M_z ,

$$|\psi_{M_z(m)}(t)\rangle = \sum_{N_m} Y_{\{m\}}(t) |\downarrow; \{m\}\rangle + \sum_{N_{m+1}} X_{\{m+1\}}(t) |\uparrow; \{m+1\}\rangle. \quad (3)$$

Here, $|\{m\}\rangle = |\{\downarrow\}_{N-m}\rangle$, with $m=0,1,\dots,N$, denotes all arrangements of the m nuclear spins that are flipped down among the total N nuclear spins, and $(\uparrow, \downarrow)$ are the electron spin states. $X_{\{m+1\}}(t)$ and $Y_{\{m\}}(t)$ are the respective time-dependent probability amplitudes.

The solutions are obtained for each value of M_z , and the complete solution is the sum over all states $|\psi(t)\rangle = \sum_{M_z(m)} C_{M_z(m)} |\psi_{M_z(m)}(t)\rangle$. The state is normalized giving

$$1 = \sum_{M_z(m)} |C_{M_z(m)}|^2 \left[\sum_{N_m} |Y_{\{m\}}(t)|^2 + \sum_{N_{m+1}} |X_{\{m+1\}}(t)|^2 \right]. \quad (4)$$

The time dependences of the average electron spin components are

$$\langle s_z(t) \rangle = \frac{1}{2} \sum_{M_z(m)} |C_{M_z(m)}|^2 \left[\sum_{N_{m+1}} |X_{\{m+1\}}(t)|^2 - \sum_{N_m} |Y_{\{m\}}(t)|^2 \right], \quad (5)$$

$$\langle s_+(t) \rangle = \sum_{M_z(m)} \sum_{M_z(m-1)} C_{M_z(m-1)}^* C_{M_z(m)} \sum_{N_m} X_{\{m\}}^*(t) Y_{\{m\}}(t), \quad (6)$$

$$\langle s_-(t) \rangle = \langle s_+(t) \rangle^*, \quad (7)$$

where $s_{\pm} = s_x \pm i s_y$.

It is straightforward to show that for a given M_z , the Schrödinger equation with the Hamiltonian in Eq. (1) gives the following closed set of simultaneous equations for the amplitudes:

$$i\dot{Y}_{\{m\}}(t) = -B_{\{m\}} Y_{\{m\}}(t) + \sum_{j \neq \{m\}} \frac{A_j}{2} X_{\{m+1\},j}(t), \quad (8)$$

$$i\dot{X}_{\{m+1\},j}(t) = B_{\{m+1\},j} X_{\{m+1\},j}(t) + \sum_{i=1}^{\{m\}} \frac{A_i}{2} Y_{\{m+1\},i}(t). \quad (9)$$

Here, $B_{\{m\}} = (\epsilon + A - \sum_{i=1}^{\{m\}} A_i)/2$, $B_{\{m+1\},j} = B_{\{m\}} - A_j/2$, and $A = \sum_{i=1}^N A_i$. The sum in Eq. (8) $\sum_{j \neq \{m\}}^N$ is over all nuclei excluding the ones flipped down for the arrangement $\{m\}$, and the sum in Eq. (9) $\sum_{i=1}^{\{m\}}$ is over all nuclei flipped down for the arrangement $\{m\}$. We have introduced the following notations: $X_{\{m+1\},j}$ is the amplitude associated with the electron spin up and $m+1$ nuclear spins down, which is obtained by flipping one of the $N-m$ up nuclear spins, labeled by j , thus adding one more nuclear spin to the previous m -down spins; $Y_{\{m+1\},i}$ is the amplitude of a state with m nuclear spins down obtained by flipping the i th nuclear down spin to up in the group of $m+1$ down nuclear spins. Equations (8) and (9) are converted to linear simultaneous algebraic equations with general initial conditions at $t=0$ by taking Laplace transforms. The exact solutions for the amplitudes are found as follows:

$$\bar{Y}_{\{m\}}(\omega) = \frac{1}{i\omega + B_{\{m\}} - \sum_{j \neq \{m\}} \frac{(A_j/2)^2}{i\omega - B_{\{m+1\},j}}} \times \left[iY_{\{m\}}(0) + \sum_{l \neq \{m\}}^N \frac{iX_{\{m+1\},l}(0)A_l/2}{i\omega - B_{\{m+1\},l}} + \sum_{i=1}^{\{m\}} \sum_{l \neq \{m\}}^N \frac{A_i A_l \bar{Y}_{\{m+1\},l}(\omega)/4}{i\omega - B_{\{m+1\},l}} \right], \quad (10)$$

$$\bar{X}_{\{m+1\}}(\omega) = \frac{1}{i\omega - B_{\{m+1\}}} \left[iX_{\{m+1\}}(0) + \sum_{i=1}^{\{m\}} \frac{Y_{\{m+1\},i}(0)A_i/2}{i\omega + B_{\{m+1\},i}} + \sum_{i=1}^{\{m\}} \frac{A_i/2}{i\omega + B_{\{m+1\},i}} \sum_{l \neq \{m\}}^N \frac{A_l \bar{X}_{\{m+1\},l}(\omega)}{2} \right]. \quad (11)$$

