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Phonon-Mediated Electron-Spin Phase Diffusion in a Quantum Dot

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An effective spin relaxation mechanism that leads to electron spin decoherence in a quantum dot is proposed. In contrast with the common calculations of spin-flip transitions between the Kramers doublets, we take into account a process of phonon-mediated fluctuation in the electron spin precession and subsequent spin phase diffusion. Specifically, we consider modulations in the longitudinal g factor and hyperfine interaction induced by the phonon-assisted transitions between the lowest electronic states. Prominent differences in the temperature and magnetic field dependence between the proposed mechanism and the spin-flip transitions are expected to facilitate its experimental verification. Numerical estimation demonstrates highly efficient spin relaxation in typical semiconductor quantum dots.

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Recently, there has been much interest in the study of electron spin decoherence mechanisms in quantum dots (QDs) since they are a natural candidate for the qubit operations of quantum computing. A typical approach to this problem is to calculate the spin transition probability caused by the electron spin interaction with a thermal bath [1–3]. A thermal reservoir composed of nuclear spins is not an effective dissipative system due to the long nuclear spin relaxation; however, the nuclei can assist in the phonon-induced electron spin relaxation [4–7].

Since spin-lattice relaxation (SLR) is assumed to be associated with spin-flip transitions between different electron spin states, the nondiagonal spin-flip matrix elements must be taken into account. Most studies of SLR in the literature have analyzed various mechanisms responsible for such transitions. Specifically, there are two approaches to this problem. The first is based on the spin-orbital interaction which is particular to III-V [1] or Si/Ge [2,3] QDs. The second approach incorporates the nuclear hyperfine interaction (HFI) [5–7] as a factor, leaving the time-reversal symmetry for the electron spin Hamiltonian [4].

Actually, there are no principal differences between the aforementioned approaches to the SLR in QDs and the early pioneering works performed in the 1960s for shallow donor relaxation (with the exception of the specific electronic energy structure and the influence of strain). Both the early works [8–10] and the later studies of QD SLR [1–3] deal with the *longitudinal* (or energy) relaxation accompanying the exchange between the Zeeman and phonon reservoirs. Note, however, that quantum computing is qualitatively limited by other relaxation processes which result in the destruction of the electron spin phase coherence that can occur without energy relaxation. Development of *transversal* (or phase) relaxation was not the goal of the early studies nor the recent SLR research in QDs.

In this Letter, we show that there are decoherence mechanisms specific to phase relaxation, which can be rather more effective than energy relaxation. To make this assertion clearer, let us consider an electron spin $\vec{s}$ under the influence of a magnetic field directed along the z axis with a randomly fluctuating strength. In this case, the projection of electron spin on the z axis s_z is conserved and no longitudinal relaxation occurs. Nevertheless, the phase of electron spin will change randomly with the Zeeman frequency fluctuation $\delta\Omega$ resulting in a decoherence rate of $T_2^{-1} \sim \delta\Omega^2\tau_c$. Here, τ_c is the correlation time of these fluctuations [11]. This leads to a very important conclusion that the phase relaxation time may not be determined by the matrix elements between the Kramers doublets (i.e., spin-flip transitions), the most important restriction on the longitudinal SLR.

In order to calculate the fluctuations in the Zeeman frequency Ω, we consider the phonon-induced transitions between the lowest electronic states $|k\rangle$ which possess different spin splitting and are not linked by the time reversal operator. The resulting electron spin phase diffusion due to the spin precession in a fluctuating field is expected to be an efficient relaxation mechanism if (i) the electron significantly changes the spin precession frequency when the transitions between different states $|k\rangle$ occur and (ii) the transitions do not occur too frequently [11]. In the case of a QD, these two conditions can be easily realized due to the shallow energy levels and the g factor or hyperfine constant dependence on the orbital electronic states.

