Phonon-induced decay of the electron spin in quantum dots

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We study spin relaxation and decoherence in a GaAs quantum dot due to spin-orbit interaction. We derive an effective Hamiltonian which couples the electron spin to phonons or any other fluctuation of the dot potential. We show that the spin decoherence time $T_2$ is as large as the spin relaxation time $T_1$ under realistic conditions. For the Dresselhaus and Rashba spin-orbit couplings, we find that, in leading order, the effective magnetic field can have only fluctuations transverse to the applied magnetic field. As a result, $T_2 = 2T_1$ for arbitrarily large Zeeman splittings, in contrast to the naively expected case $T_2 \ll T_1$. We show that the spin decay is drastically suppressed for certain magnetic field directions and values of the Rashba coupling constant. Finally, for the spin coupling to acoustic phonons, we show that $T_2 = 2T_1$ for all spin-orbit mechanisms in leading order in the electron-phonon interaction.

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Phase coherence of spin in quantum dots (QDs) is of central importance for spin-based quantum computation in the solid state. Sufficiently long coherence times are needed for implementing quantum algorithms and error correction schemes. If the qubit is operated as a classical bit, its decay time is given by the spin relaxation time $T_1$, under realistic conditions. For the Dresselhaus and Rashba spin-orbit couplings, we find that, in leading order, the effective magnetic field can have only fluctuations transverse to the applied magnetic field. As a result, $T_2 = 2T_1$ for arbitrarily large Zeeman splittings, in contrast to the naively expected case $T_2 \ll T_1$. We show that the spin decay is drastically suppressed for certain magnetic field directions and values of the Rashba coupling constant. Finally, for the spin coupling to acoustic phonons, we show that $T_2 = 2T_1$ for all spin-orbit mechanisms in leading order in the electron-phonon interaction.

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\[ H_d = \frac{p^2}{2m^*} + U(r), \]  
\[ H_{SO} = \beta(p_x\sigma_z + p_y\sigma_y) + \alpha(p_x\sigma_y - p_y\sigma_x), \]  
\[ H_Z = \frac{1}{2} \mu_B \mathbf{B} \cdot \mathbf{\sigma}, \]

where \( p = -i\hbar \nabla + (e/c)\mathbf{A}(r) \) is the electron 2D kinetic momentum, \( U(r) \) is the lateral confining potential, with \( r = (x, y) \), and \( \mathbf{\sigma} \) are the Pauli matrices. The axes \( x \) and \( y \) point along the main crystallographic directions in the (001) plane of GaAs. The spin-orbit coupling is given by the Hamiltonian \( H_{SO} \), where the term proportional to \( \beta \) originates from the bulk Dresselhaus spin-orbit coupling, which is due to the absence of inversion symmetry in the GaAs lattice; the term proportional to \( \alpha \) represents the Rashba spin-orbit coupling, which can be present in quantum wells if the confining potential (in our case along the z-axis) is asymmetric. The magnetic field \( \mathbf{B} = B(\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta) \) defines the spin quantization axis via the Zeeman term \( H_Z \). The phonon potential is given by

\[ U_{ph}(r) = \sum_{q_j} \frac{F(q_j)e^{iq_jr}}{\sqrt{2\rho_c\omega_{q_j}/\hbar}}(e\beta_{q_j} - iq\Xi_{q_j})(b_{-q_j}^\dagger + b_{q_j}) \]

where \( b_{q_j}^\dagger \) creates an acoustic phonon with wave vector \( q = (q_x, q_y, q_z) \), branch index \( j \), and dispersion \( \omega_{q_j} \); \( \rho_c \) is the sample density (volume is set to unity in \( \Xi_{q} \)). The factor \( F(q_j) \) equals unity for \( |q_j| \ll d^{-1} \) and vanishes for \( |q_j| \gg d^{-1} \), where \( d \) is the characteristic size of the quantum well along the z-axis. We take into account both piezo-electric (\( \beta_{q_j} \)) and deformation potential (\( \Xi_{q_j} \)) kinds of electron-phonon interaction. For \( \beta_{q_j} \) and \( \Xi_{q_j} \), we refer the reader to Ref. \[4\]. Next, we derive an effective Hamiltonian, which describes the spin dynamics, decoherence and relaxation at low temperatures \( T < \hbar \omega_0 \).

