Correlation of phonon decay with localized electron spin-phase diffusion

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A spin decoherence mechanism is proposed for localized electrons. The irregular phonon phase disturbances originated from phonon relaxation can influence electron spin precession with a net effect of spin phase decay. A quantitative analysis demonstrates relatively high efficiency of this mechanism in the low temperature and low magnetic field regime compared to the spin-flip processes.

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I. INTRODUCTION

Recently, much attention has been devoted to electron spin relaxation in quantum dots (QDs) since they provide a natural candidate for the qubits in quantum computing. A typical approach to this problem is to calculate the spin transition probability associated with the spin-flip processes, i.e., longitudinal spin relaxation. However, quantum computing is qualitatively limited by the processes that result in the destruction of electron spin phase coherence. For example, phase diffusion of localized electron spin can be characterized by relaxation mechanisms that are not related to spin-flip processes under certain conditions. Hence, a further investigation of *transversal* (or phase) relaxation T_2 is crucial for an accurate understanding.

One such mechanism was proposed in Ref. 1 where a random change of spin precession and subsequent spin phase diffusion is associated with the transitions between electronic quantum states with different g factors. Although generally efficient, this process is frozen out at low temperatures due to its phonon-mediated nature and the direct spin-flip is expected to be the dominant mechanism of phase relaxation. However, the spin-flip relaxation reveals a very strong (4th to 5th power) dependence on the magnetic field,[2,3] becoming rather ineffective at low fields. Hence, it is necessary to explore other potential sources of decoherence, particularly in the low field and low temperature regime. In this work, we show that the spin-phonon interaction, which heretofore was considered mainly with respect to the resonant processes, can provide such a mechanism if a finite phonon damping is taken into account.

Our analysis is based on the representation of spin-phonon interaction in terms of fluctuating effective magnetic field $\vec{\Omega}$ (in units of energy) acting on the electron spin $\vec{s}$. This field is assumed to be composed of additive contributions $\vec{\Omega}_p$ from each phonon $p=\{\vec{q},\varkappa\}$ with a wave vector $\vec{q}$ and polarization $\varkappa$, i.e., $\vec{\Omega}=\Sigma_p\vec{\Omega}_p$. For the moment, let us focus on a single phonon contribution. Then, in the frame of reference rotating with the Zeeman frequency, the electron spin performs precession around the small $\vec{\Omega}_p$, which oscillates with a phonon frequency ω_p. No alteration in the electron spin phase occurs due to such a harmonic perturbation with a possible exception of spin phase shift $\Delta\phi_0$ acquired at the initial period of interaction $0<t<2\pi/\omega_p$ due to a random phonon phase θ_p.[4]

A different situation can be realized when a phonon harmonic oscillation is interrupted and resumes at a series of instant times t_{1i} and t_{2i} $(i=0,1,\ldots)$, respectively. The reason of such phonon fluctuations can be lattice anharmonicity, phonon scattering at the impurities or lattice defects, etc. These irregular phonon perturbations affect the electron spin precession resulting in the phase shift $\Delta\phi_i$ at each interval of time $t_{2i}-t_{1i}$. Subsequently, the net effect of spin phase change $\phi_p(t)$ due to a phonon mode p can be expressed as $\phi_p(t)=\Sigma_i\Delta\phi_i$, $(t_{2i}<t)$.

Note that for a large number of small changes $\Delta\phi_i$, their total effect can be described by a diffusion equation. Its solution leads to an exponential decay of electron spin phase with a relaxation rate $T_p^{-1}=1/2\langle\Delta\phi_i^2\rangle\tau_p^{-1}$, where τ_p is the mean time between sequential instants t_{1i} (or t_{2i}).[5] To estimate the spin phase change $\Delta\phi_i$ caused by a phonon perturbation during the $t_{2i}-t_{1i}$, it is helpful to recognize that a single oscillator influence does not change a spin phase during its full period $\Delta t_p=2\pi/\omega_p$ as well as for any n integer periods $n2\pi/\omega_p$. Hence, $\Delta\phi_i$ can be approximated as a spin rotation $\Omega_p\Delta t_p$ in an effective field $\vec{\Omega}_p$ independently on duration $t_{2i}-t_{1i}$. With the mean value $\langle\Delta\phi_i\rangle$ on the order of Ω_p/ω_p, one can expect $T_p^{-1}\sim\tau_p^{-1}\Omega_p^2/\omega_p^2$ for the phonon mode p and $T_2^{-1}\sim\Sigma_p N_p\tau_p^{-1}\Omega_p^2/\omega_p^2$ when the contributions of all phonons (with the population factor N_p) are taken into account.

