Decoherence of Nuclear Spin Quantum Memory in Quantum Dot

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Recently an ensemble of nuclear spins in a quantum dot have been proposed as a long-lived quantum memory. A quantum state of an electron spin in the dot can be faithfully transferred into nuclear spins through controlled hyperfine coupling. Here we study the decoherence of this memory due to nuclear spin dipolar coupling and inhomogeneous hyperfine interaction during the storage period. We calculated the maximum fidelity of writing, storing and reading operations. Our results show that nuclear spin dynamics can severely limit the performance of the proposed device for quantum information processing and storage based on nuclear spins.

INTRODUCTION

Electronic and nuclear spins in semiconductor nanostructures have been studied intensively because of their potential applications in quantum computation and spintronics. In a series of proposals of solid state quantum computers, electron or/and nuclear spins have been suggested as qubits [1,2], the information carriers. Hyperfine coupling, the main interaction between an electron and the surrounding nuclei in semiconductors, can be utilized as a tool to polarize the nuclei by optical pumping [3] or spin-polarized transport [4,5,6]. On the other hand, it could also be a major decoherence channel for electron spins [3,5,6].

In a recent study, long-lived quantum memory of the electron spin in both quantum dots [7] and quantum Hall semiconductor nanostructures [8,9], with the help of hyperfine interaction, is proposed using highly polarized nuclei in these structures. The idea is to transfer the electron spin state to nuclear spins coherently by tuning to the resonance condition of the nuclei-electron hyperfine flip-flop term; this process is referred to as information writing. Since nuclear spins have a long relaxation time $T_1$ (up to minutes), it is hoped that the information of electron spin can be stored in nuclei for a much longer period of time. The reverse process (reading) can be activated on demand later by tuning back to the resonance point so as to transfer the collective nuclear spin state back to the electron spin. Partially polarized nuclei which uniformly couple to the electron have been shown not to depolarize the performance of the quantum memory dramatically [10]. Internal dynamics of nuclear spins has so far been neglected.

However, nuclear spins are not static. Their evolution during the storage time, when their flip-flop with the electron is turned off by either changing the external magnetic field or simply removing the electron from the dot, will induce decoherence in the quantum memory. In this paper we study the influence of nuclear spin dynamics on the information transfer and storage. Two types of spin interactions of nuclei could be in effect, namely the dipole-dipole coupling and the inhomogeneous hyperfine coupling with the electron (effective field seen by nuclei).

During the writing and reading processes, the dipolar interaction can be ignored, since the operation time $\tau$ is determined by the hyperfine coupling constant (see below), so that $\tau H_d/\hbar$ is much less than 1, where $H_d$ is the dipolar Hamiltonian. However, it could be critical when the storage time approach the nuclear transverse relaxation time $T_2$, which is usually in the order of $10^{-3}$ to $10^{-4}$ second.

In this paper we assume that initially the nuclear spins are fully polarized in a pure state $|I, \cdots, I\rangle$, where $I$ represent the magnitude of nuclear spins, and $m_I$ means complete polarization. The system will undergo non-trivial evolution when a spin down electron is put into the dot. Once spin transfer is achieved at $\tau_w$, we turn off the resonance condition and let the nuclear spin system evolves for a long time $\tau_s$. Finally we read the information by putting back a spin up electron and turning on the resonance. Our objective then is to study how the maximal reading fidelity vary as a function of the storage time $\tau_s$.

COHERENT SPIN DYNAMICS OF THE EFFECTIVE TWO-LEVEL SYSTEM

In the writing and reading stage, the electron-nuclei spin system can be approximated with a simple Hamiltonian

$$H = \hbar \omega_z S_z - \hbar \omega_I \sum_{i=1}^{N} I_i z + H_h,$$

$$H_h = \sum_{i=1}^{N} A_i [I_i z S_z + \frac{1}{2} (I_i S_+ + I_i S_-)]. \quad (1)$$

The first two terms in $H$ are the Zeeman energies of the electron spin and nuclear spins. $H_h$ describes the hyperfine interaction in which a single electron interacts with an ensemble of $N$ nuclear spins where $N \approx 10^4 - 10^5$ for a small quantum dot; $A_i = \langle \psi(R_i) | \psi(R_i) \rangle^2$ is the local strength of hyperfine coupling and $\psi(R_i)$ is the electronic envelope wavefunction at the $i$th nucleus. In the Hamiltonian $H$ we neglect the nuclear dipole-dipole interaction as we have previously explained. The combined system
of nuclear and electron spins can be taken as an effective
two-level system, where the electron spin could be either
up or down. The time-dependent wavefunction reads

\[ |\Psi(t)\rangle = \alpha(t)|\Psi_0\rangle + \sum_{k=1}^{N} \beta_k(t)|\Psi_k\rangle, \quad (2) \]

