

# Analytical solution of electron spin decoherence through hyperfine interaction in a quantum dot

Changxue Deng and Xuedong Hu

Department of Physics, University at Buffalo, SUNY, Buffalo, New York 14260-1500, USA

(Received 18 April 2006; published 7 June 2006)

We analytically solve the non-Markovian single-electron spin dynamics due to hyperfine interaction with surrounding nuclei in a quantum dot. We use the equation-of-motion method assisted with a large field expansion, and find that virtual nuclear spin flip-flops mediated by the electron contribute significantly to a complete decoherence of the transverse electron spin correlation function. Our results show that a 90% nuclear polarization can enhance the electron spin time  $T_2$  by almost two orders of magnitude. In the long time limit, the electron spin correlation function has a nonexponential  $1/t^2$  decay in the presence of both polarized and unpolarized nuclei.

DOI: [10.1103/PhysRevB.73.241303](https://doi.org/10.1103/PhysRevB.73.241303)

PACS number(s): 73.21.La, 03.67.Lx, 72.25.Rb, 85.35.Be

Spins in semiconductor nanostructures are promising qubit candidates for a solid state quantum computer because of their long decoherence times and potential scalability.<sup>1</sup> To demonstrate the feasibility of a spin qubit, electron spin decoherence in semiconductor quantum dots (QDs) has been widely studied both theoretically and experimentally.<sup>1-5</sup> The level discretization in a QD ensures that spin-orbit-interaction-induced spin relaxation is quite slow in QDs,<sup>6</sup> which leaves the environmental nuclear spins, particularly abundant in III-V semiconductors such as GaAs ( $10^4-10^6$  depending on the actual size of the QD), as the main source of decoherence for the electron spins.

It has been shown that static thermal polarization of nuclear spins leads to inhomogeneous broadening of electron spins (which can be corrected using the spin echo technique<sup>7</sup>) at a time scale of 10 ns,<sup>4,5,8</sup> and nuclear magnetic dipolar coupling leads to electron spin spectral diffusion and dephasing at a time scale of 10  $\mu$ s.<sup>9,10</sup> For the intervening period of time, the hyperfine interaction between the electron and nuclear spins can also lead to electron spin decoherence, which is in general non-Markovian because nuclear dynamics is slower than the hyperfine dynamics.

The study of the non-Markovian electron spin dynamics in the presence of hyperfine interaction is a complicated problem due to its quantum many-body nature (one electron spin and  $N$  nuclear spins), and has drawn widespread attention recently.<sup>11-14</sup> Analytically, an exact solution has been found in the case of a fully polarized nuclear reservoir,<sup>11</sup> while for the rest of the parameter regimes (in terms of nuclear polarization and external field), perturbative theory<sup>12</sup> or effective Hamiltonians<sup>13,14</sup> have been used to study the problem. Numerically, only small systems with typically fewer than 20 spins have been explored because of the extremely large Hilbert space.<sup>13,15,16</sup>

In this Rapid Communication we focus on the problem of the spin decoherence of a single electron due to hyperfine interaction with the surrounding nuclear spins. Although at a finite magnetic field the direct electron-nuclear-spin flip-flop is highly unlikely due to the Zeeman energy mismatch, higher-order processes where electron spins do not flip are possible. For example, conduction-electron-mediated nuclear spin interaction [Ruderman-Kittel-Kasuya-Yosida (RKKY)] has been studied for a long time in both metals and semiconductors.<sup>7,17</sup> Here our focus is the backaction of the

electron-mediated RKKY interaction between nuclear spins on the *single* mediating electron spin. We start from the exact electron-nuclear-spin Zeeman and hyperfine Hamiltonian and use the equation-of-motion approach in the Heisenberg picture. Helped by a systematic large field expansion, we solve the full quantum mechanical problem analytically and reveal the crucial importance of the electron-mediated nuclear spin flip-flop processes in the decoherence of an electron spin.

