Decoherence of coupled electron spins via nuclear spin dynamics in quantum dots

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In double quantum dots, the exchange interaction between two electron spins renormalizes the excitation energy of pair-flips in the nuclear spin bath, which in turn modifies the non-Markovian bath dynamics. As the energy renormalization varies with the Overhauser field mismatch between the quantum dots, the electron singlet-triplet decoherence resulting from the bath dynamics depends on sampling of nuclear spin states from an ensemble, leading to the transition from exponential decoherence in single-sample dynamics to power-law decay under ensemble averaging. In contrast, the decoherence of a single electron spin in one dot is essentially the same for different choices of the nuclear spin configuration.

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Decoherence draws a boundary between the microscopic quantum world and the macroscopic classical world. It is also a main obstacle in quantum technologies such as quantum computation. Thus, both for understanding crossover from the quantum to the classical world and for exploiting quantum coherence of large systems, it is desirable to comprehend how decoherence develops with scaling up the size of a system. The very initial step toward such a purpose is to examine the difference between a two-level system (the simplest quantum object, called a qubit in quantum computation) and two coupled ones. For a system in a Markovian bath (which has broad-band fluctuation), the decoherence is described by the Lindblad formalism. For a system in a non-Markovian bath, there are indications of nontrivial scaling behavior of decoherence, such as the non-additive decoherence in multiple baths. In this paper, we study the decoherence of a composite quantum object in a non-Markovian mesoscopic bath, based on a paradigmatic system in mesoscopic physics and quantum information science, namely, two electron spins in double quantum dots (QDs) where the nuclear spins serve as the bath.

The dynamics of a mesoscopic nuclear spin bath in a QD is conditioned on the state of the electron spin in contact with the bath. The conditional evolution of the nuclear spins establishes the electron-nuclear entanglement that causes the electron spin decoherence. When the electron spin is disturbed, the nuclear bath dynamics is altered. For example, the nuclear spin evolution can be shepherded by a structured sequence of electron spin flips so that the electron is disentangled from the bath and as a result the lost coherence is recovered. Besides external control, the disturbance may also be due to interaction with another quantum object in proximity, such as the exchange interaction between two electron spins in coupled QDs. Intuitively speaking, the disturbance due to the exchange interaction may be viewed as precessing of the electron spins about each other. For a more rigorous treatment, we should first diagonalize the electron spin Hamiltonian including the exchange interaction, and then study the bath dynamics and the decoherence in the electron eigenstate basis. To demonstrate the most essential physics, we consider only the decoherence between the singlet state $|S\rangle$ and the unpolarized triplet state $|T_0\rangle$ (by assuming that the polarized triplet states $|T_\pm\rangle$ are well split off by a large external magnetic field). The singlet-triplet (S-T) dephasing due to the Overhauser field distribution in an ensemble or a superposition state of nuclear spins has been studied in Refs. [15, 16], but the bath considered there has no dynamical fluctuation since the interaction between nuclear spins was neglected.

The role of the exchange interaction in the decoherence may be disclosed in a semiclassical spectral diffusion picture. Let us denote the local Overhauser fields for the nuclear spin configuration $|I\rangle$ by the electron Zeeman energies $\Omega_I^2$ and $\Omega_I^Z$ in dot 1 and dot 2, respectively. The pairwise nuclear spin flip-flops cause the dynamical fluctuation of the Overhauser fields and therefore a random phase of the electron spins. For a single electron spin in one QD, the Zeeman energy change due to the $k$th pair-flip is $2Z_k$. With exchange interaction, the S-T splitting is $E_{S,T} = \sqrt{J_{ex}^2 + \Delta_T^2}$, varying with the Overhauser field mismatch $\Delta_T = \Omega_I^2 - \Omega_I^Z$ [see Fig. 1(a)]. So, the S-T splitting change due to the $k$th pair-flip is

$$2Z_k \approx (2Z_k) \frac{\partial E_{S,T}}{\partial \Delta_T} \approx 2Z_k \frac{\Delta_T}{E_{S,T}}. \quad (1)$$

The exchange interaction modifies the energy fluctuation (or spectral diffusion) responsible for the decoherence.

