Spin relaxation in quantum dots

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Results are given for spin relaxation in quantum dots due to acoustic phonon-assisted flips of single spins at low temperatures. The dominant spin relaxation processes for varying dot size, temperature, and magnetic field are identified. These processes are mediated by the spin-orbit interaction and are described within a generalized effective mass treatment. Particular attention is given to phonon coupling due to interface motion, which dominates the relaxation for dots with diameters ≤ 15 nm, and also to a direct spin-phonon process that arises from valence-conduction band coupling and dominates the rates for increasing temperature. Low-temperature relaxation rates are found to be small and to depend strongly on size, on temperature, and on magnetic field. Results are illustrated with evaluations for GaAs/Al_xGa_{1-x}As systems, and a minimum in the relaxation rate is found for dot diameters ~ 20 nm.

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Spins in quantum dots (QD's) are of considerable current interest in connection with the search for solid state implementations for "qu-bits" in quantum information technologies.¹ A key challenge is obtaining long spin coherence times so that quantum information can be stored and manipulated without losses. A few experimental results have been reported² that suggest long coherence times in QD's. To date, however, it has been difficult to obtain quantitative experimental results and to interpret them. Thus it is important to have theoretical guidance for the coherence times and their dependences on system and on parameters.

In bulk semiconductors spin relaxation is quite fast, typically on the order of 10^{-11} s (Ref. 3). Bulk relaxation arises from spin-orbit coupling mediated by phonon or impurity scattering.³ Relaxation in QD's is different because the electronic states are quantized, which makes it more difficult to satisfy energy conservation in spin-flip events. Phonons typically are needed⁴ for energy conservation in QD's, and thus the coherence rates are considerably smaller.

Here we give a full treatment of the relaxation of single spins in QD's due to acoustic phonons for dots up to \sim 70 nm, of interest in a wide range of optical experiments. Results for the overall magnitude and for dependence on magnetic field, temperature, and size are given. One-phonon processes give the zero temperature contributions, and two-phonon processes give contributions that increase with temperature. Several of these contributions have been obtained earlier and evaluated for large, flat QD's.^{8,9} We show that a novel spin-phonon coupling due to interface motion dominates the relaxation for smaller dots and that a new spin-phonon interaction, which arises from coupling between valence and conduction bands, dominates for increasing temperature.

For magnetic fields up to ~ 7 T, the Zeeman spin splittings in the QD's of interest are typically ~ 0.01 -0.1 meV, which are much smaller than the separations between confined electronic states. Thus the spin relaxation rates between the two Zeeman levels are greater than those between confined states, and we consider only the former. For illustration we give results for GaAs QD's with several shapes in Al_{0.3}Ga_{0.7}As. Other materials and geometries differ mainly in quantitative details. Spin-orbit interaction in semiconductors is essential in spin relaxation processes because phonons alone do not flip spins. The effective mass Hamiltonian for a carrier in a magnetic field in a bulk III-V semiconductor with T_d symmetry, such as GaAs, is obtained from the standard Kane Hamiltonian³

$$H = \frac{1}{2} \mathbf{P} \left[\frac{1}{m(\mathbf{r})} \mathbf{P} \right] + V(\mathbf{r}) + \mu g(\mathbf{r}) \sigma_z B + \{ \gamma_c(\mathbf{r}), \sigma \cdot \mathbf{\Omega} \},$$
(1)

where $\mathbf{P} = \mathbf{p} + e\mathbf{A}/c$, $\Omega = [P_x(P_y^2 - P_z^2), P_y(P_z^2 - P_x^2), P_z(P_x^2 - P_y^2)]$, with **A** the vector potential, and $\{A,B\} = (AB + BA)/2$. The first term is the kinetic energy, the second the confining potential, the third the Zeeman interaction. The last term is the Dresselhaus spin-orbit interaction, which arises from the absence of the inversion symmetry. $\gamma_c = 2\hbar^3 \Delta/\sqrt{2mE_g}E_gm_{cv}$, where Δ , m, E_g , and m_{cv} are the energy of the spin-orbit split-off valence band, the electron mass, the band gap energy, and the Kane parameter for the coupling to higher conduction bands.³ For QD's of one material imbedded in another the parameters have spatial dependence.

Spin relaxation at low T: For T less than a few degrees Kelvin spin relaxation rates are given by one-phonon emission processes accompanied by spin flips arising from spinorbit coupling for the dot sizes and fields of interest here.¹⁰ The one-phonon spin flip rates are given by the Fermi Golden Rule.

Acoustic phonons dominate these processes at low temperatures. The two materials in QD systems of interest typically have similar acoustic properties, and thus we will use bulk phonons to a good approximation. Long wavelength phonons are involved for small Zeeman energies, and thus rates for piezoelectric coupling are greater than those from deformation potential.¹¹ The most important contribution from the bulklike region of the QD comes from the piezoelectric interaction between electrons and acoustic phonons. The coupling between acoustic piezophonons and carrier charges is L. M. WOODS, T. L. REINECKE, AND Y. LYANDA-GELLER

$$U_{\mathbf{Q}}^{piezo} = h_{14}\xi_i\xi_j u_{\mathbf{Q},l}\rho_{\mathbf{Q}}, \qquad (2)$$

where $h_{14}=1.2\times10^9$ Ev/m is the piezotensor, $\xi_{i,j}=Q_{i,j}/Q$, $u_{\mathbf{Q},l}$ is the l-th component of the phonon operator, and $\rho_{\mathbf{Q}}$ is the electron density.