We begin the quantitative analysis by defining the Hamiltonian H over the basis functions consisting of a few lowest electronic states $|k\rangle$, which are involved due to phonon-assisted transitions. We also assume that the single-electron problem in a QD without the Zeeman energy and HFI gives a doubly degenerate energy spectrum E_k with eigenstates $|k\rangle$. It is conveniently assumed (but not necessary) that the spin splitting $|\Omega|$ is small with respect to the energy intervals $|E_k - E_{k'}|$. The specific nature and type of the QD is unimportant for the analysis; in fact, the theory is applicable to any local electron center with spin $S = 1/2$. With these assumptions, the total Hamiltonian takes the form

$$H = H_s + H_e + H_{ph} + H_{e-ph}. \tag{1}$$

The first term H_s is the spin (or pseudospin) energy Hamiltonian which can be reduced to the form $H_s = \vec{\Omega}\,\vec{s}$ in the most general case. With $\Omega_z = \langle\uparrow|H_s|\uparrow\rangle - \langle\downarrow|H_s|\downarrow\rangle$, $\Omega_x = \langle\uparrow|H_s|\downarrow\rangle + \langle\downarrow|H_s|\uparrow\rangle$ and $i\Omega_y = \langle\uparrow|H_s|\downarrow\rangle - \langle\downarrow|H_s|\uparrow\rangle$ are now independent of the spin arguments. Its projection on the lowest electronic states $|k\rangle$ reads (see, for comparison, Refs. [8,9])

$$\vec{\Omega} = \sum_{k,k'} |k\rangle \vec{\Omega}^{k,k'} \langle k'|, \tag{2}$$

where $\vec{\Omega}^{k,k'}$ are the matrix elements of the effective field (in units of energy) taken between the $|k\rangle$ and $|k'\rangle$ states. The spin-independent electron energies describe the Hamiltonian H_e,

$$H_e = \sum_k E_k |k\rangle\langle k|. \tag{3}$$

The Hamiltonians of the lattice and electron-phonon interactions have the usual form

$$H_{ph} = \sum_q \omega_q \left(a_q^+ a_q + \frac{1}{2} \right); \tag{4}$$

$$H_{e-ph} = \sum_{q,k,k'} B_{k,k'}^q |k\rangle\langle k'| (a_q^+ + a_{-q}). \tag{5}$$

Here $q = \{\vec{q}, \kappa\}$ represents the wave vector and polarization of a phonon with energy ω_q; $-q \equiv \{-\vec{q}, \kappa\}$, a_q^+ and a_q are the phonon creation and annihilation operators, $B_{k,k'}^q$ is the matrix element of the electron-phonon interaction, which depends on the material parameters and the geometry of the QD. In Eq. (1), the last three terms constitute the Hamiltonian of the dissipative subsystem responsible for electron spin relaxation, $H_d = H_e + H_{ph} + H_{e-ph}$.

We are interested in the evolution of electron spin $\vec{s}(t) = \mathrm{Tr}\{\rho(t)\vec{s}\}$ [$\rho(t)$ is a density matrix] in a system with the Hamiltonian of Eq. (1). Assuming that the spin relaxation time is much longer than the correlation time of the fluctuating effective field (i.e., $T_2 \gg \tau_c$), one can derive a quantum kinetic equation (see Ref. [11])

$$\frac{d}{dt}\vec{s}(t) = \vec{\omega} \times \vec{s}(t) - \mathbf{\Gamma}(\vec{s}(t) - \vec{s}_0), \tag{6}$$

where $\vec{\omega} = \langle\vec{\Omega}\rangle$ is an effective magnetic field, $\langle\ldots\rangle = \mathrm{Tr}\{e^{-H_d/T}\ldots\}/\mathrm{Tr}\{e^{-H_d/T}\}$, and T is the temperature; $\vec{\omega}$ and T are in units of energy. The matrix $\mathbf{\Gamma}$ of relaxation coefficients is composed of Fourier transformed correlation functions $\gamma_{\mu\nu} \equiv \gamma_{\mu\nu}(\omega) = \langle\delta\Omega_\mu(\tau)\delta\Omega_\nu\rangle_\omega = \frac{1}{2\pi}\int_{-\infty}^{\infty}\langle\delta\Omega_\mu(\tau)\delta\Omega_\nu\rangle e^{i\omega\tau}d\tau$; $\mu, \nu = x, y, z$; $\delta\vec{\Omega} = \vec{\Omega} - \langle\vec{\Omega}\rangle$; $\delta\Omega_\mu(\tau) = e^{iH_d\tau}\delta\Omega_\mu e^{-iH_d\tau}$. With a provision that the correlation functions are symmetrical, $\gamma_{\mu\nu}(\omega) = \gamma_{\nu\mu}(\omega)$, the matrix $\mathbf{\Gamma} = \pi\|\Gamma'_{\mu\nu}\|$ has a simple form in the frame of references related to the direction of the effective field $\vec{\omega} \parallel \hat{z}$: $\Gamma'_{xx} = \gamma_{zz}^0 + n\gamma_{yy}$, $\Gamma'_{yy} = \gamma_{zz}^0 + n\gamma_{xx}$, $\Gamma'_{zz} = n(\gamma_{xx} + \gamma_{yy})$, $\Gamma'_{\mu\nu} = -n\gamma_{\mu\nu}$, ($\mu \neq \nu$), where $\gamma_{zz}^0 = \gamma_{zz}(0)$, $n \equiv n(\omega) = (1 + e^{\omega/T})/2$, $\vec{s}_0 = -\frac{1}{2}\tanh(\omega/2T)\{0, 0, 1\}$.