The qualitative consideration provided above shows that electron spin phase relaxation can be strongly affected by phonon phase damping of any origin such as phonon decay. Moreover, since this mechanism does not involve energy exchange, only the longitudinal (with respect to the external magnetic field $\vec{B}$) component Ω_z of the effective fluctuating field is relevant to our case. These characteristics qualitatively distinguish the mechanism under consideration from other processes, most of which are determined by fluctuations of transversal components Ω_x and Ω_y at the resonant frequency with the Zeeman splitting.

II. THEORETICAL MODEL

For a detailed quantitative analysis of the proposed mechanism, let us start with the spin–phonon interaction operator,

$$H_{s-ph} = \vec{\Omega}\vec{s}, \tag{1}$$

where the αth component ($\alpha = x,y,z$) of the fluctuating field takes a form linear in the creation and annihilation operators $a_p^\dagger$ and a_p of the phonon mode p [$-p \equiv \{-\vec{q}, \varkappa\}$]; i.e.,

$$\Omega_\alpha = \sum_p V_\alpha^p Q_p \equiv \sum_p V_\alpha^p (a_p^\dagger - a_{-p}), \tag{2}$$

with a matrix element V_α^p of the spin–phonon interaction. The specific form of V_α^p will be discussed later.

Now we focus on the spin evolution caused by random fluctuations of Ω_α . Obviously electron spin follows each of such fluctuations that result in its irregular behavior at the time scale τ_c of the Ω_α fluctuations. Actually a random single spin fluctuation associated with each phonon scattering is expected to be very small and drops out of the problem; instead, the total result of these small fluctuations averaged over the time scale Δt ($\tau_c \ll \Delta t \ll T_2$) is the subject of our investigation. The time evolution of mean spin value $\vec{s}$ can be described by the quantum kinetic equation[5] in the case of anisotropic medium and interaction H_{s-ph} [Eq. (1)],

$$\frac{d}{dt}\vec{s}(t) = \vec{\omega} \times \vec{s}(t) - \mathbf{\Gamma}[\vec{s}(t) - \vec{s}_0], \tag{3}$$

where $\vec{\omega}$ is an effective field with components $\omega_i = \Sigma_j\, g_{ij}\mu_B B_j$.[6] As usual, g_{ij} are the components of a g tensor, the subscripts i and j relate to the crystalline coordinate system, μ_B is the Bohr magneton, $\langle \ldots \rangle = \mathrm{Tr}\{e^{-H_d/T}\ldots\}/\mathrm{Tr}\, e^{-H_d/T}$ where H_d is the Hamiltonian of the dissipative subsystem (lattice vibrations in our case), and T is the temperature. $\vec{\omega}$ and T are expressed in units of energy. The matrix $\mathbf{\Gamma}$ of relaxation coefficients is composed of Fourier transformed correlation functions,

$$\gamma_{\mu\nu} \equiv \gamma_{\mu\nu}(\omega) = \langle \Omega_\mu(\tau)\Omega_\nu \rangle_\omega = \frac{1}{2\pi}\int_{-\infty}^{\infty} \langle \Omega_\mu(\tau)\Omega_\nu \rangle e^{i\omega\tau} d\tau, \tag{4}$$