Calculating Green's functions with the equations of motion is an old technique in solid state physics.<sup>18</sup> In our current study we use this venerable technique to attack the problem of spin decoherence, which is generally studied using quantum master equations for the density operator.<sup>19</sup> This traditional approach originating from quantum optics is more adapted to dealing with weak interactions between a system and its reservoir. We demonstrate in this study that a properly defined correlation function can be used to fully characterize the decoherence properties of a two-level system, and the equation-of-motion approach can be a powerful tool in studying non-Markovian dynamics.

We model the coupled electron-nuclear-spin system by the Hamiltonian<sup>7</sup>

$$H = \omega_0 S^z + \sum_k A_k I_k^z S^z + \frac{1}{2} \sum_k A_k (I_k^+ S^- + I_k^- S^+), \quad (1)$$

where  $S$  and  $I$  represent the electron and nuclear spin operators, respectively,  $\omega_0$  is the external magnetic field,  $A_k$  is the hyperfine coupling constant with the  $k$ th nucleus, and  $\hbar=1$ . In this Rapid Communication we assume  $I=\frac{1}{2}$  for simplicity, though all calculations can be generalized for arbitrary  $I$ . For a two-dimensional QD with a Gaussian electron wave function,  $A_k$  has the simple form  $A_k=A_0 e^{-k/N}$  with  $k \in (0, \infty)$ .<sup>12</sup> For convenience we assume  $A_0=1$  so that time is measured in the unit of  $1/A_0$ .

To describe the decoherence between electron spin  $|\uparrow\rangle$  and  $|\downarrow\rangle$  states, we introduce a retarded transverse spin correlation function

$$G_{\perp}(t) = -i\theta(t)\langle\Psi_0|S^-(t)S^+(0)|\Psi_0\rangle. \quad (2)$$

Here  $\theta(t)$  is the usual step function, and  $|\Psi_0\rangle$  is the initial wave function of the system where the electron and nuclear

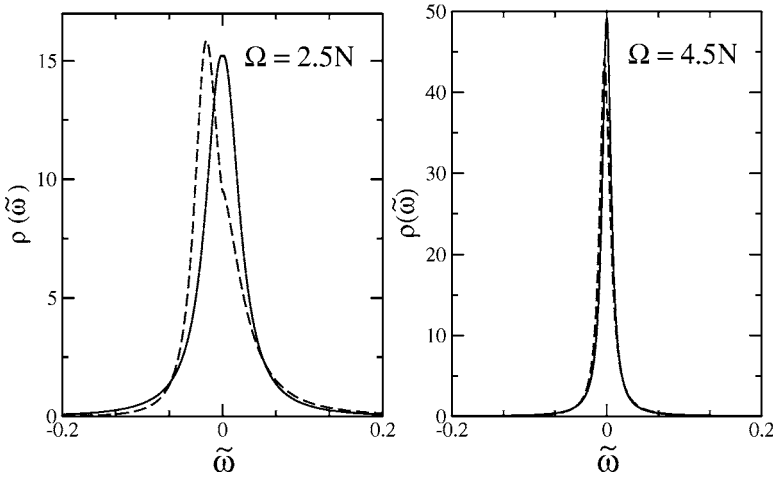


FIG. 1. Spectral function  $\rho(\tilde{\omega})$  (where  $\tilde{\omega} = \omega - \Omega - N/8\Omega$ ) with  $P=0$  for  $\Omega=2.5N$  (left panel) and  $4.5N$  (right panel). In both panels the solid lines represent the spectral functions calculated by considering only self-energy  $\Sigma_1(\tilde{\omega})$ . The dashed lines show the total contributions from  $\Sigma_1(\tilde{\omega})$  and  $\Sigma_2(\tilde{\omega})$ . The original delta functions  $\delta(\tilde{\omega})$  in the spectral densities are broadened due to the effects of the flip-flopping of nuclear spins.