We now consider the solutions of Eqs. (10) and (11) in time using an inverse Laplace transform. The large number of nuclei precludes obtaining exact results. However, we note that in the case of predominant initial nuclear polarization $m \ll N$ one can make useful approximations for the amplitudes. The set of equations for the amplitudes $\bar{X}_{\{m\}}(\omega)$ and $\bar{Y}_{\{m\}}(\omega)$, and subsequently for $X_{\{m\}}(t)$ and $Y_{\{m\}}(t)$, can actually be given as an infinite series of terms.¹² There are dimensionless contributions in $X_{\{m\}}(t)$ and $Y_{\{m\}}(t)$ proportional to $A_i A_j$ with denominators that are second order in quantities such as $B_{\{m\}}$. For large N and $m \ll N$, these terms are small to the order of $1/N^2$ and we drop them. In addition, there are “self-energy” corrections to quantities such as $B_{\{m\}}$ of order A_i^2 in the denominators of $\bar{X}_{\{m\}}(\omega)$ and $\bar{Y}_{\{m\}}(\omega)$, and we also drop them. Here, we use the fact that the individual $A_i \sim A/N$ are small compared to A , and $\epsilon + A$ dominates when it appears in combination with any subset of A_i when $m \ll N$. In effect, we have replaced a series of poles for the amplitudes $\bar{X}_{\{m\}}(\omega)$ and $\bar{Y}_{\{m\}}(\omega)$ by a single pole, hence, the term “pole approximation” for this approach. Then, the solutions are found to be

$$\begin{aligned}
Y_{\{m\}}(t) &\approx Y_{\{m\}}(0)e^{iB_{\{m\}}t/2} \\
&\times \left[1 - \sum_{j=1}^{\{m\}} \sum_{l \neq \{m\}}^N \frac{A_j A_l}{8(\epsilon + A)(B_{\{m\}} - B_{\{m+1\}/l})} \right] \\
&\times (1 - e^{i(B_{\{m\}} - B_{\{m+1\}/l})t/2}) - X_{\{m+1\}}(0)e^{iB_{\{m\}}t/2} \\
&\times \sum_{l \neq \{m\}}^N \frac{A_l}{4(\epsilon + A)} (1 - e^{-i(B_{\{m\}} + B_{\{m+1\}/l})t/2}), \quad (12)
\end{aligned}$$

$$\begin{aligned}
X_{\{m+1\}}(t) &\approx X_{\{m+1\}}(0)e^{-iB_{\{m+1\}}t/2} \\
&\times \left[1 - \sum_{j=1}^{\{m+1\}} \sum_{l \neq \{m+1\}}^N \frac{A_j A_l}{8(\epsilon + A)(B_{\{m+1\}/l} - B_{\{m+1\}/j})} \right] \\
&\times (1 - e^{-i(B_{\{m+1\}/l} - B_{\{m+1\}/j})t/2}) + Y_{\{m\}}(0)e^{-iB_{\{m+1\}}t/2} \\
&\times \sum_{j=1}^{\{m+1\}} \frac{A_j}{4(\epsilon + A)} (1 - e^{i(B_{\{m\}} + B_{\{m+1\}/j})t/2}). \quad (13)
\end{aligned}$$

We note that the results in Eqs. (12) and (13) give the normalization in Eq. (4) correctly for all frequencies to order A^2 and that these results give the exact result for the short time limit $t \rightarrow 0$. This approach gives accurate results for large N and $m \ll N$ providing that $\epsilon + A$ in the denominator does not become small.