with $\Omega_\mu(\tau) = \exp(iH_d\tau)\Omega_\mu \exp(-iH_d\tau)$. It has a canonical form in the frame of references $\hat{x}$, $\hat{y}$, $\hat{z}$ with $\hat{z}$ directed along $\vec{\omega}$ (so that μ, $\nu = x, y, z$). With a provision that the correlation functions are symmetrical, $\gamma_{\mu\nu}(\omega) = \gamma_{\nu\mu}(\omega)$, the matrix Γ has a simpler form: $\Gamma_{xx} = \pi(\gamma_{zz}^0 + n\gamma_{yy})$, $\Gamma_{yy} = \pi(\gamma_{zz}^0 + n\gamma_{xx})$, $\Gamma_{zz} = \pi n(\gamma_{xx} + \gamma_{yy})$, $\Gamma_{\mu\nu} = -\pi n \gamma_{\mu\nu}$, $(\mu \neq \nu)$, where $\gamma_{zz}^0 = \gamma_{zz}(0)$, $n \equiv n(\omega) = (1 + e^{-\omega/T})/2$, $\vec{s}_0 = -1/2\hat{z}\tanh(\omega/2T)$, $\omega = \omega_z = (\Sigma_i\, \omega_i^2)^{1/2}$.

One can see that the coefficients Γ_{xx} and Γ_{yy} responsible for transversal relaxation consist of two parts, $T_{2,\omega}^{-1} = \pi n \gamma_{yy}$ (or $\pi n \gamma_{xx}$) and $T_{2,0}^{-1} = \pi \gamma_{zz}^0$. A comparison with the longitudinal relaxation coefficient Γ_{zz} shows that the term $T_{2,\omega}^{-1}$ stems from the contribution of spin-flip processes involving energy exchange between the Zeeman and phonon reservoirs. Since the longitudinal relaxation has been the subject of a number of recent studies,[2,3,7,8] we focus on the analysis of the $T_{2,0}^{-1}$ term.

The correlation function Fourier image [Eq. (4)] of the effective field $\vec{\Omega}$ is expressed in terms of the phonon operators according to Eq. (2). In turn, the Fourier image of phonon correlation functions $\varphi_p(\omega) = \langle Q_p(\tau)Q_{-p} \rangle_\omega$ is Lorentzian-like since the corresponding Green function satisfies the equation $G_p(\omega) = (\omega_p/\pi)[\omega^2 - \omega_p^2 - 2\omega_p M_p(\omega)]^{-1}$, where the "mass" operator $M_p(\omega)$ depends on the phonon interaction (see, for example, Ref. 9). In the most general case, this correlation function takes the form[10]

$$\varphi_p(\omega) = \frac{1}{\pi}\frac{(2N_p + 1)\Gamma_p(\omega)}{(\omega^2 - \omega_p^2)^2/\omega_p^2 + \Gamma_p^2(\omega)}, \tag{5}$$

where $\Gamma_p(\omega) = \mathrm{Im}\, M_p(\omega)$ depends on the specific mechanism of phonon scattering. In such a manner, $\Gamma_p(\omega)$ is a function of temperature due to the anharmonicity of the third and fourth order; furthermore, there are contributions by other sources of phonon scattering (point defects, isotopes, dislocations, crystal boundaries, and interfaces) that reveal different dependencies on ω and ω_p. Hence, an evaluation of the relaxation coefficients becomes too complicated to be approached analytically. Instead, to proceed further, we utilize the phonon relaxation time that can be extracted from the thermal conductivity measurements (see Ref. 11 and the references therein). An expression appropriate for the correlation function Fourier image $\gamma_{\mu\nu}(\omega)$ was derived in Ref. 5 in the relaxation time approximation. For our particular case of $\omega = 0$ and $\mu = \nu = z$, it can be reduced to

$$\gamma_{zz}^0 = \sum_{p_1,p_2} V_z^{p_1} V_z^{p_2} \langle Q_{p_1} Q_{p_2} \rangle \frac{1}{\pi}\frac{\tau_{p_1}^{-1}}{\omega_{p_1}^2 + \tau_{p_1}^{-2}}, \tag{6}$$

where $\tau_p = 1/\Gamma_p(\omega_p)$ is the relaxation time of phonon mode p (i.e., phonon lifetime). In most cases, one can assume $\omega_p \gg \tau_p^{-1}$ and neglect the second term in the denominator of Eq. (6). Then, along with the definition of the operator Q_p [see Eq. (2)], one can express the nonresonant phonon contribution to the transversal spin relaxation rate in the form

$$T_{2,0}^{-1} = \sum_p |V_z^p|^2 (2N_p + 1)\frac{\tau_p^{-1}}{\omega_p^2}, \tag{7}$$

which is in accordance with the qualitative analysis discussed earlier in this paper. The phonon population factor N_p is given as $[\exp(\omega_p/T) - 1]^{-1}$.