Thus, the problem of spin relaxation is reduced to the calculation of correlation functions of the effective field operator with the Hamiltonian H_d of the dissipative subsystem. These calculations strongly depend on the specific form of H_d, the energy spectrum, and the quantity of electron states considered. Keeping this context in mind, we consider the simpler problem of electron fluctuations between only two discreet states $|k\rangle = |g\rangle$ or $|e\rangle$ corresponding to the ground state and the first exited (by an interval δ_0) electronic energy level with Zeeman frequencies $\vec{\Omega}^g$ and $\vec{\Omega}^e$. Such a simplification allows us to easily perform all the necessary calculations in an analytical form. In addition, most of the important physics of the new mechanism under consideration can be obtained in the framework of this two-level model.

Hereafter, it is convenient to introduce Pauli matrices $\sigma_1, \sigma_2, \sigma_3$ on the basis $|e\rangle, |g\rangle$, where according to the definition, $\sigma_1, \sigma_2, \sigma_3$ are invariant with respect to the coordinate system rotation in contrast to actual spin matrices $\vec{s}$. In terms of Pauli matrices, the Hamiltonian of a dissipative subsystem takes the form $H_d = H_{ph} + \frac{1}{2}\delta_0\sigma_3 + \Sigma_q B_q \sigma_1 (a_q^+ + a_{-q})$. The electron spin Hamiltonian now assumes the form $H_s = \frac{1}{2}(\vec{\Omega}^e + \vec{\Omega}^g)\vec{s} + \frac{1}{2}(\vec{\Omega}^e - \vec{\Omega}^g)\sigma_3\vec{s}$, which defines the fluctuating part of the effective field $\delta\vec{\Omega} = \frac{1}{2}(\vec{\Omega}^e - \vec{\Omega}^g)(\sigma_3 - \langle\sigma_3\rangle)$ with $\langle\sigma_3\rangle = -\tanh(\delta_0/2T)$ and gives the correlation functions in the form [see the description following Eq. (6) for reference]

$$\gamma_{\mu\nu}(\omega) = \tfrac{1}{4}(\Omega_\mu^e - \Omega_\mu^g)(\Omega_\nu^e - \Omega_\nu^g) J_\omega(T); \tag{7}$$

$$J_\omega(T) = \langle\!\langle[\sigma_3(\tau) - \langle\sigma_3\rangle](\sigma_3 - \langle\sigma_3\rangle)\rangle\!\rangle_\omega. \tag{8}$$

This correlation function $\langle\!\langle[\sigma_3(\tau) - \langle\sigma_3\rangle](\sigma_3 - \langle\sigma_3\rangle)\rangle\!\rangle$ can be calculated by using the double-time Green's function $G(t, t') = \langle\!\langle\sigma_3(t); \sigma_3(t')\rangle\!\rangle$. Once solved with the Hamiltonian H_d in a usual mean field approximation, $\langle\!\langle a_q^+ a_q \sigma_3; \sigma_3\rangle\!\rangle \approx \langle a_q^+ a_q\rangle G(t, t')$, the resulting Green's function Fourier image $G(\omega)$ is related to Eq. (8) as $G(\omega + i0) - G(\omega - i0) = -i(1 + e^{\omega/T})[J_\omega(T) + \langle\sigma_3\rangle^2]$ [12]. The final expression takes the following form for the case $\omega \ll \delta_0$:

$$J_\omega(T) = \frac{1 - \langle\sigma_3\rangle^2}{\pi n(\omega)} \frac{\tau_c}{\omega^2\tau_c^2 + 1}; \tag{9}$$

$$\tau_c^{-1} = 2\pi \sum_q |B_q|^2 (2n_q + 1)\delta(\omega_q - \delta_0), \tag{10}$$

where $n_q = \langle a_q^+ a_q\rangle$ is the phonon population factor for mode q. The parameter τ_c has the simple physical meaning of the correlation time caused by phonon-assisted transitions between the $|g\rangle$ and $|e\rangle$ states.

Actually, Eqs. (7)–(10) describe the problem under consideration in a very general form. Before we specify the electron spin interaction, which fluctuates due to the phonon-induced transitions between the $|g\rangle$ and $|e\rangle$ states, we provide an analysis of the SLR temperature dependence. In doing so, we note that the correlation time τ_c given in Eq. (10) can be written as $\tau_\delta \tanh(\delta_0/2T)$, where $\tau_\delta = [2\pi\Sigma_q|B_q|^2\delta(\omega_q - \delta_0)]^{-1}$ is the lifetime of the excited electron state with respect to the transition to the ground state through phonon emission in the limit $T \rightarrow 0$. Thus, for the $\omega = 0$ component (e.g., γ_{zz}^0), the temperature dependence of SLR is reduced to

$$J_0(T) = \frac{\tau_\delta}{\pi} F\left(\frac{\delta_0}{2T}\right); \qquad F(x) = (1 - \tanh^2 x)\tanh x. \quad (11)$$

As shown in Fig. 1, the pronounced maximum in the temperature dependence of $F(\delta_0/2T)$ around $T = \delta_0$ has a simple physical meaning. The left of the peak corresponds to the reduced hopping from the $|g\rangle$ to the $|e\rangle$ state that decreases the difference $\vec{\Omega}^g - \langle\vec{\Omega}^g\rangle$ (or the amplitude of fluctuations). So, in the limit $T \ll \delta_0$ the fluctuations are frozen out and our mechanism becomes noneffective as $F(\delta_0/2T) \rightarrow \exp(-\delta_0/T)$. The negative slope on the right side (high temperature) arises due to the well-known effect of fluctuation dynamical averaging, which becomes more pronounced with an increase in temperature.

As noted above, the correlation function $\pi\gamma_{zz}^0 = \frac{\pi}{4}(\Omega_z^e - \Omega_z^g)^2 J_0(T)$ describes the rate T_2^{-1} of spin relaxation if longitudinal fluctuations dominate over transversal ones, $\gamma_{zz}^0 \gg \gamma_{xx}(\omega), \gamma_{yy}(\omega)$. Let us apply the general theory discussed above to the mechanism of phase relaxation, which stems from the hopping between excited and ground states with different g factors. In the most general case, the reason for such a difference is the g-factor dependence on the energy separation between the electron's discrete level and the nearest spin-orbital split electronic band. For technologically significant III-V compounds, where the interaction with the valence band edge determines the deviation of the electron g factor from the free electron Landé factor $g_0 \approx 2$ [13], one can find the amplitude of the fluctuation $\Delta g = \delta_0(g_0 - g)(\Delta_{so} + 2E_g)/E_g(\Delta_{so} + E_g)$, where E_g is the band gap and Δ_{so} the spin-orbital splitting of the valence band; we also assume inequality $\delta_0 \ll E_g$. In the case of a Si QD, E_g is a splitting E_{15} of the Δ point in the Brillouin zone [9]. The relaxation mechanism due to the g-factor anisotropy of the Δ band is not effective for spin flip in Si QDs because of the specific valley orbital structure [3]. The final equation for the phonon-assisted rate of phase relaxation caused by the Zeeman energy fluctuations is given by

