Field Dependence of the Electron Spin Relaxation in Quantum Dots

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Interaction of the electron spin with local elastic twists due to transverse phonons has been studied. Universal dependence of the spin relaxation rate on the strength and direction of the magnetic field has been obtained in terms of the electron gyromagnetic tensor and macroscopic elastic constants of the solid. The theory contains no unknown parameters and it can be easily tested in experiment. At high magnetic field it provides parameter-free lower bound on the electron spin relaxation in quantum dots.

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Relaxation of the electron spin in solids is a fundamental problem that has important applications. Among them are electron spin resonance and quantum computing. In a semiconductor quantum dot the relaxation time of the electron spin is determined by its interaction with phonons, nuclear spins, impurities, etc. While impurities and nuclear spins can, in principle, be eliminated, the interaction with phonons cannot. Thus, spin-phonon interactions provide the most fundamental upper bound on the lifetime of electron spin states. The existing methods of computing electron spin-phonon rates in semiconductors rely upon phenomenological models of spin-orbit interaction, see, e.g., Refs. 1, 2, 3, 4, 5, 6, 7, 8, 9, 10. These models contain unknown constants that must be obtained from experiment. Meantime, as has been noticed more than 50 years ago by Elliot (see also Ref. 11), the spin-orbit coupling in semiconductors determines the difference of the electron g-factor from the free electron value of \( g_0 = 2.0023 \). The question then arises whether the effect of the spin-orbit coupling on spin-phonon relaxation can be expressed via the difference between the electron gyromagnetic tensor \( g_{\alpha\beta} \) (\( \alpha, \beta = x, y, z \)) and the vacuum tensor \( g_{0 \alpha \beta} \). Since \( g_{0 \alpha \beta} \) can be measured independently, this would enable one to compare the computed relaxation rates with experiment without any fitting parameters. In this Letter we show that this, indeed, can be done under certain reasonable simplifying assumptions.

Zeeman interaction of the electron with an external magnetic field, \( \mathbf{B} \), is given by the Hamiltonian

\[
\hat{H}_Z = -\mu_B g_{\alpha\beta} \mathbf{s}_\alpha \mathbf{B}_\beta ,
\]

where \( \mu_B \) is the Bohr magneton and \( \mathbf{s} = \mathbf{\sigma}/2 \) is the dimensionless electron spin with \( \mathbf{\sigma}_\alpha \) being Pauli matrices. One can choose the axes of the coordinate system along the principal axes of the tensor \( g_{\alpha\beta} \). Then \( g_{\alpha\beta} \) is diagonal,

\[
g_{\alpha\beta} = g_{\alpha} \delta_{\alpha\beta} ,
\]

represented by three numbers, \( g_x \), \( g_y \), and \( g_z \) that can be directly measured. Perturbation of Eq. (1) by phonons has been studied in the past by writing all terms of the expansion of \( g_{\alpha\beta} \) on the strain tensor, \( u_{\alpha\beta} \), permitted by symmetry. This gives spin-phonon interaction of the form \( A_{\alpha\beta\gamma\delta} u_{\alpha\beta} \mathbf{s}_\gamma \mathbf{B}_\delta \) with unknown coefficients \( A_{\alpha\beta\gamma\delta} \). To avoid this uncertainty we limit our consideration to local rotations generated by transverse phonons. The argument for doing this is three-fold. Firstly, the rate of the transition accompanied by the emission or absorption of a phonon is inversely proportional to the fifth power of the sound velocity. Since the velocity of the transverse sound is always smaller than the velocity of the longitudinal sound, the transverse phonons must dominate the transitions. Secondly, for a dot that is sufficiently rigid to permit only tiny local rotations as a whole under an arbitrary elastic deformation, the emission or absorption of a quantum of the elastic twist will be the only spin-phonon relaxation mode. Finally, we notice that interaction of the electron spin with a local elastic twist generated by a transverse phonon does not contain any unknown constants. Consequently, it gives parameter-free lower bound on the electron spin relaxation rate.