Equation (7) is the starting point of our investigation on the proposed spin relaxation mechanism. However, this still requires detailed knowledge of the phonon dispersion ω_p and the relaxation time τ_p for each phonon mode p. By taking into account the conditions frequently encountered in quantum computation utilizing semiconductor QDs, we restrict our consideration to the case when the radius a_0 of the electron state is much larger than the lattice constant and the temperature is sufficiently low. Since the spin–phonon interaction matrix V_z^p is significant only for the phonon wave vector $q \lesssim 1/a_0$, a large a_0 essentially limits the summation of Eq. (7) to long wavelength phonons. Subsequently τ_p, which is a complex function of the temperature and phonon frequencies,[12] can be considered in the long wavelength limit. Moreover, at low enough temperatures $T \lesssim T_{bs}$ ($T_{bs} \approx 10$ K in the case of Ref. 12), only one term originating from the boundary scattering survives for phonon relaxation.[11] Since this mechanism is insensitive to the tem-

perature as explained by Ref. 11, it is adequate to assume a constant phonon relaxation time $\tau_p \simeq \tau_{ph}$ for long wavelength phonons at $T \lesssim T_{bs}$. This permits us to avoid the problems associated with the complex dependence of ω_p and τ_p, which can be very specific for each particular sample.

A. Effect of *g*-factor fluctuation

To evaluate V_z^p, we consider the spin–lattice interaction via phonon modulation of g factor. In general, the spin–lattice interaction Hamiltonian can be written in terms of the tensor A_{ijkl}:[13,14,3]

$$H_{s-ph} = \sum_{ijkl} A_{ijkl}\mu_B B_i s_j \overline{u_{kl}}, \tag{8}$$

where $\overline{u_{kl}}$ is the strain tensor u_{kl} averaged over the electron ground state $|g\rangle = \psi_g(\vec{r})$: $\overline{u_{kl}} = \langle g|u_{kl}|g\rangle$. By way of an important example, we consider a z-directed magnetic field and a localized electron with the axial symmetry with respect to the z-axis. This reduces Eq. (8) to the form of Eq. (1) with $\Omega_z = [(A_{33} - A_{31})\overline{u_{zz}} + A_{31}\overline{\Delta}]\mu B$; here, Δ denotes the dilatation $\Delta = \Sigma_i u_{ii}$ and the Voigt notation is adopted ($A_{33} = A_{zzzz}$, $A_{31} = A_{zzxx}$, $A_{66} = A_{xyxy}$). Then, the matrix element of the spin–phonon interaction takes the expression

$$V_z^p = i\left(\frac{\hbar}{2\rho V\omega_p}\right)^{1/2} [(A_{33} - A_{31})e_z^p q_z + \delta_{\varkappa,L}A_{31}q]\Phi(\vec{q})\mu_B B, \tag{9}$$

where ρ is the mass density of the crystal, V is the volume of the sample structure, $\vec{e}^p$ the polarization vector of the phonon mode p, $=L,T$, and $\Phi(\vec{q}) = \langle g|e^{i\vec{q}\cdot\vec{r}}|g\rangle$. The spin-lattice relaxation rate in Eq. (7) can be calculated by treating the phonon modes based on the isotropic elastic continuum model with the longitudinal and transverse sound velocities c_L and c_T. Assuming the axial symmetry for the local electron center, i.e., $\Phi(\vec{q}) = \Phi(x,z)$ ($x = qa_0/2$, $z = q_z/q$, the parameter a_0 represents the electron state radius as mentioned before), one can obtain

$$T_{2,0}^{-1} = \tau_{ph}^{-1}\xi(B)\int_0^{x_{\max}} x\frac{\tau_p^{-1}}{\tau_{ph}^{-1}}\left[\coth\left(\frac{T_T^{eff}}{T}x\right)F_T(x) + \frac{c_T^3}{c_L^3}F_L(x)\coth\left(\frac{T_L^{eff}}{T}x\right)\right]dx, \tag{10}$$