The electron spin coupled to phonons due to the spin-orbit interaction \[49\]. For typical GaAs QDs the spin-orbit length \( \lambda_{SO} = \hbar/m^*\beta \) is much larger than the electron orbit size \( \lambda \). In 2D, there is no linear in \( \lambda/\lambda_{SO} \) contribution to the spin-phonon coupling at zero Zeeman splitting \[10\], \[11\], \[16\], \[17\]. Here, we consider a finite magnetic field, \( m^* \beta^2 < \mu_B B < \hbar \omega_0 \), for which the linear in \( \lambda/\lambda_{SO} \) contribution to the spin–phonon coupling dominates the spin decay. Using perturbation theory (or Schrieffer-Wolff transformation), we obtain the effective Hamiltonian \[12\]

\[ H_{eff} = \frac{1}{2} \mu_B (\mathbf{B} + \delta \mathbf{B}(t)) \cdot \mathbf{\sigma}, \]

\[ \delta \mathbf{B}(t) = 2\mathbf{B} \times \mathbf{\Omega}(t), \]

where \( \mathbf{\Omega}(t) \) is defined as follows

\[ \mathbf{\Omega}(t) = \langle \psi | \left( \hat{L}_d^{-1} \mathbf{\xi}, U_{ph}(t) \right) | \psi \rangle. \]

Here, \( | \psi \rangle \) is the electron orbital wave function, \( \hat{L}_d \) is the dot Liouvillian, \( \hat{L}_d A = [H_d, A] \). The vector \( \mathbf{\xi} \) has a simple form in the coordinate frame: \( x' = (x+y)/\sqrt{2}, \)

\[ y' = (y-x)/\sqrt{2}, \]

\[ z' = z, \]

namely, \( \mathbf{\xi} = (y'/\lambda_z, x'/\lambda_x, 0) \), where \( 1/\lambda_z = m^*(\beta \pm \alpha)/\hbar \). Eq. \( (7) \) contains one of our main results: in 1st order in spin-orbit interaction, there can be only transverse fluctuations of the effective magnetic field, i.e. \( \delta \mathbf{B}(t) \cdot \mathbf{B} = 0 \). This statement holds true for spin coupling to any fluctuations, be it the noise of a gate voltage or coupling to particle-hole excitations in a Fermi sea. Next, we consider the decay of the electron spin, \( \mathbf{S} = \sigma/2 \), governed by the Hamiltonian \[48\].

The phonons which are emitted or absorbed by the electron leave the dot during a time \( \tau_c \), \( d/s \lesssim \tau_c \lesssim \lambda/s \), where \( s \) is the sound velocity. The electron spin decays over a much longer time span in typical structures, and, therefore, undergoes many uncorrelated scattering events. In this regime, the dynamics and decay of the spin is governed by the Bloch equation \[20\]

\[ \dot{\mathbf{S}} = \omega \times (\mathbf{S}) - \Gamma \mathbf{S} + \mathbf{Y}, \]

where \( \omega = \omega_l \), with \( \omega = \mu_B B/\hbar \) and \( l = \mathbf{B}/B \). For a generic \( \delta \mathbf{B}(t) \), we find that in the Born-Markov approximation \[20\], \[21\] the tensor \( \Gamma_{ij} \) can be written as \( \Gamma = \Gamma^r + \Gamma^d \), with

\[ \Gamma^r_{ij} = \delta_{ij} \left( \delta_{pq} - l_p l_q \right) J_{pq}^r(\omega) - (\delta_{ip} - l_i l_p) J_{pq}^r(\omega) - \delta_{ij} \varepsilon_{kpq} l_q I_{pq}^r(\omega) + \varepsilon_{ipq} l_p I_{pq}^r(\omega), \]

\[ \Gamma^d_{ij} = \delta_{ij} l_p J_{pq}^d(\omega) - l_i l_p J_{pq}^d(\omega), \]

where \( J_{ij}^r(\omega) = \text{Re} \left[ J_{ij}(\omega) \pm J_{ij}(-\omega) \right] \) and \( I_{ij}^d(\omega) = \text{Im} \left[ J_{ij}(\omega) \pm J_{ij}(-\omega) \right] \) are given by the spectral function

\[ \hat{J}_{ij}(\omega) = \frac{g^2 \mu_B^2}{2\hbar^2} \int_0^{\infty} \langle \delta \mathbf{B}(0) \delta \mathbf{B}(t) \rangle e^{-i\omega t} dt. \]