The angle of the local rotation of the crystal lattice in the presence of the deformation, \( \mathbf{u}(r) \), is given by

\[
\delta \phi = \frac{1}{2} \nabla \times \mathbf{u} ,
\]

and the local angular velocity is \( \delta \phi \). The analysis of the effect of the rotation on the electron spin can be done in the coordinate frame that is rigidly coupled to the crystal lattice. In that coordinate frame the effect of the rotation is two-fold. Firstly, it results in the opposite rotation of the external magnetic field felt by the spin. The corresponding perturbation of the magnetic field is given by \( \delta \mathbf{B} = \mathbf{B} \times \delta \phi \). Secondly, the Hamiltonian in the rotating frame acquires a kinematic term \( -\hbar \mathbf{s} \cdot \delta \phi \). The spin-phonon interaction in the lattice frame (marked by prime) is then given by

\[
\hat{H}_{s-ph}' = -\hbar \Omega' \cdot \mathbf{s} , \quad \Omega'_\alpha = \delta \phi'_{\alpha} \pm (\mu_B/\hbar) g_{\alpha\beta} [\mathbf{B} \times \delta \phi]'_{\beta} .
\]
In these formulas \( \delta \phi \) should be understood as an operator. Summation over repeated indices is implied. The canonical quantization of phonons and Eq. (3) yield

\[
\mathbf{u} = \sqrt{\frac{\hbar}{2MN}} \sum_{k\lambda} \mathbf{e}_{k\lambda} \frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{\sqrt{\omega_{k\lambda}}} \left( a_{k\lambda} + a_{-k\lambda}^\dagger \right)
\]

(5)

\[
\delta \phi = \frac{1}{2} \sqrt{\frac{\hbar}{2MN}} \sum_{k\lambda} \left[ i \mathbf{k} \times \mathbf{e}_{k\lambda} \right] \frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{\sqrt{\omega_{k\lambda}}} \left( a_{k\lambda} + a_{-k\lambda}^\dagger \right),
\]

(6)

where \( M \) is the mass of the unit cell, \( N \) is the number of cells in the crystal, \( \mathbf{e}_{k\lambda} \) are unit polarization vectors, \( \lambda = l_1, l_2, l \) denotes polarization, and \( \omega_{k\lambda} = v_{k\lambda} \) is the phonon frequency. The total Hamiltonian in the lattice frame is

\[
\hat{H}' = \hat{H}_0 + \hat{H}'_{s-\text{ph}}, \quad \hat{H}_0 = \hat{H}_Z + \hat{H}_{\text{ph}},
\]

(7)

where \( \hat{H}_Z \) is Zeeman Hamiltonian of Eq. (11) unperturbed by phonons and \( \hat{H}_{\text{ph}} \) is Hamiltonian of free phonons.

Spin-phonon transitions occur between the eigenstates of \( \hat{H}_0 \). These eigenstates are direct products of the spin and phonon states

\[
| \Psi_\pm \rangle = | \psi_\pm \rangle \otimes | \phi_\pm \rangle.
\]

(8)

Here \( | \psi_\pm \rangle \) are the eigenstates of \( \hat{H}_Z \) with energies \( E_\pm \) and \( | \phi_\pm \rangle \) are the eigenstates of \( \hat{H}_{\text{ph}} \) with energies \( E_{\text{ph},\pm} \), satisfying

\[
E_+ + E_{\text{ph},+} = E_- + E_{\text{ph},-}.
\]

(9)

For \( \hat{H}'_{s-\text{ph}} \) of Eq. (11), which is linear in phonon amplitudes, the states \( | \phi_\pm \rangle \) differ by one emitted or absorbed phonon with a wave vector \( \mathbf{k} \). We will use the following designations

\[
| \phi_+ \rangle \equiv | n_{\mathbf{k}} \rangle, \quad | \phi_- \rangle \equiv | n_{\mathbf{k}} + 1 \rangle.
\]

(10)

We need to compute the matrix element corresponding to the decay of the spin \( | \Psi_+ \rangle \rightarrow | \Psi_- \rangle \). With the help of Eq. (11) we get:

\[
\langle \Psi_- | \hat{H}'_{s-\text{ph}} | \Psi_+ \rangle = \mathbf{K} \cdot \langle \phi_- | \delta \phi | \phi_+ \rangle,
\]

(11)

where components of vector \( \mathbf{K} \) are given by

\[
K_\gamma \equiv -\mu_B (g_\alpha - g_\beta) B_\beta \epsilon_{\alpha\beta\gamma} \langle \psi_- | s_\alpha | \psi_+ \rangle,
\]

(12)

and the principal components of the gyromagnetic tensor, \( g_\alpha \), are defined by Eq. (2). To obtain Eq. (12), we have used the relation

\[
\delta \phi = i \frac{\hbar}{\epsilon} [\hat{H}_{\text{ph}}, \delta \phi],
\]

(13)

to eliminate \( \delta \phi \) from Eq. (11), and the energy conservation, Eq. (12). Note that for the isotropic \( g \)-factor \( K_\gamma = 0 \) and thus phonons do not couple to the spin.