spins are assumed to be in a product state, with the electron having spin down initially, i.e.,  $|\Psi_0\rangle = |\downarrow; I_{k_1}^z, I_{k_2}^z, \dots, I_{k_N}^z\rangle$ .<sup>20</sup> This spin correlation function represents the phase fluctuations between electron spin up and down states in the presence of the nuclear spin reservoir, which can be most clearly seen in the Schrödinger picture,

$$\begin{aligned} G_{\perp}(t) &= -i\theta(t)\langle \downarrow; I_{k_1}^z, \dots, I_{k_N}^z | e^{iHt/\hbar} S^- e^{-iHt/\hbar} S^+ | 0 \rangle \\ &\quad \times |\downarrow; I_{k_1}^z, \dots, I_{k_N}^z\rangle \\ &= -i\theta(t)\{ \langle \downarrow; I_{k_1}^z, \dots, I_{k_N}^z | e^{iHt/\hbar} \rangle S^- \\ &\quad \times \{ e^{-iHt/\hbar} | \uparrow; I_{k_1}^z, \dots, I_{k_N}^z \rangle \}. \end{aligned} \quad (3)$$

The term in the first curly brackets represents the evolution of the electron spin-down state in the presence of the hyperfine interaction, while the term in the second curly brackets represents the evolution of the electron spin-up state in the same environment. If no electron spin flip occurs, any decay in the calculated average will be due solely to dephasing between the electron spin-up and -down states. Obviously, electron spin flip will also cause decay of the correlation function. Therefore,  $G_{\perp}(t)$  contains the complete decoherence information for the electron spin in consideration.

An iterative equation of motion (EOM) for the spin correlation function  $G_{\perp}(t)$  can be obtained by differentiating  $G_{\perp}(t)$  with respect to time and then performing the Fourier transform. In general, for two arbitrary operators  $A$  and  $B$ ,

$$\omega \langle \langle A; B \rangle \rangle_{\omega} = \langle \Psi_0 | A(0)B(0) | \Psi_0 \rangle + \langle \langle [A, H]; B \rangle \rangle_{\omega}, \quad (4)$$

where  $\langle \langle A; B \rangle \rangle_{\omega}$  is the Fourier transform of  $\langle \Psi_0 | A(t)B(0) | \Psi_0 \rangle$ . For  $G_{\perp}(t)$ , we use  $G_{\perp}(\omega)$  to represent its Fourier transform, so that

$$\omega G_{\perp}(\omega) = 1 + \langle \langle [S^-, H]; S^+ \rangle \rangle_{\omega}. \quad (5)$$

The second terms on the right-hand side of Eqs. (4) and (5) involves the calculation of higher-order correlation functions. A cutoff or decoupling scheme has to be applied to eventually close the set of EOMs.

After  $G_{\perp}(\omega)$  is obtained, real time dynamics of  $G_{\perp}(t)$  can be easily calculated by an inverse Fourier transform using the spectral function defined as

$$\rho(\omega) = -\text{Im} G_{\perp}(\omega)/\pi. \quad (6)$$

In general there are two types of contribution to the spectral function after performing analytical continuation ( $\omega \rightarrow \omega + i0^+$ ):<sup>18</sup> a  $\delta$  function  $Z_p \delta(\omega - \omega_p)$ , and a nonvanishing imaginary part of the self-energy resulting from branch cuts, which results from the integration of continuous poles. The  $\delta$  function leads to a coherent oscillation with a single frequency  $\omega_p$ , while the continuous part leads to dephasing in the time evolution of the spin correlation function  $G_{\perp}(t)$ .

We consider the general case of partially polarized and unpolarized nuclear spin reservoir where both the numbers of spin-up and -down nuclei are of order  $N$ . The difference in the numbers of the two spin species is characterized by an effective polarization  $P = (N_{\uparrow} - N_{\downarrow})/N$ , where  $N_{\uparrow}$  ( $N_{\downarrow}$ ) are the number of nuclear spins in the up (down) states. Now the effective magnetic field takes the form  $\Omega = \omega_0 + \sum_k A_k \langle I_k^z \rangle$ , where  $\langle I_k^z \rangle$  represents time averaging of  $I_k^z(t)$ . We consider the physically relevant case of large effective fields ( $\Omega \sim N$ , requiring that we have either a reasonably large external field, or a nuclear reservoir with finite polarization), and focus on the spectral broadening near  $\omega = \Omega$ , which leads to dephasing of transverse electron spin magnetization.