The inhomogeneous part in Eq. \( (9) \) is given by

\[ 2\mathbf{T}_{ij} = l_i J_{ij}(\omega) - l_j J_{ij}(\omega) + \varepsilon_{ijk} k_p I_{pq}^r(\omega) + \varepsilon_{ijk} k_p I_{pq}^d(\omega), \]

where \( \varepsilon_{ijk} \) is the anti-symmetric tensor (with Einstein summation convention) and we have assumed that \( \langle \delta \mathbf{B}(t) \rangle = 0 \). Eq. \( (9) \) describes spin decay in a number of problems, such as electron scattering off impurities in bulk systems, nuclear spin scattering \[20\], etc. In our notation, the spin decay comes from the symmetric part of \( \Gamma \), whereas the anti-symmetric part leads to a correction to \( \omega \) in Eq. \( (9) \). The tensor \( \Gamma^r \) describes spin decay due to processes of energy relaxation, such as, e.g., emission/absorption of a phonon. Therefore, the \( T_1 \) time is entirely determined by \( \Gamma^r \), see below. The tensor \( \Gamma^d \) can be non-zero only due to elastic scattering of spin, i.e. due to dephasing. \( \Gamma^d \) contributes to the decoherence time \( T_2 \), and so does \( \Gamma^r \). In many cases, however, the latter contribution is negligible, and \( \Gamma^d \) entirely dominates the spin decoherence \[20\]. This is in strong contrast to what we
We now consider a harmonic confinement, \( U(r) = m^* \omega_0^2 r^2 / 2 \), and evaluate \( \Omega(t) \) of Eq. 8 for the ground state \( \psi(r) = \exp\left( -r^2 / 2 \lambda^2 \right) / \lambda \sqrt{\pi} \), where \( \lambda = h^{-1} \sqrt{(m^* \omega_0)^2 + (e B_\perp / 2c)^2} \). Using the identity

\[
y = \frac{2}{m^* \omega_0^2} \left( \frac{\partial}{\partial y} + \frac{ie B_z}{\hbar c} x \right) ,
\]

we find an expression for \( \Omega_{\perp} \), which can be obtained from the r.h.s. of Eq. 5 by replacing

\[
\exp(iq \cdot r) \to \frac{2iq_y}{m^* \omega_0^2} \exp\left( -q_y^2 \lambda^2 / 4 \right) .
\]

Furthermore, an expression for \( \Omega_{\parallel} \) is obtained in the same way, using the substitution with a different prefactor, namely with \( (q_y / \lambda -) \to (q_y / \lambda+) \). Finally, for the real part of the correlator \( J_{ij}(w) \), we obtain

\[
\text{Re} J_{XX}(w) = \frac{\omega^2 w^3 (N_w + 1)}{(\Lambda + m^* \omega_0^2)^2} \sum_{\rho \sigma} \frac{\pi h^3}{\rho \sigma} \int_0^{\pi / 2} d \theta \sin^2 \theta \times e^{-\omega \lambda \sin \theta} \theta / 2 w \int_{-\infty}^{\infty} F\left( \frac{|w|}{s_j} \cos \theta \right) \left( e^{\mu_R + \mu_I} \right) (\omega \lambda \sin \theta)^{3/2} (w \lambda)^{3/2},
\]

where \( N_w = (e^{hw/\omega} - 1)^{-1} \), and \( s_j \) is the sound velocity for branch \( j \). For GaAs, we use \( s_1 \approx 4.7 \times 10^5 \text{ cm/s} \) and \( s_2 \approx s_3 \approx 3.37 \times 10^5 \text{ cm/s} \). Furthermore, \( \Xi_j = \delta_{j1} \Xi_0 \) with \( \Xi_0 \approx 7 \text{ eV} \), and \( \Xi_{j1} = 3 \sqrt{2} \pi h_1 \kappa^{-1} \sin^2 \theta \cos \theta, \Xi_{j2} = \sqrt{2} \pi h_1 \kappa^{-1} \sin(3 \cos^2 \theta - 1) \sin \theta, \) with \( h_1 \approx 0.16 \text{ C/m}^2 \) and \( \kappa \approx 13 \). The effective spin-orbit length \( \Lambda_+ \) in Eq. 17 is given by

\[
\frac{2}{\Lambda_+^2} = 1 - \frac{1}{\lambda^2} + \frac{1}{\lambda^2} \pm \left( 1 - \frac{1}{\lambda^2} + \frac{1}{\lambda^2} \right)^2 - 4 \sqrt{\lambda^2 + \lambda^2} .
\]