where \(|\Psi_0\rangle = |\downarrow; I, \cdots, I\rangle\) and \(|\Psi_k\rangle = |\uparrow; I, \cdots, (I-1)k, \cdots, I\rangle\). Substituting Eq. 2 into Schrödinger equation,

\[ i\hbar \frac{\partial |\Psi(t)\rangle}{\partial t} = H|\Psi(t)\rangle, \]

we find

\[ i\dot{\alpha} = \frac{1}{2}(\delta + \gamma)\alpha + \sqrt{\frac{2}{\hbar}} \sum_j A_j \beta_j, \]

\[ i\dot{\beta}_k = \left( \frac{1}{2} \delta - \gamma + \omega_I \right) \beta_k - \frac{1}{2} \frac{A_k}{\hbar} \beta_k + \frac{\sqrt{2}}{\hbar} \frac{A_k}{2} \alpha, \quad (3) \]

where \(\delta = \omega_e + AI/\hbar\) and \(\gamma = NI\omega_I\). Under the resonance condition, the electron-nuclei flip-flops are greatly
enhanced since energies are conserved during these pro-
cesses. The resonance condition can be obtained by equating the total energies before and after the scatter-
ing. We arrive at

\[ \delta = \frac{A}{2Nh} - \omega_I. \quad (4) \]

In deriving Eq. 4 we have assumed that the hyperfine
coupling constant is \(A/N\). Since \(N \approx 10^3 \gg 1\) and \(\omega_I \approx 10^{-3}\omega_e < \omega_e\), Eq. 4 can be reduced to \(\delta = \omega_e + AI/\hbar \approx 0\), i.e., the resonance condition can be satisfied by adjusting the external magnetic field.

An exact solution of Eq. 8 was found using Laplace transformation [7]. However, the non-trivial complex in-
tegral (the inverse Laplace transformation) in the solution makes it less transparent. In the following we will apply a different approach. For later convenience we de-
define

\[ y_m = \sum_j \frac{A_j^m}{\hbar^m} \beta_j. \quad (5) \]

Using the resonance condition \(\delta = 0\) and Eq. 8 Eq. 3 can be transformed into

\[ i\dot{\alpha} = -\gamma \alpha + \sqrt{\frac{2}{\hbar}} y_1, \]

\[ i\dot{y}_m = -\gamma y_m - \frac{1}{2} y_{m+1} + \sqrt{\frac{2}{\hbar}} \sum_j \frac{A_j^{m+1}}{\hbar^{m+1}} \alpha. \quad (6) \]

Without further approximation Eq. 8 are equivalent to Eq. 2. However, if we write \(y_2\) as

\[ y_2 = \sum_j \frac{A_j^2}{\hbar^2} \beta_j \approx \frac{A}{Nh} y_1, \quad (7) \]

which is exact if the hyperfine coupling is uniform. The group of differential equations are now closed with only
\(\alpha\) and \(y_1\). This approximation is appropriate because the decoherence induced by the inhomogeneous hyperfine
coupling is significant only when the time scale is as long as \(N/A\) for fully polarized nuclei [8]. The spin transfer
process in which we are interested is much shorter. The argument will become clear in the following calculations.

We consider two initial conditions corresponding to the
writing and reading processes respectively. For the initial state with a spin down \(\Psi(0) = |\downarrow; I, I, \cdots, I\rangle; \alpha(0) = 1\) and \(\beta_1 = \cdots = \beta_N = 0\) and \(y_1(0) = 0\). The solution of Eq. 3 and Eq. 4 is

\[ \alpha(t) = e^{\gamma t} \cos \left( \frac{\Omega}{2} t \right), \quad (8) \]

where \(\Omega = \sqrt{2I \sum_j A_j^2}\) is the Rabi flopping frequency of the effective two-level system. Substituting Eq. 8 back
into Eq. 8 one find the solution of \(\beta_k\) is

\[ \beta_k(t) = i \frac{A_k}{\sqrt{\sum_j A_j^2}} e^{\gamma t} \sin \left( \frac{\Omega}{2} t \right). \quad (9) \]

We see that the system oscillates coherently between state \(|\Psi_0\rangle\) and state \(\sum_k A_k|\Psi_k\rangle/\sqrt{\sum_j A_j^2}\). If we keep \(y_2\) in Eq. 8 the solution will be different from Eq. 8. However the corrections of the amplitude and the frequency for \(\alpha\) are \(\sim 1/\sqrt{N}\) and \(A/N\) respectively. In other words, the approximation Eq. 8 we made should be OK as long as the time scale is much smaller than \(N/A\). If we let \(\tau_w = \pi/\Omega, |\alpha(t_w)|^2 = 0\) so that complete spin trans-
fering is achieved. The condition \(\tau_w < N/A\) is consistent with our approximation. For the initial state with a spin up electron, \(\Psi'(0) = \sum_k \beta_k'(0)|\Psi_k\rangle; \alpha'(0) = 0\) and \(y_1(0) = \sum_j A_j \beta_j'(0)/\hbar\). We find the solution in this case is

\[ \alpha'(t) = \frac{-i\hbar}{\sqrt{\sum_j A_j^2}} \frac{y_1(0)}{e^{\gamma t}} \sin \left( \frac{\Omega}{2} t \right). \quad (10) \]