$$\xi(B) = \frac{(A_{33} - A_{31})^2\mu_B^2 B^2}{2\pi^2\hbar\rho c_T^3 a_0^2},$$

$$F_L(x) = \int_{-1}^{1}(z^2 + \zeta)^2\Phi^2(x,z)dz,$$

$$F_T(x) = \int_{-1}^{1} z^2(1 - z^2)\Phi^2(x,z)dz, \tag{11}$$

where τ_{ph}^{-1} is an average phonon relaxation rate, $T_\varkappa^{eff} = \hbar c/k_B a_0$ is the effective temperature, and $\zeta \equiv A_{31}/(A_{33} - A_{31}) = -1/3$ if one assumes that the strain induced part of the effective g-tensor $\tilde{g}_{ij} = \Sigma_{k,l}A_{ijkl}u_{kl}$ is characterized by zero trace, i.e., $A_{33} + 2A_{31} = 0$. When a_0 is much larger than the lattice constant, the upper limit $x_{\max}$ in the integral of Eq. (10) may be taken to infinity since $\Phi(\vec{q})$ restricts the actual phonon wave vectors to $q \lesssim 1/a_0$ as discussed above.

Let us evaluate spin relaxation of a shallow donor with an effective Bohr radius a_B ($=a_0$) and $\Phi(x,z) = (1 + x^2)^{-2}$. Utilizing the constant phonon relaxation time approximation $\tau_p \simeq \tau_{ph}$ for $T \lesssim T_{bs}$, the integral in Eq. (10) can be evaluated analytically,

$$T_{2,0}^{-1} = \frac{2\xi(B)\tau_{ph}^{-1}}{45}\left(\sqrt{1 + \frac{T^2}{T_\perp^2}} + \frac{2c_T^3}{3c_L^3}\sqrt{1 + \frac{T^2}{T_\parallel^2}}\right), \tag{12}$$

where $T_{\parallel(\perp)} = (16/15\pi)T_{L(T)}^{eff}$. Note that Eq. (12) is obtained with $\vec{B}\parallel[001]$. In the case of cubic symmetry [where only two constants A_{66} and $A_{33} = -2A_{31}$ in Eq. (8) describe the effect of spin–phonon coupling], an expression $T_{2,0}^{-1}$ for an arbitrarily directed $\vec{B}$ can be obtained in terms of the direction cosines $l = B_x/B$, $m = B_y/B$, $n = B_z/B$. Our calculations show that this is achieved by multiplying the factor

$$f(\vec{B}/B) = 1 + \left(\frac{4}{9}\frac{A_{66}^2}{A_{33}^2} - 1\right)P \tag{13}$$

to Eq. (12); $P = 3(l^2m^2 + m^2n^2 + n^2l^2)$, $0 \leqslant P \leqslant 1$. One can see that the angular dependence of our mechanism does not result in zero relaxation under any direction of $\vec{B}$. Moreover, the directions along the principal axes ([001], etc.) can result in maximal relaxation, while the same directions sometimes forbid the spin-flip processes.[13,15]

As an example, we consider a phosphorus shallow donor in Si with $a_B = 1.8$ nm. The phonon relaxation time can be extracted from the low temperature measurements of Si thermal resistivity[16] in terms of the theory developed in Refs. 17 and 11 ($\tau_{ph} = 2.4 \times 10^{-8}$ s). The spin–phonon coupling constants were estimated in the works of Refs. 13 and 15. However, we believe that direct determination of coupling constants by means of EPR measurements of Si:P under an applied stress gives more reliable data. A corresponding experiment was performed in Ref. 18, where the constant $A_{66} = 0.44$ was found. Similarly, our estimation obtained $A_{33} = 0.31$ and $A_{31} = -0.155$ that gives $T_{2,0}^{-1} = 1.3 \times 10^{-4}$ s^{-1} at the magnetic field of 1 T and low temperatures $T \ll T_{\parallel(\perp)} \simeq 10$ K.

In another important case of a Si shallow donor in $Al_{0.4}Ga_{0.6}As$, the data on EPR under a uniaxial stress[19] provide rather strong spin–phonon constants of $A_{33} = 19.6$ and $A_{31} = -9.8$. This gives the estimation $T_{2,0}^{-1} = 6.1 \times 10^{-2}$ s^{-1} and 6.1×10^{-4} s^{-1} for the magnetic fields of 1 T and 0.1 T, respectively, at $T = 4$ K under the assumption that phonon lifetimes are identical in these crystals.