Re \( J_{YY}(w) \) is obtained from Eq. 17 by substituting \( \Lambda_+ \to \Lambda_- \), and \( J_{ZZ}(w) = 0 \). Im \( J_{XX}(w) \) and Im \( J_{YY}(w) \) are irrelevant for our discussion, see further. From Eq. 10, we obtain \( \Gamma_{XX} = \Gamma_{YY}^{\perp} \), \( \Gamma_{YY} = \Gamma_{XX}^{\perp} \), \( \Gamma_{ZZ} = \Gamma_{XX}^{\perp} + \Gamma_{YY}^{\perp} \), \( \Gamma_{XX}^{\perp} = -\Gamma_{YY}^{\perp} \), \( \Gamma_{YY}^{\perp} = -\Gamma_{XX}^{\perp} \). Since \( \Gamma_j / \omega \sim \omega^2 / \omega_0 \Lambda_+^2 \ll 1 \), we can solve the secular equation iteratively and obtain

\[
\frac{1}{T_1} = \frac{1}{T_2} = \frac{1}{2} (\delta_{pq} - l_p l_q) \Gamma_{pq} = \frac{1}{2} (\Gamma_{XX} + \Gamma_{YY}) .
\]

Then, the solution of Eq. 8 reads \( \langle S_X(t) \rangle = S_L e^{-1/T_2} \sin(\omega t + \phi) \), \( \langle S_Y(t) \rangle = S_L e^{-1/T_2} \cos(\omega t + \phi) \), and \( \langle S_Z(t) \rangle = S_T + (S_{x0}^2 - S_T) e^{-1/T_1} \), with the thermodynamic value of spin being \( S_T = (l_T T_1) T_1 = -(4/2) \tan(h_0 / 2K_B T) \), and the initial value \( \langle S(0) \rangle = (S_{x0} \sin \phi, S_{x0} \cos \phi, S_{y0}^2) \). For our special situation with purely transverse fluctuations \( \Gamma^d = 0 \), we obtain

\[
\frac{1}{T_1} = \frac{2}{T_2} = J_{XX}(\omega) + J_{YY}(\omega) .
\]
Keeping in mind the setup of Ref. 3, we plot the relaxation rate $1/T_1$ as a function of $B$ for $\theta = \pi/2$ and $\alpha = 0$ on Fig. 1 (solid curve). We find that $1/T_1$ has a plateau in a wide range of magnetic fields (cf. Ref. 3), due to a crossover from the piezoelectric-transverse (dashed curve) to the deformation potential (dot-dashed curve) mechanism of electron-phonon interaction. For arbitrary $\varphi$, $\theta$, and $\alpha$, we have $1/T_1 = f/T_1(\theta = 0, \alpha = 0) = 0$ with

$$f = \frac{1}{\beta} \left[ ((\alpha^2 + \beta^2)(1 + \cos^2 \theta) + 2\alpha \beta \sin^2 \theta \sin 2\varphi) \right] \quad (22)$$