Previous studies<sup>11,12</sup> indicate that when only the direct electron-nuclei spin flip-flop is considered, the decay amplitude of the electron spin correlation function is of the order  $O(1/N)$ , and the correlation function has almost undamped oscillations. Clearly, such direct processes are energetically unfavorable in high effective magnetic fields. However, if the higher-order virtual processes (electron-mediated nuclear spin flip-flop) are included, we expect that nuclear field fluctuation will give rise to complete decoherence in the electron spin. In other words, the  $\delta$  function (indicating no damping) in the spectral function would be broadened (decoherence) after the virtual processes are included. The spectral weight in the low-energy region where  $\omega \sim O(1)$  has been found to be negligible.<sup>21</sup>

In the following calculation we treat the nuclear field  $\sum_k A_k I_k^z$  within the adiabatic approximation, which is physical since  $S^-(t)$  has an oscillation frequency  $\Omega \sim N$  while the nuclear field varies in a much longer time scale, so that in the Fourier transform of  $\langle \Psi_0 | \sum_k A_k I_k^z(t) S^-(t) S^+(0) | \Psi_0 \rangle$  we can

simply replace  $\Sigma_k A_k I_k^z(t)$  by  $\Sigma_k A_k \langle I_k^z \rangle$ .  $G_{\perp}(\omega)$  is related to the higher-order correlation function  $\langle\langle I_k^{\dagger} I_{k'}^{\dagger} S^-; S^{\dagger} \rangle\rangle_{\omega}$  through the following equation:

$$\left(\omega - \Omega - \frac{N}{8\Omega}\right) G_{\perp}(\omega) = 1 + \frac{1}{2\Omega} \sum_{k \neq k'} A_k A_{k'} \langle\langle I_k^{\dagger} I_{k'}^{\dagger} S^-; S^{\dagger} \rangle\rangle_{\omega}. \quad (7)$$

Here  $\langle\langle I_k^{\dagger} I_{k'}^{\dagger} S^-; S^{\dagger} \rangle\rangle_{\omega}$  represents nuclear spins  $k$  and  $k'$  flip-flopping with each other while the electron spin returns to its initial state ( $\downarrow$ ).

Calculating this higher-order correlation function requires a cutoff to terminate the iteration. The structure of the iterative equations reveals that a natural cutoff does exist. The correlation function with one flip-flopped nuclear spin pair has two contributions to the self-energy, one of them proportional to  $N^2/(4\Omega)^2$ , the other to  $N^3/(4\Omega)^3$ . For two pairs of flip-flopped nuclear spins the two contributions are proportional to  $N^4/(4\Omega)^4$  and  $N^5/(4\Omega)^5$ , respectively. This geometrical series converges quite fast when  $\Omega > N$  (*large field expansion*). Physically, the expansion parameter  $N/\Omega$  appears because the intermediate high-energy state where the electron spin is flipped requires energy  $\Omega$ , and  $N$  comes from

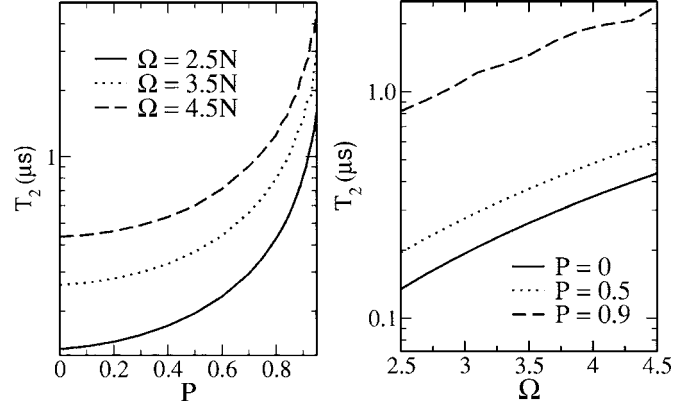


FIG. 2.  $T_2$  determined from the halfwidth of the peak of spectral functions for different nuclear polarizations ( $P$ ) and effective fields ( $\Omega$ ). In all cases the spectral functions are calculated using both  $\Sigma_1(\tilde{\omega})$  and  $\Sigma_2(\tilde{\omega})$ . We have chosen  $N=10^5$  and  $A=\Sigma A_k=92 \mu\text{eV}$ .