Similar calculations can be performed for an electron localized in a QD of $L_{xy} = 2a_0$ in the lateral width and $L_w = \epsilon L_{xy}$ in the thickness. Under the condition $\epsilon \lesssim 0.1$, an approximate formula takes the form

$$T_{2,0}^{-1} = \xi(B)\tau_{ph}^{-1}\left(\sum_{i=L,T} b_i \sqrt{c_i^2 + d_i^2 \frac{T^2}{T_i^2}}\right), \tag{14}$$

where the fitting coefficients are $b_T=1$, $b_L=c_T^3/c_L^3$, $c_T=0.33-1.27\epsilon^2$, $d_T=0.35-0.395\epsilon^2$, $c_L=0.97-28.5\epsilon^2$, and $d_L=0.40-3.76\epsilon^2$.

Let us compare, as an example, spin phase relaxation caused by the phonon decay [Eq. (14)] with the spin-flip admixture mechanism (Ref. 2) in a GaAs QD with $L_w=3$ nm and $L_{xy}=25$ nm, assuming $\tau_{ph}=2.4\times10^{-8}$ s and $A_{33}=19.6$. For the relatively strong magnetic field of 1 T and $T=4$ K, our mechanism and the spin-flip mechanism give $T_{2,0}^{-1}\approx0.1$ s^{-1} and $1/2T_1^{-1}=T_{2,\omega}^{-1}=10$ s^{-1}, respectively, while for $B=0.1$ T both mechanisms predict almost the same rate of $\approx10^{-3}$ s^{-1}. In lower magnetic fields, our mechanism prevails.

B. Effect of hyperfine constant modulation

The g-factor modulation described in Eq. (8) is not the only possible mechanism of spin–phonon interaction. For an alternative process, let us consider the hyperfine interaction (HFI) of localized electrons with the nuclei:

$$H_{hf} = a_{hf}\sum_j |\psi(\vec{r}_j)|^2 \vec{I}_j \vec{s}, \tag{15}$$

where a_{hf} is the HFI constant and $\vec{I}_j$ is the nuclear spin situated at site j with the position $\vec{r}_j$. Lattice vibrations near the nuclear equilibrium positions can lead to effective field fluctuations and, subsequently, the spin–phonon interaction. Taking into account the long wavelength phonons with respect to the mean internuclear distance $\approx n_i^{-1/3}$ (n_i is the nuclear spin concentration), the main part of this interaction for a typical nuclear spin configuration can be represented as in Eq. (1) with

$$\vec{\Omega} = \hat{n}\sqrt{I(I+1)n_i/V_{QD}}\,a_{hf}\overline{\Delta}. \tag{16}$$

Here, the unit vector $\hat{n}$ is directed along the effective nuclear field defined by Eq. (15) and $V_{QD}=(\int|\psi(\vec{r})|^4 d^3\vec{r})^{-1}$. A calculation of the phase relaxation rate for the case of a shallow donor results in the expression, which is similar to Eq. (12),

$$T_{2,0}^{-1} = \frac{\xi_{hf}\tau_{ph}^{-1}}{3}\sqrt{1+\frac{T^2}{T_\parallel^2}}, \tag{17}$$

where the parameter

$$\xi_{hf} = \frac{I(I+1)n_i a_{hf}^2}{6\pi^2\hbar\rho V_{QD}c_L^3 a_0^2}, \tag{18}$$

is independent on the magnetic field. In the case of an electron localized in a QD, one can find the approximate rate through an analogy with Eq. (14):

$$T_{2,0}^{-1} = \xi_{hf}\tau_{ph}^{-1}\sqrt{c_{hf}^2 + d_{hf}^2\frac{T^2}{T_L^2}}, \tag{19}$$

where $c_{hf}=3.7-68\epsilon^2$, $d_{hf}=2.7-9.8\epsilon^2$, and $\epsilon\lesssim0.1$. Numerical estimations provided for a donor in Si and GaAs in terms of Eq. (17) indicate the inefficiency of this mechanism with a very long relaxation time (about 10^{14} s and 10^8 s, respectively). Hence, this mechanism can be neglected in most cases.