For a full quantum mechanical description of the decoherence, we focus on the nuclear spin dynamics which is driven by pair-flips (as elementary excitations in the bath). The pair-flips are characterized by the flip transition strength and the energy cost. For uncorrelated electrons in the double dots ($J_{ex} = 0$), the hyperfine-energy cost of a nuclear pair-flip is $\pm Z_k$ for electron spin up and down states, respectively. In presence of exchange interaction, the energy cost for the pair flip is renormalized to be $\pm Z_k$ for the triplet and singlet states, respectively. Thus the bath dynamics itself is al-
FIG. 1: (Color online) (a) The singlet and triplet energies as functions of the Overhauser field mismatch $\Delta_I$, and the (renormalized) excitation energy of a nuclear spin pair-flip $Z_k$ ($Z_k$). (b) FID of the S-T coherence for various initial nuclear spin states, indicated by $\Delta_I$. (c) FID decoherence time $T_2^*$ as a function of $\Delta_I$.

tered when non-interacting quantum objects in the bath are replaced by interacting ones. In particular, through the dependence of the bath fluctuation on the Overhauser field mismatch $\Delta_I$, the dynamics of nuclear spins in a dot are affected by the state of the nuclear spins in the other dot. Therefore, the resultant S-T decoherence time varies with sampling of the nuclear spin configuration $|I\rangle$ from an ensemble $\rho_N = \sum_T P_T |T\rangle \langle T|$. This, as will be shown later, leads to a transition from exponential decoherence to power-law decay upon ensemble average. On the contrary, the decoherence of a single electron spin in a QD is essentially independent of the static Overhauser field $|12, 14\rangle$.

We consider a gate-defined double-dot structure similar to those used in Ref. [10, 11, 15, 16]. We assume a large on-site Coulomb energy and gate voltages supporting one electron in each dot. The virtual inter-dot tunneling mediates the exchange interaction. Under a large magnetic field (such that the electron Zeeman energy $\Omega_e \gg J_{ex}$), the unpolarized states $|S\rangle$ and $|T_0\rangle$ are well separated in energy from the polarized triplet states $|T_k\rangle$. The electrons and nuclei are coupled by the contact hyperfine interaction $\hat{S}_1 \cdot \hat{h}_1 + \hat{S}_2 \cdot \hat{h}_2$, where $\hat{h}_j = \sum_n a_{j,n} \hat{I}_{j,n}$, with $a_{j,n}$ being the hyperfine coefficient for the $n$th nuclear spin at the $j$th dot. The off-diagonal hyperfine interaction (terms with $\hat{S}_k^j, \hat{S}_k^j$) couples $|S\rangle$ and $|T_0\rangle$ to $|T_k\rangle$, which, however, has negligible effect under a strong external magnetic field ($\Omega_e \gg a_{j,n}$) [20]. The Overhauser field mismatch $\Delta = h_1^2 - h_2^2$ couples $|T_0\rangle$ and $|S\rangle$, which causes longitudinal $T_1$ relaxation. For a relatively large exchange splitting (e.g., $J_{ex} \gtrsim 10 \Delta_I$), the $T_1$ process is suppressed $[13, 10]$, but virtual S-T flips induce a self-energy correction

$$\hat{H}_{S,T} = \left( E_{S,T} + \frac{\Delta_I}{E_{S,T}} \mathbf{\delta}_T \right) \otimes |T_0\rangle \langle T_0| - |S\rangle \langle S|, \tag{2}$$