the summation over all nuclear spins. In the limit of  $\Omega \gg N$ , only the first-order term of the self-energy contributes significantly. Neglecting  $N^4/\Omega^4$  and higher-order terms, the final expression of the spin correlation function is

$$G_{\perp}(\omega) = \frac{1}{\tilde{\omega} - [(P^2 - 1)N^2/16\Omega^2]\Sigma_1(\tilde{\omega}) - [(1 - P^2)N^3/32\Omega^3]\Sigma_2(\tilde{\omega})}, \quad (8)$$

with the lowest-order self-energy taking the form

$$\Sigma_1(\tilde{\omega}) = \frac{2}{3} \left( \tilde{\omega}(4\tilde{\omega}^2 - 3) \ln \left| 1 - \frac{1}{4\tilde{\omega}^2} \right| + \tilde{\omega} + \ln \left| \frac{2\tilde{\omega} - 1}{2\tilde{\omega} + 1} \right| \right) + i \frac{2\pi}{3} (4|\tilde{\omega}|^3 - 3|\tilde{\omega}| + 1), \quad (9)$$

for  $|\tilde{\omega}| < 1/2$ . Here  $\tilde{\omega} = \omega - \Omega - N/8\Omega$ . The exact form of  $\Sigma_2(\tilde{\omega})$  is also found.<sup>21</sup> Both self-energy terms have branch cuts or nonvanishing imaginary parts when  $|\tilde{\omega}| < 1/2$ , leading to dephasing when calculating  $G_{\perp}(t)$ . Another significant feature of  $G_{\perp}(\omega)$  is that it does not have a  $\delta$ -function component any longer, indicating that the decoherence of  $G_{\perp}(t)$  will be complete. In addition, Eq. (8) indicates that the amplitude of  $G_{\perp}(\omega)$  is  $\sim O(1)$ , in contrast to the fully polarized case, where  $G_{\perp}(\omega) \sim O(1/N)$ .

Figure 1 shows the calculated electron spin spectral functions for different effective fields. We compare the results of including only  $\Sigma_1(\tilde{\omega})$  (solid lines) and those with both  $\Sigma_1(\tilde{\omega})$  and  $\Sigma_2(\tilde{\omega})$  (dashed lines) for  $\Omega=2.5N$  and  $4.5N$ . The two panels clearly show the validity of the large field expansion for  $\Omega \geq 2.5N$ . For smaller  $\Omega$  more higher-order terms need to be included to attain convergence. Indeed, even if  $\Omega < N$  there is no divergence in our theory, since there could be at most  $N_{\downarrow}$  ( $N_{\downarrow} < N_{\uparrow}$ ) flip-flopped nuclear pairs in the system, so

that there is an upper limit to the number of EOMs and terms in self-energy. The right panel of Fig. 1 (for  $\Omega=4.5N$ ) shows that the contribution of  $\Sigma_2(\tilde{\omega})$  is now completely negligible. Using hyperfine coupling constant of bulk GaAs,<sup>22</sup> we estimate that  $\Omega=2.5N$  corresponds to a magnetic field of 5 T. Figure 1 also explicitly shows that the original  $\delta$  function in the spectral function is now broadened after taking into account the electron-mediated flip-flop of nuclear spins. According to Eq. (8), in the limit  $\Omega \gg N$  or  $P=1$ , both self-energy terms go to zero, so that the  $\delta$ -function form of  $\delta(\tilde{\omega})$  of the spectral functions would have been recovered, and there would have been no decoherence effect.