Suppose nuclear spin dynamics is frozen during the storage
time, \(y_1(0) = \sum_j A_j \beta_j'(0)/\hbar = \sum_j A_j \beta_j(t_w)/\hbar\) and \(|\alpha'(\pi/\Omega)|^2 = 1\). However, if we allow the nuclear spins to evolve during the storage time, \(\beta_k(t)\) will be differ-
cent from \(\beta_k(t_w)\), which will make \(|\alpha'(\pi/\Omega)|^2\) less than 1. In the next section, we shall investigate how the deco-
herence induced by the nuclear spin dynamics affect the coherent electron spin transfer in the read out process.

**NUCLEAR SPIN DYNAMICS**

During the period of storage we can either turn off the
resonance by varying the external magnetic field or simply remove the electron from the dot. In the first case the electron-nuclei flip-flops are largely suppressed; nuclei
only feel the effective magnetic field generated by the term $A_j I_z S_z$, which acts like an inhomogeneous field for nuclei at different lattice sites. The effective Hamiltonian of nuclear spins is

$$H_n = \sum_i \left( \frac{A_i}{2} - \hbar \omega_I \right) I_z + H_d,$$

$$H_d = \sum_{i \neq j} 2b_{ij} I_z I_j - \sum_{i \neq j} b_{ij} I_i I_j .$$  \hspace{1cm} (11)

Here $H_d$ describes the secular terms of the dipole-dipole interaction; $b_{ij} = \hbar^2 \gamma_i^2 (1 - 3\cos^2 \theta_{ij}) R_{ij}^{-3} / 4$, where $R_{ij}$ is the distance between the nuclei at site $i$ and site $j$ and $\theta_{ij}$ is the angle between $R_{ij}$ and the external magnetic field. The second term in $H_d$ is the flip-flop term which introduces the nuclear spin diffusion \cite{14}. If the nucleus at the center of the dot is flipped down at $t = \tau_w$, then the down state of the nuclei will propagate to the nuclei at larger radius through their mutual flip-flops. After a sufficiently long period of time, the nucleus with flipped spin state may be at the very boundary of the dot. If one turns on the electron-nuclei resonance at this time, the injected spin-up electron has minimal chance to interact with the nucleus with flipped spin, because the probability of the electron at the boundary is negligible. This leads effectively to decoherence of the nuclear spin memory.

Since the truncated dipolar Hamiltonian $H_d$ conserve the total spin of all the nuclei in the quantum dot, the general time-dependent spin wavefunction can be expressed in terms of a linear combination of $|\psi_k\rangle$

$$|\psi(t)| = \sum_k \beta_k(t)|\psi_k\rangle,$$  \hspace{1cm} (12)

where $|\psi_k\rangle = |I, \cdots, (I-1)k, \cdots, I\rangle$. The Schrödinger equation is

$$i \dot{\beta}_k = \eta \beta_k - \frac{A_k}{2\hbar} \beta_k - 2I \sum_{m(k)} \frac{b_{km}}{\hbar} \beta_m,$$  \hspace{1cm} (13)

where $\sum_{m(k)}$ means the summation over all $m$ except for $m = k$,

$$\eta = \frac{AI}{2} - (NI - 1)\omega_I + 4I(I-1) \sum_{m(k)} \frac{b_{mk}}{\hbar},$$

$$+ 2I^2 \sum_{m(k)} \sum_{n(m,k)} \frac{b_{mn}}{\hbar},$$  \hspace{1cm} (14)

which is a constant. The reason that we do not drop the second term on the right hand side of Eq. (13) although it is much less than $\eta$, is that we are interested in the large time behavior ($\gg \pi / \Omega$) where non-uniform hyperfine coupling may have significant effect.