$$T_{2,Z}^{-1} = \frac{(g_0 - g)^2}{4g^2}\left(\frac{\delta_0(\Delta_{so} + 2E_g)}{E_g(\Delta_{so} + E_g)}\right)^2 \omega_0^2 \tau_\delta F\left(\frac{\delta_0}{2T}\right), \quad (12)$$

where $\hbar\omega_0 = g\mu_B B$ and μ_B is the Bohr magneton. One can see that our mechanism reveals a quadratic dependence of $T_{2,Z}^{-1}$ on the applied magnetic field B in contrast to the $B^4 \sim B^5$ dependence found in the previous calculations of longitudinal SLR through the direct processes between the Kramers doublets (with absorption or emission of one resonant phonon) [1–3,8,9]. Note that the spin-flip transitions via excited states involve nonresonant phonons and can be characterized by a quadratic dependence on B. However, numerical estimations show that these processes can be efficient only at high temperatures [3,14].

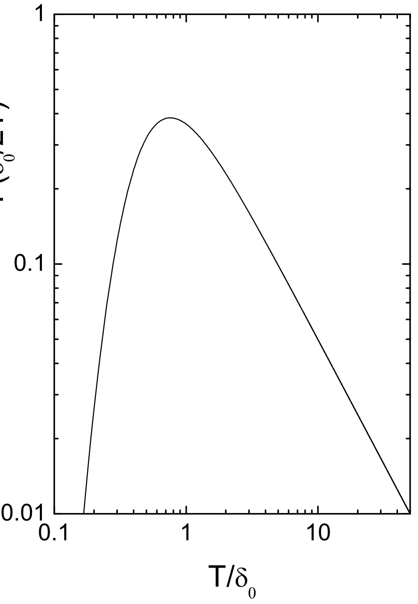

FIG. 1. Decoherence factor $F(\delta_0/2T)$ [Eq. (11)] as a function of temperature.

An estimation of excited state lifetime τ_δ can be performed in terms of a deformation potential interaction and a model of lateral carrier confinement [3]. The matrix element of electron-phonon interaction between the $|g\rangle$ and $|e\rangle$ states in this model is $B_q = iC\sqrt{\hbar q/2\rho v_\parallel V_0} J_{osc}$, where $J_{osc} = J_{osc}(\vec{q})$ is a corresponding form factor calculated in Ref. [3], C is the deformation potential, and ρ, $v_\parallel$ and V_0 are the density, longitudinal sound velocity, and volume of crystal, respectively. A straightforward calculation of inverse lifetime results in the expression

$$\tau_\delta^{-1} = \frac{C^2 q_\delta^3 \alpha}{32\pi^2\hbar\rho v_\parallel^2}\int_0^1 (1 - z^2)e^{-\alpha(1-z^2)}dz, \qquad (13)$$

where $q_\delta = \delta_0/\hbar v_\parallel$, $\alpha = \delta_0/2m_e v_\parallel^2$, and m_e is a lateral effective mass.

To show the efficiency of the mechanism under consideration, we assume $\delta_0 = 2$ meV [3] and calculate the relaxation parameters of Eqs. (13) and (12) and for a GaAs QD under the magnetic field $B = 1$ T. We find that $\tau_c \simeq \tau_\delta = 5.8 \times 10^{-9}$ s, and $T_{2,Z} = 2.0$ s, 1.8×10^{-5} s, and 5.4×10^{-8} s for $T = 1$ K, 2 K, and 4 K, respectively. Similar calculations were provided for a Si QD with the same magnitudes of δ_0, B, and T: $\tau_c = 3.5 \times 10^{-9}$ s, and $T_{2,Z} = 2.6 \times 10^6$ s, 24 s and 0.073 s. Comparing these data with the T_1 calculation of Ref. [3] shows that in spite of the strong suppression of relaxation in the Si QD due to a small deviation $g_0 - g$, the phonon-induced g-factor fluctuation via excited states can control the phase relaxation (i.e., $T_{2,Z} < T_1$) at $T \gtrsim 2$ K. The slight anisotropy of the g factor [10] results in some T_2 dependence on the magnetic field direction. However, this effect is expected to be small (less than 25%) in contrast to the strong magnetic anisotropy of T_1 [3].