The decoherence time  $T_2$  for the electron spin can be determined from the halfwidth ( $\Delta\tilde{\omega}$ ) of the spectral peak ( $T_2 = 1/\Delta\tilde{\omega}$ ). Figure 2 shows  $T_2$  as functions of the nuclear spin polarization  $P$  and the effective magnetic field  $\Omega$ . It is clear that  $T_2$  only increases slowly with the external magnetic field, but is much more sensitive to the nuclear polarization. If the nuclear polarization  $P$  is raised to 0.9 from 0,  $T_2$  increases by almost two orders of magnitude. Physically this is quite reasonable, as increasing polarization reduces the phase space for nuclear spin flip-flops, while increasing external field slowly reduces the cross section of these processes.

The real time dynamics of  $G_{\perp}(t)$  is obtained with the inverse Fourier transform  $G_{\perp}(t) = -i\theta(t) \int \rho(\omega) e^{-i\omega t} d\omega$  using the spectral function calculated with both  $\Sigma_1(\tilde{\omega})$  and  $\Sigma_2(\tilde{\omega})$ .

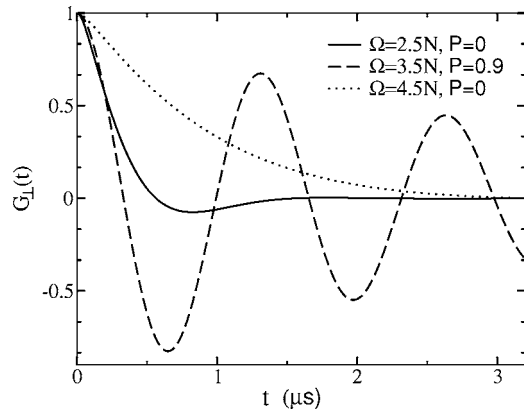


FIG. 3. Time evolution of the envelope of  $\text{Re}\{G_{\perp}(t)\}$  for various regimes of  $T_2$ .  $G_{\perp}(t)$  is obtained from the spectral function by inverse Fourier transform. It should be noted that the real evolution is modulated by the fast oscillation term  $e^{i\Omega t}$ . We have assumed  $N = 10^5$  and  $A = \sum A_k = 92 \mu\text{eV}$  in these calculations.

Figure 3 plots the time evolution of the envelope of  $\text{Re}\{G_{\perp}(t)\}$  for three different parameter regimes. The solid line represents the case of fast decay with  $\Omega = 2.5N$  and no polarization. The dotted line shows that increasing the magnetic field can increase the coherence time moderately. If the nuclei in the QD are polarized to 90%, the amplitude of the fast oscillation in electron spin (with frequency  $\Omega$ ) could be maintained for a much longer time as indicated by the dashed line. Notice that here the amplitude of  $G_{\perp}(t)$  does decrease as quickly as in the previous two cases initially, but

electron spin quantum coherence is only partially lost so that clear revival phenomenon is visible even after several microseconds in Fig. 3.

The long-time asymptotic behavior of  $G_{\perp}(t)$  can also be extracted from  $\rho(\omega)$ , which is nonzero only when  $|\tilde{\omega}| < 1/2$ . Calculating the inverse Fourier transform at  $t \gg 1$ , we find

$$G_{\perp}(t) \propto \frac{144\Omega^2}{\pi^2(1-P^2)N^2} \frac{1}{t^2}. \quad (10)$$

The  $1/t^2$  power-law decay here can be compared to the  $1/t$  power-law decay found in Refs. 11 and 12 for large magnetic fields, where electron-mediated nuclear spin flip-flops are not taken into account, and the exponential decay found in Ref. 14, where an effective Hamiltonian for the nuclear spin flip-flop is considered.

In summary, we have presented a detailed analytical study of transverse electron spin decoherence using large field expansion. We find that electron-mediated nuclear spin flip-flops contribute significantly to electron spin dephasing by generating fluctuations in the Overhauser field (the nuclear field) for the electron spin. We find that 80–90% nuclear polarization can enhance the electron spin  $T_2$  time by two orders of magnitude into the microsecond time scale in a 5 T external field. We also show that the long-time asymptotic behavior of the spin decoherence is  $1/t^2$ .