C. Two phonon process

So far, we primarily considered the influence of phonon decay on spin phase relaxation via linear spin–phonon interaction as given in Eq. (8). Namely, the effect of phonon scattering with an electron spin on phonon relaxation has not been considered (i.e., electron spin-induced phonon decay). The Hamiltonian of this process can be derived in terms of spin–two-phonon interaction $H_{s-ph}^{(2)}=\Sigma D_{ijklmn}\mu_B B_i s_j u_{kl} u_{mn}$ with the spin–phonon coupling constants D_{ijklmn}. Now the fluctuating effective field takes the form $\Omega_\alpha=\Sigma_{p,p'} W_\alpha^{p,p'} Q_p Q_{p'}$ ($W_\alpha^{p,p'}$ are the matrix elements of $H_{s-ph}^{(2)}$), so the correlation function Fourier image $\gamma_{\mu\nu}(\omega)$ [Eq. (4)] is expressed in terms of phonon correlation functions $\langle(Q_{p_1}Q_{p_2})(\tau)Q_{p_3}Q_{p_4}\rangle_\omega$. Its calculation performed in a harmonic approximation leads to a simple expression $\delta(\omega_{p_1}-\omega_{p_2})(\delta_{p_1,p_3}\delta_{p_4,p_2}+\delta_{p_2,p_3}\delta_{p_4,p_1})(2N_{p_1}N_{p_2}+N_{p_1}+N_{p_2})$. Substituting this function for $\gamma_{\mu\nu}(\omega)$ and a parameter D for the dominant contribution among the coupling constants D_{ijklmn}, the spin phase relaxation rate for the two-phonon process is given at low temperatures ($T<\hbar c_T/k_B a_0$) approximately as

$$T_{2,0}^{-1} = \frac{\mu_B^2 B^2 D^2}{21\rho^2 c_T^3}\left(\frac{k_B T}{\hbar c_T}\right)^7. \tag{20}$$

Parameter D can be estimated as $D=3(g-2)C^2/E_g^2$ (g, C, and E_g are the electron g factor, deformation potential and energy gap).[1] The numerical evaluation of Eq. (20) at low temperatures ($T=4$ K) predicts a long relaxation time. In the case of GaAs at $B=1$ T, one can find $T_{2,0}\approx3\times10^5$ s, which is too long to be of any experimental or practical interest.

III. DISCUSSION

To illustrate the significance of the mechanism under consideration, let us briefly survey the most important spin decoherence mechanisms reported in the literature: the HFI and spin–lattice interactions. In the presence of the HFI, an electron spin performs precession around the sum of the external magnetic field $\vec{B}$ and the effective field $\vec{B}_{hf}$ caused by the HFI. The dispersion of $\vec{B}_{hf}$ over an ensemble of QDs results in a relatively fast electron spin phase diffusion (see Refs. 20 and 21); it causes also a partial longitudinal relaxation ($<67\%$) that can be essentially eliminated as $B\gg B_{hf}$ ($B_{hf}<1$ G for typical Si QDs).

In the case of a single electron in a QD, the electron spin can change its phase through the HFI since the nuclei also perform precession around the effective field caused by the electron spin. This field proportional to $|\psi(\vec{r}_j)|^2$ [see Eq. (15)] is inhomogeneous over the QD volume, which distorts the mutual correlation of nuclei spin configuration and subsequently causes an alteration in the direction and strength of $\vec{B}_{hf}$.[20,22] However, this relaxation is rather long and can be

suppressed if $B \gg B_{hf}$. In addition, it can be further reduced in the case of full nuclei spin polarization[22] and/or isotope purification. Hence, the spin–lattice (i.e., phonon) interaction provides the most fundamental and unavoidable source of electron spin decoherence.

Among the spin–lattice interaction mechanisms, the phonon-mediated transitions between the ground and excited states modulate the precession velocity leading to very effective decoherence,[1] when their energy separations are small enough. However, under the assumption $k_B T \ll \delta_0$ this relaxation is reduced as $\exp(-\delta_0/k_B T)$. Thus, the spin-flip processes and the phonon-decay induced mechanism considered in this paper provide the main contributions at low temperatures. Moreover, these two mechanisms differ in the magnetic field dependence. When the magnetic field decreases, the spin-flip process yields to spin phase diffusion induced by phonon relaxation as mentioned above. The estimated magnetic field strength for this cross-over (e.g., $\lesssim 0.1$ T) is well within the range of practical importance.