where the mean-field part ($\Delta_I = \langle I | \Delta | T \rangle$) is singled out from the quantum fluctuation ($\mathbf{\delta}_T = \Delta - \Delta_I$) of the Overhauser field, and is included non-perturbatively. This self-energy correction is responsible for the renormalized energy cost ($Z_k$) of a nuclear spin pair-flip driven by the intrinsic nuclear-nuclear spin interaction $H_N$. Now the Hamiltonian of the electron-nuclear spin system is $\hat{H} = \hat{H}_k \otimes |T_0\rangle \langle T_0| + \hat{H}_T \otimes |S\rangle \langle S|$, where $\hat{H}_k = \hat{H}_N \pm E_{S,T}/2 \pm \hat{H}_Z$ with $\hat{H}_Z = \mathbf{\delta}_T \Delta_I/(2E_{S,T})$. Such a block-diagonal Hamiltonian induces no $T_1$ relaxation but only pure S-T dephasing. We emphasize that the intrinsic nuclear spin interaction $\hat{H}_N$ is essential to the decoherence. Otherwise, without $\hat{H}_N$, the pure dephasing caused by the hyperfine interaction is totally eliminated from spin echo signals $[12, 13, 14, 21]$.

The S-T decoherence is caused by the electron-nuclear entanglement, established during the evolution of the nuclear spin state (on the electron states: Suppose the electrons are initially in a superposition state $|\alpha\rangle |S\rangle + |\beta\rangle |T_0\rangle$ and the nuclear state $|I\rangle$ is one randomly chosen from a thermal ensemble. The conditional nuclear spin evolution $|I\rangle \rightarrow |I^\pm (\tau)\rangle \equiv e^{-i\hat{H}_I \tau} |I\rangle$ establishes an entangled state $|\alpha\rangle |S\rangle \otimes |T^\pm\rangle + |\beta\rangle |T_0\rangle \otimes |T^+\rangle$. The S-T coherence is given by $L_I(\tau) = \langle I^\pm (\tau) | T^+ (\tau) \rangle$. To calculate the nuclear spin evolution $|I^\pm (\tau)\rangle$, we employ the pair-correlation approximation in which all possible pair-flips from the initial configuration $|I\rangle$ are taken as independent of each other. The pair-correlation approximation is justified by a mesoscopic nuclear spin bath with a sufficiently random configuration where within the dephasing timescale the occurred pair-flips are much fewer than the available pairs to be flipped and therefore have little probability to be in neighborhood of each other or to be correlated $[12, 13, 14, 18, 19, 22, 23]$. A pair-flip is characterized by a transition due to the off-diagonal nuclear spin interaction $D_k = \langle I | \hat{H}_N | I, k \rangle$, an energy cost due to the diagonal nuclear spin interaction $D_k = \langle I, k | \hat{H}_N | I, k \rangle - \langle I | \hat{H}_N | I \rangle$, and a hyperfine energy cost $\pm Z_k = \pm \left( \langle I, k | \hat{H}_Z | I, k \rangle - \langle I | \hat{H}_Z | I \rangle \right)$ for the triplet and the singlet state, respectively, where $|I, k\rangle$ denotes the nuclear spin state after the $k$th pair-flip. An independent pair-flip is mapped to be a pseudo-spin $\hat{s}_k$ precessing about a pseudo-field $\chi_k^\pm \equiv (2B_k, 0, D_k \pm Z_k)$, initially pointing to the down direction $[12, 13, 14]$. The nuclear Hamiltonian in the pseudo-spin representation is $\hat{H}_k = \pm E_{S,T}/2 + \sum_k \chi_k^\pm \cdot \hat{s}_k$.

Within the pair-correlation approximation, the S-T coherence in free-induction decay (FID) is

$$L_I(\tau) = e^{-iE_{S,T}/2} \prod_k \left| \left\langle 1 \left| e^{+i\chi_k^\pm \hat{s}_k \tau} e^{-i\chi_k^\pm \hat{s}_k \tau} | 1 \right\rangle \right| \right|, \tag{3}$$