As an independent test, one can consider the problem in the laboratory frame. In the presence of the local rotation given by Eq. (3), the gyromagnetic tensor in the laboratory frame becomes

\[
\delta g_{\alpha\beta} = \delta_{\alpha\beta} - \epsilon_{\alpha\beta\gamma} \delta \phi_\gamma.
\]

(15)

Substituting Eq. (14) into Eq. (11) and using the orthogonality of the rotation matrix, \( \mathbb{R}_{\alpha\beta} = R_{\alpha\beta}^{-1} \), we get for the Zeeman Hamiltonian in the presence of phonons

\[
\hat{H}_Z^{(\text{ph})} = -\mu_B g_{\alpha'\beta'} (\mathbb{R}_{\alpha'\alpha}^{-1} s_\alpha) (\mathbb{R}_{\beta'\beta}^{-1} B_\beta).
\]

(16)

In the linear order in \( \delta \phi \) with the help of Eq. (15) one obtains the full Hamiltonian in the laboratory frame

\[
\hat{H} = \hat{H}_0 + \hat{H}_{s-\text{ph}}, \quad \hat{H}_0 = \hat{H}_Z + \hat{H}_{\text{ph}},
\]

(17)

where

\[
\hat{H}_{s-\text{ph}} = -\hbar \Omega \cdot \mathbf{s},
\]

(18)

and

\[
\Omega_\alpha = (\mu_B/\hbar) (g_\alpha - g_\beta) B_\beta \epsilon_{\alpha\beta\gamma} \delta \phi_\gamma.
\]

(19)

One can see that the spin-phonon matrix element in the laboratory frame, \( \langle \Psi_- | \hat{H}_{s-\text{ph}} | \Psi_+ \rangle \), is the same as that in the lattice frame, Eqs. (11) and (12).

To obtain the relaxation rate one can use the Fermi golden rule. With the help of Eq. (16) the transition matrix element of Eq. (11) can be expressed as

\[
\langle \Psi_- | \hat{H}_{s-\text{ph}} | \Psi_+ \rangle = \frac{\hbar}{N} \sum_{k\lambda} V_{k\lambda} \langle n_{k'} \mid a_{k\lambda} + a_{-k\lambda}^\dagger \mid n_{k'} \rangle,
\]

(20)

where

\[
V_{k\lambda} \equiv \frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{\sqrt{8M/\hbar\omega_{k\lambda}}} | \mathbf{k} \times \mathbf{e}_{k\lambda} |.
\]

(21)

Note that only the transverse phonons contribute to the relaxation process. The decay rate \( W_{+\to} \) of the upper spin state into the lower state, accompanied by the emission of a phonon, and the rate \( W_{-\to} \) of the inverse process are given by

\[
\begin{pmatrix}
W_{+\to} \\
W_{-\to}
\end{pmatrix} = W_0 \begin{pmatrix} n_{\omega_0} + 1 \\ n_{\omega_0} \end{pmatrix},
\]

(22)

where \( n_{\omega_0} = (e^{\hbar\omega_0/(k_B T)} - 1)^{-1} \) is the phonon occupation number at equilibrium,

\[
\hbar\omega_0 \equiv E_+ - E_- = \mu_B \left( \sum_{\gamma} g_\gamma^2 B_\gamma^2 \right)^{1/2}.
\]

(23)
is the distance between the two spin levels, and

\[ W_0 = \frac{1}{N} \sum_{k \lambda} |V_{k \lambda}|^2 2\pi \delta(\omega_{k \lambda} - \omega_0). \quad (24) \]