A qualitatively different situation arises in the case of HFI modulation by the phonon-assisted transitions. The distinctive feature of this mechanism is an uncontrolled dispersion of the local nuclear field over the ensemble of QDs due to the random distribution of nuclear spins. This dispersion accounts for the fast (but partial) loss of initial electronic polarization of the QD aggregate without spin coherence loss [6,7]. Thus, we take into account the spin relaxation of a typical QD with a mean value of nuclear field dispersion $\delta\Omega_n = a\sqrt{\frac{2}{3}I(I+1)\varkappa n_I/V_{\mathrm{QD}}}$, where I and n_I are the nuclear spin and its concentration in a QD of volume V_{QD}; the dimensionless parameter $\varkappa = V_{\mathrm{QD}} \int(|\Psi_e(\vec{r})|^2 - |\Psi_g(\vec{r})|^2)^2 d^3\vec{r}$ is equal to $9/16\pi$ within the approximations of Ref. [3] and a is a constant in the HFI Hamiltonian $H_{hf} = a\vec{s}\,\vec{I}\,|\Psi(\vec{R})|^2$ for the nuclear spin $\vec{I}$ located at a point $\vec{R}$ and an electron with its envelope function $\Psi(\vec{r})$. If we set $\Omega_z^e - \Omega_z^g = \delta\Omega_n$ in Eq. (7), we readily find the following estimation:

$$T_{2,hf}^{-1} = \frac{\varkappa}{6} I(I+1) \frac{a^2 n_I}{\hbar^2 V_{\mathrm{QD}}} \tau_\delta F\left(\frac{\delta_0}{2T}\right). \tag{14}$$

This equation shows the independence of $T_{2,hf}$ on the magnetic field in the case of HFI-induced transitions. An estimation of Eq. (14) (with an appropriate HFI averaging over ^{69}Ga, ^{71}Ga, and ^{75}As [15]) for a GaAs QD with a typical size of 50 Å × 500 Å × 500 Å and $\delta_0 = 2$ meV gives $T_{2,hf} = 34$ s, 3.2×10^{-4} s, and 1.0×10^{-6} s for $T = 1$ K, 2 K, and 4 K, respectively. A similar estimation performed for a Si QD with the same dimensions and temperatures results in $T_{2,hf} = 4.1 \times 10^7$ s, 3.8×10^2 s, and 1.2 s, respectively. One can see that for the considered cases of both GaAs and Si QDs, the contribution of HFI to the phonon-assisted relaxation is small in comparison to that of the g-factor modulation mechanism at $B = 1$ T. However, the role of HFI modulation mechanism described in Eq. (14) should prevail at weaker magnetic fields ($B < 0.1$ T). The common property of Eqs. (12) and (14) are their inverse proportionality to the rate of phonon-induced transitions unlike those based on the spin-flip transition probabilities [4].

Our formulas were obtained under the assumption of a two-level electronic structure (with regard to the orbital degree of freedom). In a similar manner, one can consider a many-level case. For example, the N_e excited electron levels close together with an identical g factor or HFI can be taken into account by replacing the $\tanh x$ term with a function $(N_e e^x - e^{-x})/(N_e e^x + e^{-x})$ in Eq. (11).

When considering the HFI as a mechanism of spin relaxation, we restricted our investigation to the SLR of single, but typical, QD with a mean configuration of nuclear spins. It is important to bear in mind that the relaxation measurements performed on a large ensemble of QDs correspond to averaging the relaxation curves over random $T_{2,hf}$ instead of the single spin relaxation with a mean $T_{2,hf}$ value. As a result, our mechanism would predict nonexponential decay of initial electron magnetization in a QD ensemble.

In conclusion, we considered an efficient mechanism of electron spin decoherence in a single QD due to the fluctuating precession of the longitudinal (with respect to the direction of external magnetic field) effective magnetic field. Compared to the SLR controlled by the spin-flip transitions, our mechanism does not involve transitions between the Kramers doublets, which leads to effective spin relaxation characterized by a smooth dependence on the applied magnetic field. On the other hand, the mechanism under consideration reveals an exponential dependence on the temperature when $T \ll \delta_0$. Although detailed estimation of the spin relaxation rate depends on the specific properties of the respective QDs, the analysis on the typical cases illustrates an advantage of Si QDs for the quantum computing applications due to their very long relaxation time.

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