The angular dependence of $1/T_1$ for $\alpha = 0$ has been obtained before [10]. Note that $\sqrt{T}$ describes an ellipsoid in the frame $(x', y', z)$, i.e. $f = x'^2 + y'^2 + z'^2$, with dimensionless $x', y', z$ obeying $(x'/\alpha^2) + (y'/\beta^2) + (z/c)^2 = 1$, where $\alpha = 1 + \alpha/\beta$, $b = 1 - \alpha/\beta$, and $c = \sqrt{\alpha^2 + b^2}$. Note that, if $\alpha = \beta$, then $b = 0$, i.e. $1/T_1$ vanishes if the $B$-field is along $y'$. The same is true along the $x'$ axis for $\alpha = -\beta$. This special case ($\alpha = \pm \beta$) has been discussed previously for extended electron states in a two-dimensional electron gas (2DEG) [27]. Note that the Hamiltonian [1] conserves the spin component $\sigma_{y}(x')$ for $\alpha = \beta$ ($\alpha = -\beta$) and $B || y'(x')$. This spin conservation results in $T_1$ being infinite to all orders in the spin-orbit Hamiltonian [3]. At the same time, the decoherence rate $1/T_{2D}$ reduces to the next order contribution of [3]. However, as we show below, a single-phonon process is inefficient in inducing dephasing and, therefore, $1/T_2$ can be non-zero only in the next order in electron-phonon interaction. Next, we note that a long lived spin state also occurs in a different GaAs structure, namely, for a 2DEG grown in the (110) crystallographic direction. Then, the normal to the 2DEG plane component of spin is conserved [26], provided there is no Rashba coupling.

We discuss now other spin-orbit mechanisms. In Eq. 3, we did not include the so-called $k^3$-terms of the Dresselhaus spin-orbit coupling [26], i.e. $H_{SO} \propto \beta d^2 (\sigma_z k_x k_y^2 - \sigma_y k_x k_y^2)$. They are parametrically small ($d^2/\lambda^2 \ll 1$) in the 2D limit, compared to the retained ones. However, their contribution to the spin decay can be important, if $g \lessapprox d^2/\lambda^2$ and $\lambda^2 \ll d\lambda_{SO}$, since the orbital effect of the magnetic field contributes here in the first place. Still, for typical GaAs QDs, these mechanisms are negligible.

Additional spin decay mechanisms arise from the direct spin-phonon interaction [10]. The strain field produced by phonons couples to the electron spin via the spin-orbit interaction, resulting in the term $\Delta H' = (V_0/4)\varepsilon_{ijk}\sigma_i u_{uj} p_k$, where $p_k$ is the bulk momentum, $u_{uj}$ is the phonon strain tensor, and $V_0 = 8 \times 10^7$ cm/s for GaAs. A similar mechanism occurs in a $B$-field, due to $g$-factor fluctuations caused by lattice distortion. This yields $\Delta H'' = \tilde{g}h B \sum_{i \neq j} u_{uj} \sigma_i B_j$, where $\tilde{g} \approx 10$ for GaAs. The contribution of these mechanisms to the spin-flip rates in QDs has been estimated in Ref. 10. Except for the $\alpha = \pm \beta$ cases discussed above, the direct mechanisms are usually negligible in QDs. Here, we find that such spin-phonon couplings do not violate the equality $T_2 = 2T_1$. For this, we note that $1/T_{ij} = 0$ for a generic $\delta B_i = \sum_q M_i(q) b_q + b_q^*$ in Eq. 6, if $q |M(q)|^2 \rightarrow 0$ at $q \rightarrow 0$. Obviously, this condition is satisfied for the direct spin-phonon mechanisms, since the strain tensor $u_{ij}$ vanishes at $q = 0$. The same follows for the Hamiltonian [1] with the phonon potential [3] and an arbitrary $H_{SO}$; the physical explanation being that the potential of long-wave phonons is constant over the dot size and, thus, commutes with $H_{SO}$. Finally, we note that, at temperatures $T \sim \hbar \omega_0$, there can be dephasing mechanisms [28], which can result in $T_2 \ll T_1$.

In conclusion, we have shown that the decoherence time $T_2$ of an electron spin in a GaAs QD is as large as the relaxation time $T_1$, for the spin decay based on spin-orbit mechanisms. We acknowledge support from the Swiss NSF, NCCR Basel, EU RTN ‘Spintronics’, US DARPA, ARO, and ONR.

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We fix the quadrant of $\chi$ by requiring that the sign of $\sin 2\chi$ coincides with the one of the numerator of Eq. (13).

A similar identity for $x$ is obtained from Eq. (15) by replacing $(x, y)$ by $(-y, x)$.

The decay rates are defined by the secular equation:
\[ \det |\Gamma_{ij} - E\delta_{ij} + \varepsilon_{ijk}\omega_k| = 0, \] as the real part of $E$. In the secular approximation, one iterates the secular equation, assuming $\Gamma_{ij}/\omega \ll 1$.

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