where
The short-time behaviour (for $\tau \ll Z_k^{-1}$) is a quartic exponential decay, $L_{I}(\tau) \approx \exp(-iE_{S-T}\tau / \sum_k B_k^2 Z_k^2 \tau^4 / 2) \equiv e^{-iE_{S-T}\tau / (\tau / T_{I}^2)^4}$. The decoherence time $T_{I}^2 \propto (Z_k)^{-1/2} \propto (\Delta_{I}/E_{S-T})^{-1/2}$, varying with sampling of the nuclear spin configuration from the ensemble $\sum_{I} P_{I} |I\rangle \langle I|$. For comparison, the decoherence of a single electron spin in a QD [24], except for a trivial global phase factor related to the inhomogeneous broadening, is essentially independent of choice of the initial state $|I\rangle$, since the excitation energy of a nuclear pair-flip there $Z_k = \langle I, k | \hat{h}_{z}^2 / 2 | I, k \rangle - \langle I | \hat{h}_{x}^2 / 2 | I \rangle$ is independent of $|I\rangle$. Also, the S-T decoherence time is longer than the single spin decoherence time by a factor of $\sqrt{E_{S-T}/\Delta_{I}}$ because of the reduction of the excitation energy.

In numerical evaluation, we take a symmetric GaAs double-dot structure with height 6 nm, Fock-Darwin radius 70 nm for a parabolic confinement potential, and center-to-center separation 137 nm, under a perpendicular magnetic field $B = 1$ T at temperature $T = 1$ K. The calculated variance of the local Overhauser field mismatch is $\Gamma \approx 0.12 \mu$eV, corresponding to an inhomogeneous dephasing time $T_{2} = \sqrt{\Delta_{I}/\Gamma \approx 8}$ ns. The exchange energy $J_{ex} \approx -1 \mu$eV is determined with the Hund-Mulliken limit [13], consistent with experimental values [14].

Figure 1(b) shows the FID decoherence for several nuclear spin initial states $|I\rangle$ randomly chosen from the thermal ensemble. The suppression of the decoherence with decreasing the Overhauser field mismatch $\Delta_{I}$ is evident. The dependence of the decoherence time on the Overhauser field mismatch $\Delta_{I}$ is verified in Fig. 1(c). Note that for vanishing mismatch $\Delta_{I} \to 0$, the energy cost $Z_k$ vanishes in the leading order of $Z_k$ and the second-order correction $Z_k = Z_k^2 / J_{ex}$, resulting in a saturation of the decoherence time at a large value.

In ensemble dynamics, the signal is to be averaged by $L(\tau) = \sum_{I} P_{I} L_{I}(\tau)$. The inhomogeneous broadening of the Overhauser field results in an effective dephasing $\sum_{I} P_{I} e^{-iE_{S-T} \tau}$ with a nanosecond timescale [15, 16], much faster than the entanglement-induced decoherence $L_{I}(\tau)$ in single-sample dynamics. So below we study the ensemble-averaged coherence in spin-echo configurations where the inhomogeneous broadening effect is eliminated.

In the single-pulse Hahn echo ($\sigma_{x} \tau \sigma_{x} \tau$-echo), the S-T coherence at the echo time $2\tau$ for a nuclear spin state $|I\rangle$ is

$$L_{I}^{(1)}(2\tau) = \prod_{k} \left| \left\langle \hat{U}_{k}^{(1)}(\tau) \right| \hat{U}_{k}^{(1)}(\tau) \right| ,$$

where $\hat{U}_{k}^{(1)}(\tau) \equiv e^{-i\hat{H}_{k}\tau} e^{-i\hat{Z}_{k}^{2}\tau}$. The short-time behavior (for $\tau \ll Z_k^{-1}$) is $L_{I}^{(1)}(2\tau) \approx \exp(-2 \sum_k B_k^2 Z_k^2 \tau^4) \equiv e^{-2(2\tau / T_{I}^2)^4}$ with the single-sample decoherence time $T_{I}^2 = (\sum_k B_k^4 Z_k^2 / 8)^{-1/4} = \sqrt{2T_{I}^2}$. The ensemble dynamics is studied by averaging over a large number of samples from a Gaussian distribution of the Overhauser field. With the approximation $E_{S-T} \approx J_{ex}$, the ensemble-averaged result is analytically obtained for $\tau \ll Z_k^{-1}$,