We acknowledge financial support by NSA, LPS, ARO, and NSF.

- <sup>1</sup>X. Hu and S. Das Sarma, *Phys. Status Solidi B* **238**, 360 (2003); X. Hu, in *Quantum Coherence*, edited by W. Potz, J. Fabian, and U. Hohenester, Lecture Notes in Physics Vol. 689 (Springer, Berlin, 2006), pp. 83–114; S. Das Sarma, R. de Sousa, X. Hu, and B. Koiller, *Solid State Commun.* **133**, 737 (2004).
- <sup>2</sup>D. Loss and D. P. DiVincenzo, *Phys. Rev. A* **57**, 120 (1998).
- <sup>3</sup>T. Fujisawa, D. G. Austing, Y. Tokura, Y. Hirayama, and S. Tarucha, *Nature (London)* **419**, 278 (2002).
- <sup>4</sup>F. H. L. Koppens, J. A. Folk, J. M. Elzerman, R. Hanson, L. H. Willems van Beveren, I. T. Vink, H. P. Tranitz, W. Wegscheider, L. P. Kouwenhoven, and L. M. K. Vandersypen, *Science* **309**, 1346 (2005).
- <sup>5</sup>J. R. Petta, A. C. Johnson, J. M. Taylor, E. A. Laird, A. Yacoby, M. D. Lukin, C. M. Marcus, M. P. Hanson, and A. C. Gossard, *Science* **309**, 2180 (2005).
- <sup>6</sup>V. N. Golovach, A. Khaetskii, and D. Loss, *Phys. Rev. Lett.* **93**, 016601 (2004).
- <sup>7</sup>C. P. Slichter, *Nuclear Magnetic Resonance* (Springer-Verlag, Berlin, 1996).
- <sup>8</sup>I. A. Merkulov, A. L. Efros, and M. Rosen, *Phys. Rev. B* **65**, 205309 (2002).
- <sup>9</sup>R. de Sousa and S. Das Sarma, *Phys. Rev. B* **68**, 115322 (2003).
- <sup>10</sup>W. M. Witzel, R. de Sousa, and S. Das Sarma, *Phys. Rev. B* **72**, 161306(R) (2005).
- <sup>11</sup>A. Khaetskii, D. Loss, and L. Glazman, *Phys. Rev. Lett.* **88**, 186802 (2002); *Phys. Rev. B* **67**, 195329 (2003).
- <sup>12</sup>W. A. Coish and D. Loss, *Phys. Rev. B* **70**, 195340 (2004).
- <sup>13</sup>N. Shenvi, R. de Sousa, and K. B. Whaley, *Phys. Rev. B* **71**, 224411 (2005).
- <sup>14</sup>W. Yao, R. B. Liu, and L. J. Sham, cond-mat/0508441 (unpublished); cond-mat/0604634 (unpublished).
- <sup>15</sup>J. Schliemann, A. V. Khaetskii, and D. Loss, *Phys. Rev. B* **66**, 245303 (2002).
- <sup>16</sup>S. I. Erlingsson and Y. V. Nazarov, *Phys. Rev. B* **70**, 205327 (2004).
- <sup>17</sup>N. Bloembergen and T. J. Rowland, *Phys. Rev.* **97**, 1679 (1955).
- <sup>18</sup>G. D. Mahan, *Many-Particle Physics* (Plenum Press, New York, 1990).
- <sup>19</sup>U. Weiss, *Quantum Dissipative Systems* (World Scientific, Singapore, 2001).
- <sup>20</sup>We assume that the nuclear spins are initially in a product state. For mixed initial nuclear states, the result of this paper can be directly used in the sum over all the product states in the ensemble.
- <sup>21</sup>C. Deng and X. Hu (unpublished).
- <sup>22</sup>D. Paget, G. Lampel, and B. Sapoval, *Phys. Rev. B* **15**, 5780 (1977).