IV. CONCLUSION

We considered spin phase diffusion of a localized electron through anharmonic phonon disturbances. In contrast to the spin-flip process where only the resonant (with the Zeeman energy) phonons are relevant, electron spin phase acquires random shifts when relaxation of any (resonant or nonresonant) phonon occurs. A quantitative analysis shows that the considered phase relaxation reveals a relatively weak dependence on the magnetic field strength and the temperature compared to the direct spin relaxation processes or other mechanisms that involve the excited electron states. In addition, a specific dependence on the magnetic field direction [Eq. (13)] is attributed to this mechanism. Thus, one can expect that at low temperatures and magnetic fields the spin phase diffusion mediated by the phonon relaxation can become dominant over the spin-flip processes. As for quantitative estimation of the relaxation rate, the decisive role belongs to the phonon lifetime τ_{ph}. In the present study, we estimated τ_{ph} from the experiments conducted in bulk Si. It is not apparent if this estimation is applicable to the case of QDs. Moreover, the phonon lifetime may be a function of geometry and composition of the structure under consideration. However, the qualitative signatures of the proposed mechanism is expected to persist and may provide a ground for experimental verification. It should also be pointed out that the framework of the developed theoretical model allows more accurate estimation when the detailed information on phonon dispersion and relaxation is taken into account.

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[1] Y.G. Semenov and K.W. Kim, Phys. Rev. Lett. **92**, 026601 (2004).
[2] A.V. Khaetskii and Y.V. Nazarov, Phys. Rev. B **64**, 125316 (2001).
[3] B.A. Glavin and K.W. Kim, Phys. Rev. B **68**, 045308 (2003).
[4] P.G. Klemens, *Solid State Physics* (Academic, New York, 1958), Vol. 7, p. 1.
[5] Y.G. Semenov, Phys. Rev. B **67**, 115319 (2003).
[6] The anisotropy of Eq. (3) as well as the correlation between all spin components of the $\vec{s}$ are the main distinctions between these equations and the well-known Bloch-Redfield equations [see, for instance, K. Blum, *Density Matrix Theory and Applications* (Plenum, New York, 1996)].
[7] C. Tahan, M. Friesen, and R. Joynt, Phys. Rev. B **66**, 035314 (2002).
[8] S.I. Erlingsson, Y.V. Nazarov, and V.I. Fal'ko, Phys. Rev. B **64**, 195306 (2001).
[9] D.N. Zubarev, Usp. Fiz. Nauk **71**, 71 (1960); [Sov. Phys. Usp. **3**, 320 (1960)].
[10] K.N. Pathak, Phys. Rev. **137**, A1569 (1965).
[11] Y.J. Hanand P.G. Klemens, Phys. Rev. B **48**, 6033 (1993).
[12] P. Carruthers, Rev. Mod. Phys. **33**, 92 (1961).
[13] L. Roth, Phys. Rev. **118**, 1534 (1960).
[14] N.G. Koloskova, Fiz. Tverd. Tela (Leningrad) **5**, 61 (1963); [Sov. Phys. Solid State **5**, 40 (1963)].
[15] H. Hasegawa, Phys. Rev. **118**, 1523 (1960).
[16] C.J. Glassbrenner and G.A. Slack, Phys. Rev. **134**, A1058 (1964).
[17] J. Callaway, Phys. Rev. **113**, 1046 (1959).
[18] D.K. Wilson and G. Feher, Phys. Rev. **124**, 1068 (1961).
[19] E.R. Glaser, T. A. Kennedy, B. Molnar, R.S. Sillmon, M.G. Spencer, M. Mizuta, and T.F. Kuech, Phys. Rev. B **43**, 14 540 (1991).
[20] I.A. Merkulov, A.L. Efros, and M. Rosen, Phys. Rev. B **65**, 205309 (2002).
[21] Y.G. Semenov and K.W. Kim, Phys. Rev. B **67**, 073301 (2003).
[22] A.V. Khaetskii, D. Loss, and L. Glazman, Phys. Rev. Lett. **88**, 186802 (2002).