$$L^{(1)}(2\tau) \approx \left[ 1 + (2\tau / T_{H})^4 \right]^{-1/2} ,$$

with a power-law decay profile, where the ensemble decoherence time $T_{H} = T_{I} \left| \Delta_{I} = \sqrt{Z} \right.$, i.e., the decoherence time for a nuclear spin configuration with the Overhauser field mismatch equal to $\sqrt{2}$ times the standard variance. Such a transition from an exponential decay in single-sample dynamics to a power-law decay in ensemble dynamics is shown in Fig. 2. In contrast, the echo signal of a single electron spin in a QD is unchanged by ensemble averaging, i.e., $L^{(1)} = T_{I}^{(1)}$.

We now study the S-T decoherence under concatenated control which is designed to preserve the coherence [13, 14, 20]. The coherence preserved after the $m$th order concatenated pulse sequence $L_{I}^{(m)}(\tau_{m})$ is obtained by substituting $\hat{U}_{k}^{(m),\pm}$ for $\hat{U}_{k}^{(1),\pm}$ in Eq. (1), where $\tau_{m} \equiv 2^{m}\tau$, and $\hat{U}_{k}^{(m),\pm}$ is recursively defined as $\hat{U}_{k}^{(1),\pm} U_{k}^{(m-1),\pm}$ for $m > 1$. The short-time profile (for $\tau \ll Z_k$) is $L_{I}^{(m)}(\tau_{m}) \approx \exp(-\tau_{m} / (2T_{H}^{(m)}))$, with the decoherence time $T_{H}^{(m)} \approx 2\tau_{m} (2^{m+1})^{1/2} |Z_k B_k^{m+2}(\Delta_{I}/E_{S-T})|^{-1/4}$. Again, the ensemble average leads to a power-law decay

$$L^{(m)}(\tau_{m}) \approx \left[ 1 + (\tau_{m}/T_{H}^{(m)})^{4m+2} \right]^{-1/4} ,$$

with $T_{H}^{(m)} = T_{H}^{(m)} \left| \Delta_{I} = \sqrt{Z} \right.$ in contrast, for a single electron spin, the ensemble-averaging has negligible effect.
decoherence in one dot (\(L^{(m)}_1(\tau_m)\)) and for the S-T decoherence in two coupled dots (\(L^{(m)}_2(\tau_m)\)), as indicated by the concatenation level. (b) Logarithmic plot of (a).

i.e.,\(L^{(m)}_j(\tau_m) = L^{(m)}_j(\tau_m) \approx e^{-(\tau_m/T_{2,m})^{2m+2}}\), where the decoherence time \(T_{2,m}\) and for the S-T decoherence time \(T_{2,m}\) by a factor \(\sim (J_{\text{ex}}/\Gamma)^{1/(m+1)}\). Fig. 3 compares the S-T decoherence to the single spin decoherence, showing the suppression of the coherence and the crossover to a power-law decay due to the coupling between the electron spins.

In conclusion, the exchange interaction between two electron spins in double QDs modifies the nuclear spin bath dynamics through renormalizing the pair-flip excitation energy. As the renormalized excitation energy varies with the Overhauser field mismatch between the two dots, the nuclear spin dynamics in one dot becomes dependent on the nuclear spin state in the other dot, regardless of nonexistence of inter-dot nuclear-spin interaction in the considered situation. Consequently, the singlet-triplet decoherence due to the electron-nuclear entanglement depends on choice of the nuclear spin configuration from an ensemble, leading to a power-law decay of ensemble-averaged coherence, in contrast with the exponential decoherence of a single electron spin which is insensitive to sampling of the nuclear spin ensemble. The dependence of the S-T decoherence on the Overhauser field mismatch may be observed by tuning the mismatch with an inhomogeneous external field. The exchange interaction also enhances the S-T decoherence time by suppressing the fluctuation in the nuclear spin bath.

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