Even for such a simple situation, no analytical solution of Eq. (13) can be found. Therefore we have to rely on some numerical solutions. For this purpose we perform the following transformation for $\beta_k(t)$

$$\beta_k(t) = e^{-i(\eta - \frac{A_k}{2\hbar})t} \bar{\beta}_k(t).$$  \hspace{1cm} (15)

Eq. (13) then becomes correspondingly

$$i \dot{\bar{\beta}}_k = -2I \sum_{m(k)} \frac{b_{mk}}{\hbar} e^{i(A_m - A_k)t} \beta_m .$$  \hspace{1cm} (16)

This set of transformed equations shows that the factor $\eta$ only contributes an overall phase factor for every $\beta_k$, so it will not affect our calculation of the probabilities. We set the initial condition as $\beta_k(0) = i A_k / \sum_j A_j^2$, i.e., the system starts right after the spin of the electron is...
transferred to the nuclei (see, Eq. [11]). We assume in our calculations that

$$A(r) = e^{-\frac{(x^2 + y^2)}{2a^2}} e^{-\frac{z^2}{2a^2}},$$  \hspace{1cm} (17)$$

where $a = 5.65$ Å is the lattice constant for GaAs. The coordinates $x, y$ and $z$ are in units of $a$. $L_r$ and $L_z$ are two constants determined by the geometry of the quantum dot. $I = 3/2$ for all the nuclei in GaAs; $\gamma_{As} = 4.58 \times 10^3 \, \text{s}^{-1} \, \text{cm}^{-1}$. The time scale for dipolar interaction is $a^3/\gamma^2 h \approx 10$ milliseconds. We use both leapfrog and iterative Crank-Nicolson method to solve Eq. (16).

We find that the leapfrog scheme is better in maintaining the unitarity of the coherent evolution for the nuclear spins, the total probability is preserved in the evolution. We assume a closed boundary so that spin can not diffuse out of the dot. For simplicity we also assume that a single species of nuclei resides on a simple cubic lattice.

Let’s now consider the case where the electron is removed from the dot so that the evolution of the nuclear spins is only governed by the flip-flop term (nuclear spin diffusion). In Fig. 1 and Fig. 2 we show the probability $|\beta|^2$ as a function of spatial coordinates $x$ and $z$. The slightly different behaviors observed in Fig. 1 and Fig. 2 are due to the sharper confinement along the $z$ direction; we have also integrate Eq. (16) assuming the external field is in the $z$ direction. Similar results as in Fig. 1 and 2 are found. After time $\tau_s$, the coherent spin dynamics of electron can be activated by sending an electron with up spin into the dot. To calculate the fidelity of the reading process, we use the definition given in Ref. 11:

$$F = \text{Tr} \{ \rho(t_r) (\hat{S}_z - \frac{1}{2})^2 \},$$  \hspace{1cm} (18)$$

where $t_r = \pi/\Omega$. The density operator is defined as

$$\rho(t) = |\Psi(t)\rangle \langle \Psi(t)|,$$

where the time-dependent wavefunction takes the form of Eq. (10). It is then found that

$$F = \frac{\hbar^2}{\sum_j A_j^2} \left| \sum_k \beta_k(t_s) A_k \right|^2.$$  \hspace{1cm} (19)$$

The calculated fidelity is shown in Fig. 3 by the solid and dashed lines for different external magnetic field orientations. We see that the fidelity drops dramatically when $\tau_s$ reaches a few milliseconds. The difference of these two curves can be understood as follows. Nuclear spin diffusion is stronger along the external field direction. If this direction is the same as $z$ direction, which has larger confinement (larger gradient) of the electronic wavefunction, the diffusion process is even faster. This explains that fidelity represented by the solid line ($z$) is less that that of the dashed line ($x$) at equal time $\tau_s$.

It appears therefore desirable to suppress the decoherence induced by spin diffusion with an inhomogeneous hyperfine coupling [8, 13]. This can be realized by leaving the electron in the dot during time $\tau_s$. This suppression is due to energy non-conservation in the flip-flop processes. However our result (the dot-dashed line in Fig. 3) shows that the coherence can only be maintained within a few microseconds, even worse than when the electron is removed. We can explain this result by identifying another decoherence channel, the inhomogeneous hyperfine coupling itself. The nucleus at different lattice sites have different precession frequencies due to their interaction with the electron, so the phases of the wavefunction $\beta_k(t)$ will not be uniform throughout the dot (see Eq. (13)). This induces destructive interference of the function $\sum_k A_k \beta_k(t)$. The time scale of this decoherence is determined by the hyperfine coupling constant which is three order of magnitudes larger than the dipolar constant.

### CONCLUSION

In conclusion, we have studied nuclear-spin dynamics induced decoherence of the long-lived quantum memory proposed in Ref. [11]. We find that storage time is limited to a few milliseconds (without hyperfine coupling) and a few microseconds (with hyperfine coupling $A_i I_z S_z$). The performance of nuclear spin memory is limited by the two decoherence channels: nuclear spin dipolar coupling and inhomogeneous hyperfine coupling. We only consider the fully polarized nuclei. Partially polarized nuclei give rise to further decoherence. To make the device work as a truly long-lived quantum memory, one has to improve the device by combining it with other techniques such as refocusing.
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