The balance equation for normalized populations of the upper spin state \( n_+ \) and the lower spin state \( n_- \) (satisfying \( n_+ + n_- = 1 \)) is

\[ \dot{n}_+ = -W_+ n_+ + W_- n_- = -\Gamma n_+ + W_+ \quad (25) \]

where the relaxation rate is given by

\[ \Gamma = W_+ + W_- = W_0 \coth \left( \frac{\hbar \omega_0}{2k_B T} \right). \quad (26) \]

Using Eq. 21 and replacing \((1/N) \sum_{k} \cdots \) by \(v_0 \int d^3 k/(2\pi)^3 \ldots \) in Eq. 21, \(v_0\) being unit-cell volume, one obtains

\[ W_0 = \frac{1}{12\pi^2 Mv_0^2 \omega_D^3} = \frac{1}{12\pi^2} \frac{\hbar^2}{\rho v_0^5}. \quad (27) \]

Here \( v_0 \) is the velocity of the transverse sound, \( \rho \) is the mass density, \( \omega_D = v_0 \sqrt{\frac{\gamma}{k_B T}} \) is the Debye frequency for the transverse phonons, and \(|\mathbf{K}|^2 = \sum K_x^2 K_y^2 K_z^2 \). With the help of Eq. 12 we get

\[ |\mathbf{K}|^2 = \mu_B^2 \sum_{\alpha \beta} (g_\alpha - g_\beta)^2 (B_\alpha^2 T_{\alpha \alpha} + B_\beta B_\alpha T_{\alpha \beta}), \quad (28) \]

where

\[ T_{\alpha \beta} = \frac{1}{2} \left( \langle \psi_- | s_\alpha | \psi_+ \rangle^* \langle \psi_- | s_\beta | \psi_+ \rangle + \text{c. c.} \right). \quad (29) \]

The eigenstates of \( \hat{H}_Z \) are

\[ \psi_+ = -\sin(\theta/2)e^{-i\varphi/2}|+ \rangle + \cos(\theta/2)e^{i\varphi/2}|- \rangle, \]

\[ \psi_- = \cos(\theta/2)e^{i\varphi/2}|+ \rangle + \sin(\theta/2)e^{-i\varphi/2}|- \rangle, \quad (30) \]

where \(|+ \rangle, |- \rangle\) are the eigenstates of the operator \( s_z \),

\[ s_z |\pm \rangle = \pm \frac{1}{2} |\pm \rangle, \quad (31) \]

and the spherical angles \( \theta \) and \( \varphi \) are defined through

\[ b = g_x H_x e_x + g_y H_y e_y + g_z H_z e_z = |b| (\sin \theta \cos \varphi e_x + \sin \theta \sin \varphi e_y + \cos \theta e_z). \quad (32) \]

This gives

\[ \langle \psi_- | s_x | \psi_+ \rangle = \frac{1}{2} (i \sin \varphi + \cos \theta \cos \varphi) \]

\[ \langle \psi_- | s_y | \psi_+ \rangle = \frac{1}{2} (-i \cos \varphi + \cos \theta \sin \varphi) \]

\[ \langle \psi_- | s_z | \psi_+ \rangle = -\frac{1}{2} \sin \theta. \quad (33) \]

Direct calculation then yields

\[ T_{\alpha \beta} = \frac{1}{4} \left( \delta_{\alpha \beta} - \frac{g_\alpha B_\alpha g_\beta B_\beta}{\sum_{\gamma} (g_\gamma B_\gamma)^2} \right). \quad (34) \]

Thus one obtains

\[ |\mathbf{K}|^2 = \frac{\mu_B^2}{8} \sum_{\alpha \beta \gamma} (g_\alpha - g_\beta)^2 \left( B_\alpha^2 + B_\beta^2 - \frac{(g_\alpha + g_\beta)^2 B_\alpha^2 B_\beta^2}{\sum_{\gamma} (g_\gamma B_\gamma)^2} \right). \quad (35) \]