For the purpose of illustrating these results, we choose an initial state in which m spins are flipped down and in which each arrangement of the m nuclear spins is equally likely. The initial probability is taken to be different for the two directions of the electron spin. This can be thought of as the equal probability choice of the starting state and is specified by the initial conditions $Y_{\{m\}}(t=0) = \bar{y}_m / \sqrt{N_m}$ and $X_{\{m\}}(t=0) = \bar{x}_m / \sqrt{N_m}$, where $\bar{y}_m$ and $\bar{x}_m$ are constants and all other amplitudes are zero. We find the following analytical expressions for the expectation values of the electron spin components:

$$\langle s_z(t) \rangle \approx -\frac{1}{2} [|\bar{x}_m|^2 - |\bar{y}_m|^2] \left[1 - \text{Re} \frac{1}{N_m} \sum_{\{m\}} \sum_{j=1}^{\{m\}} \frac{A_j}{2(\epsilon + A)} G_{\{m\}} \right], \quad (14)$$

$$\langle s_+(t) \rangle \approx \frac{\bar{x}_m^* \bar{y}_m}{N_m} \sum_{\{m\}} e^{2iB_{\{m\}}t/\hbar}. \quad (15)$$

Here, $\sum_{\{m\}}$ means summing over all possible arrangements of the initially down spins. The following notation is also used:

$$G_{\{m\}}(t) = \sum_{l \neq \{m\}} \frac{A_l/2}{B_{\{m\}} - B_{\{m+1\}/l}} [1 - e^{i(B_{\{m\}} - B_{\{m+1\}/l})t/2\hbar}].$$

Here, $\hbar$ is recovered to give the correct units. The function $G_{\{m\}}$ is present if there is at least one nucleus flipped down, and $G_{\{m\}}$ is zero if the nuclei are initially fully polarized. For $m \ll N$, the sum in $G_{\{m\}}$ is $\sum_{l \neq \{m\}}^N \approx \sum_l^N$, and it is converted

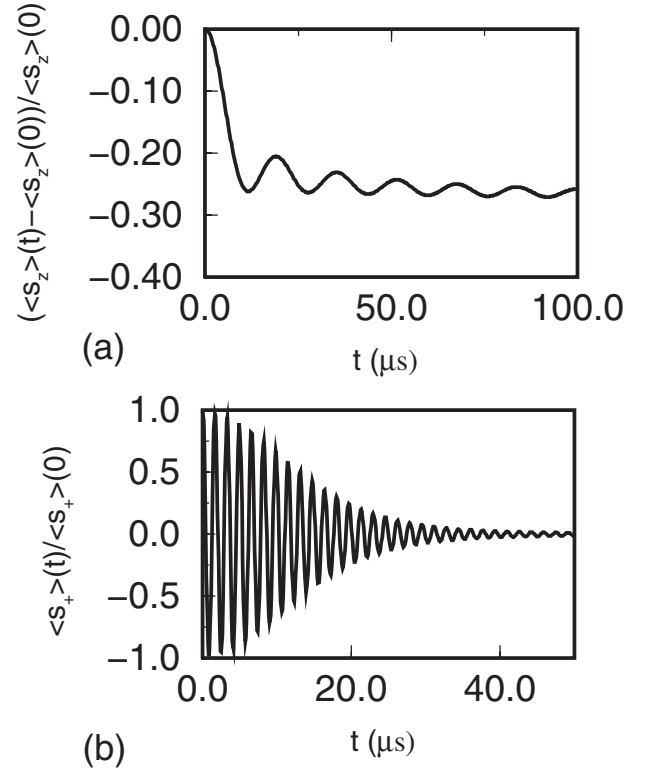


FIG. 1. (a) The time dependence of $\langle s_z \rangle$ from Eq. (13) for initial nuclear polarization of 75% is shown. (b) The time dependence of $\langle s_+ \rangle$ from Eq. (14) for initial nuclear polarization of 75% is shown. We take $A = 90 \mu\text{eV}$ (GaAs) and $\epsilon = A$.

into an integral $\sum_l^N \rightarrow \frac{1}{v_0} \int d^3r$. A similar treatment can be made for the summation in Eq. (15). Inserting the definition for $B_{\{m\}}$ and performing the summation, one obtains that $\frac{1}{N_m} \sum_{\{m\}} e^{2iB_{\{m\}}t/\hbar} \approx e^{2i(\epsilon+A)t/\hbar} J(t)^m$, where $J(t) = \frac{1}{N} \sum_{i=1}^N e^{iA_i t/\hbar}$. For these evaluations, the summations are again turned into integrations here.