With the help of Eq. 23, Eq. 24 now can be written in the final form

\[ \Gamma = \frac{1}{3\pi\hbar} \left( \frac{\mu_B B}{Mv_0^2 \omega_D} \right)^2 F_T(n) = \frac{\hbar}{3\pi \rho} \left( \frac{\mu_B B}{\hbar v_0} \right)^5 F_T(n), \quad (36) \]

where \( n = B/B \) and

\[ F_T(n) = \frac{1}{32} \left( \sum g_\gamma^2 n_\gamma^2 \right)^{3/2} \coth \left[ \frac{\mu_B B}{2k_B T} \left( \sum g_\gamma^2 n_\gamma^2 \right)^{1/2} \right] \]

\[ \times \sum_{\alpha \beta} (g_\alpha - g_\beta)^2 \left[ n_\alpha^2 + n_\beta^2 - \frac{(g_\alpha + g_\beta)^2 n_\alpha^2 n_\beta^2}{\sum_{\gamma} (g_\gamma n_\gamma)^2} \right]. \quad (37) \]

Here \( \alpha, \beta, \gamma \) run over \( x, y, z \). It is apparent from Eq. 47 that the relaxation mechanism studied in this Letter requires anisotropy of the gyromagnetic tensor. When the field is directed along the z-axis, Eq. 37 simplifies to

\[ F_T(e_z) = \frac{g^2}{16} (g_z - g_x)^2 + (g_z - g_y)^2 \coth \left( \frac{g_z \mu_B B}{2k_B T} \right). \quad (38) \]

For the theory to be valid, \( \omega_0 \) of Eq. 28 should not exceed \( \omega_D \), otherwise there will be no acoustic phonons responsible for the discussed spin-phonon relaxation mechanism. If \( g_\alpha \) are of order unity, this is equivalent to the condition that the factor \( (\mu_B B/\hbar v_0) \) in Eq. 46, that has dimensionality of the wave vector, is less than the Debye wave vector, \( k_D = \omega_D/v_0 \), for transverse phonons. This condition is almost always satisfied in the experimentally accessible field range. At \( \hbar \omega_0 \gg k_B T \) the coth factor in equations 37 and 38 tends to one. In this case \( \Gamma \propto B^5 \) while \( F_T \) depends only on the direction of the field with respect to the principal axes of \( g_{\alpha \beta} \). In the opposite limit of \( k_D T \gg \hbar \omega_0 \), the relaxation rate is proportional to \( B^4 T \) while its dependence on the direction of the field is given by the factor

\[ \frac{1}{\sum_{\alpha \beta} (g_\alpha - g_\beta)^2 \left[ n_\alpha^2 + n_\beta^2 - \frac{(g_\alpha + g_\beta)^2 n_\alpha^2 n_\beta^2}{\sum_{\gamma} (g_\gamma n_\gamma)^2} \right]} \quad (39) \]
A nice property of the spin relaxation mechanism studied above is its universal dependence on the strength and the direction of the magnetic field. Due to $B^5$ in Eq. (36) this mechanism can dominate electron spin relaxation at high fields. For, e.g., $\rho \sim 5 \text{g/cm}^3$ and $v_t \sim 2 \times 10^5 \text{cm/s}$, it gives $\Gamma \sim 3 \times 10^4 \text{s}^{-1} F_T(\mathbf{n})$. The dependence of the rate on the direction of the field, $F_T(\mathbf{n})$, is entirely determined by the difference between principal values of the tensor $g_{\alpha\beta}$. Highly anisotropic $g_{\alpha\beta}$ has been theoretically predicted in two-dimensional systems [15] and experimentally detected in GaAs quantum wells [16]. In the mK temperature range, the spin relaxation times of order $100 \mu$s have been observed [17] in GaAs electron quantum dots in the field of order 10T. Note that equations (36) - (39) allow a detailed comparison between theory and experiment for the proposed mechanism of relaxation, which, thus, can be easily confirmed or ruled out for a particular quantum dot.

In conclusion, we have studied electron spin relaxation in quantum dots due to local rotations generated by transverse phonons. This is an unavoidable relaxation channel that occurs when the electron gyromagnetic tensor, $g_{\alpha\beta}$, is anisotropic. It can dominate spin relaxation at high magnetic fields. The advantage of our theory is that it expresses the effect of unknown spin-orbit interactions on electron spin relaxation in terms of the tensor $g_{\alpha\beta}$ alone. The corresponding relaxation rate has universal dependence on the strength and direction of the field with respect to the principal axes of $g_{\alpha\beta}$. The important feature of the proposed mechanism is that it does not involve any unknown constants of the quantum dot and is entirely determined by the three principal values of $g_{\alpha\beta}$, which can be independently measured. This allows simple experimental test of the proposed theory.

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