In Fig. 1, we show the time dependences of the electron spin components for 75% initial nuclear polarization for $N = 10^4$. For illustration, we have chosen a spherical quantum dot of radius R , and use $|\psi(\mathbf{r}_i)|^2 = \frac{1}{\pi^{3/2}(R/2)^3} e^{-4r_i^2/R^2}$ for the electron wave function. We note that there are rapid oscillations in $\langle s_+(t) \rangle$. They arise from the exponential factor $e^{i(\epsilon+A)t/\hbar}$. The slower decays arise from the contributions of $A_j \sim A/N$ in both $\langle s_z(t) \rangle$ and $\langle s_+(t) \rangle$. The oscillations in the $\langle s_z(t) \rangle$ dynamics are more damped as compared to the ones in $\langle s_+(t) \rangle$ due to the prefactor $A_l/2[B_{\{m\}} - B_{\{m+1\}/l}]$. The time dependences of $\langle s_z(t) \rangle$ and $\langle s_+(t) \rangle$ are always nonexponential. Strictly speaking, the coupled system of electron and nuclear spins is secular and does not have dissipation. Revivals of the electron spin will occur at much longer times than those shown here. Nonexponential time decay was also found in other works.^{3,9} The evolution of the decay envelope also has been found to be nonexponential using analytical methods for systems with predominant initial nuclear polarization in which the nuclear interaction is switched on adiabatically¹⁰ and in studies in which numerical approaches were used.^{6,7}

A limiting case of these results is given by that for ini-

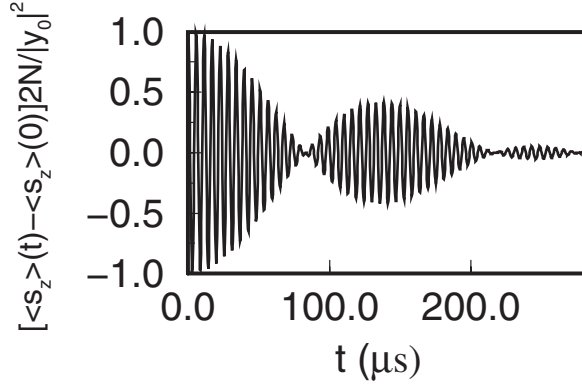


FIG. 2. The time dependence of $\langle s_z \rangle$ from Eq. (16) for the fully polarized case with hyperfine coupling $A=90 \mu\text{eV}$ and $\epsilon=A$.

tially fully polarized nuclei. For this case, we take the probability of the initial electron being up to be $\bar{x}_0$ and the probability of its being down to be $\bar{y}_0$. Then, we find that

$$\langle s_z(t) \rangle \approx \frac{1}{2} \left[|\bar{x}_0|^2 - |\bar{y}_0|^2 + |\bar{y}_0|^2 \right. \\ \left. \times \text{Re} \sum_{i=1}^N \frac{A_i^2}{4(\epsilon+A)^2} (1 - e^{-2i(\epsilon+A)t/\hbar} e^{iA_i t/2\hbar}) \right], \quad (16)$$

$$\langle s_+(t) \rangle \approx \frac{1}{2} \bar{x}_0^* \bar{y}_0^* e^{i(\epsilon+A)t/\hbar}. \quad (17)$$

In Fig. 2, the time evolution for this case is given. There are

rapid oscillations coming from contributions $\sim e^{-2i(\epsilon+A)t/\hbar}$ and slower envelop from contributions $\sim e^{iA_i t/2\hbar}$. We note that in this case, the time-dependent contributions are of order of $\sim 1/N$ smaller than $\langle s_z(t=0) \rangle$, a point that has been made earlier.^{3,9,10} In the fully polarized case, the long time envelope of $\langle s_z(t) \rangle$ has a time scale of $80 \mu\text{s}$, which is longer than the time scale for the 75% nuclear polarization above.

The long time behavior of $\langle s_z(t) \rangle$ and $\langle s_{\pm}(t) \rangle$ also can be obtained by evaluating Eqs. (14) and (16) at small distances from the center of the dot. In this way, we find that for the initially partially polarized system with m spins down, $\langle s_z(t) \rangle \sim (\hbar/At)^{3(m+1)/2}$ for long times and that for the initially fully polarized system $\langle s_z(t) \rangle \sim (\hbar/At)^{3/2}$, which is consistent with the results of Refs. 3 and 9. We find that the long time behavior of $\langle s_{z,\pm}(t) \rangle$ depends on the dimensionality of the system and on the shape of the quantum dot.^{3,9} The explicit form of the decay functions from Eqs. (14)–(16) allow one to obtain results for varying dot sizes and forms.

In summary, we have given a method to calculate the time dependence of electron spin components coupled by hyperfine interactions to a large number of nuclei with predominant initial polarization in quantum dots. This method is straightforward and accurate, it allows us to treat efficiently systems with general starting conditions, and it can be used to study relatively wide range of systems with varying dimensionality, quantum dot shape and initial nuclear spin polarization.

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¹²This is similar to the Schwinger-Dyson equation obtained in Ref. 9 where the hyperfine field is switched on adiabatically.