In addition, there is a novel coupling between spins and phonons arising from interface motion. It is analogous to that between carrier charges and phonons, sometimes called "ripple coupling."^{12,13} It arises when interface motion due to acoustic phonons changes a parameter of the system, here $\Lambda_i = 1/m, V, g$, and γ_e in Eq. (1), in a region. The coupling between charge carriers and phonons is

$$U_{\mathbf{Q},\Lambda_i} = \nabla \Lambda_i(\mathbf{r}) \cdot \mathbf{u}_{\mathbf{Q}},\tag{3}$$

where $\mathbf{u}_{\mathbf{Q}}$ is the phonon operator, $\nabla \Lambda_i(\mathbf{r}) = \Delta \Lambda_i \int_S dA \, \delta(\mathbf{r} - \mathbf{R}) \hat{\mathbf{n}}$ and $\Delta \Lambda_i = \Lambda_{i1} - \Lambda_{i2}$ for parameters in materials 1 and 2. The outward interface of the surface *S* has a normal vector $\hat{\mathbf{n}}$ at point **R**.

The matrix elements for spin-flip transitions between the Zeeman levels of the lowest QD electronic state, $\psi_{0\uparrow}$ and $\psi_{0\downarrow}$, due to the coupling to bulk piezophonons and also those due to the interface motion through variation in the parameters 1/m, V,g, and γ_e are

$$M_{\mathbf{Q},piezo} = \int d^{3} \langle \psi_{0,\downarrow} | U_{\mathbf{Q}}^{piezo} | \psi_{0,\uparrow} \rangle, \qquad (4)$$

$$M_{\mathbf{Q},\Lambda_{i}} = \int d^{3} \langle \psi_{0,\downarrow} | H_{\mathbf{Q},\Lambda_{i}} | \psi_{0,\uparrow} \rangle, \qquad (5)$$

where $H_{\mathbf{Q},V} = U_{\mathbf{Q},V}$, $H_{\mathbf{Q},g} = \mu B U_{\mathbf{Q},g}$, $H_{\mathbf{Q},m} = \frac{1}{2} \mathbf{P}(U_{\mathbf{Q},m} \mathbf{P})$, and $H_{\mathbf{Q},\gamma_c} = U_{\mathbf{Q},\gamma_c} \sigma \cdot \Omega$. The spin-orbit coupling in the last term in Eq. (1) provides the needed mixing of the spins in the wave functions. Such an admixture is not required for $M_{\mathbf{Q},\gamma_c}$, and it provides a direct spin-phonon coupling. From perturbation theory

$$|\psi_{0,\uparrow(\downarrow)}(\mathbf{r})\rangle = |u_{0,\uparrow(\downarrow)}(\mathbf{r})\rangle + \alpha \times |u_{1,\downarrow(\uparrow)}(\mathbf{r})\rangle, \qquad (6)$$

where $u_{0,1}$ are the unperturbed wave functions for the ground and first excited electronic states with energies $E_{0,1}$, $\alpha = \langle u_{1,\downarrow(\uparrow)}(\mathbf{r}) | H_{so} | u_{0,\uparrow(\downarrow)}(\mathbf{r}) \rangle / (E_1 - E_0 \pm \mu g B)$, and H_{so} is the Dresselhaus spin-orbit term. Higher lying QD states give smaller contributions and are not included.

The boundary conditions on the QD electronic functions are taken to require that the wave function and its flux $1/m(\mathbf{r}) \partial \psi / \partial n$ be continuous across the interface.¹⁴ For simplicity we choose separable wave functions. We consider dots in the shape of parallelepiped and cylinder. For a parallelepiped, the ground state in the *x* direction is $u_0 \sim \cos(k_0 x)$ for $x \leq a$ and $u_0 \sim \cos(k_0 a) e^{-\kappa_0 (|\mathbf{x}| - a)}$ for $x \geq a$, and similarly for the other directions. Here $k_0 = \sqrt{2m_1 E_0 / \hbar^2}$ and κ_0 $= \sqrt{2m_2 (\Delta E_c - E_0) / \hbar^2}$, where ΔE_c is the band offset. Two types of parallelepipeds are considered, one where the lateral extent is 2a and the height is a/2, and the second where the height is a constant 5 nm. For the cylinder $u_0 \sim J_0(k_0 \rho)$ for $\rho < R$ and $u_0 \sim J_0(k_0 R) e^{-\kappa_0(\rho - R)}$ for $\rho > R$, and in *z*-direction the function is the same as for parallelepiped.

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Then from Eqs. (4) and (6), the T=0 relaxation rates due to piezophonons for the cylindrical (cyl) and parallelepiped (para) QD's are

$$\Gamma_{piezo}^{cyl} = \frac{2\pi}{\hbar\rho s^2} \left(\gamma_c h_{14} \frac{\mu g B}{\Delta E^2} \frac{1}{R^2} \right)^2 \left(\frac{\mu g B}{\hbar s} \right)^3 K_D, \qquad (7)$$

$$\Gamma_{piezo}^{para} = \frac{2\pi}{\hbar\rho s^2} \left[\gamma_c h_{14} \frac{\mu g B}{\Delta E^2} \left(\frac{1}{a^2} - \frac{1}{c^2} \right) \right]^2 \left(\frac{\mu g B}{\hbar s} \right)^3 J_D \quad (8)$$

where $\Delta E = E_0 - E_1$, and $K_D = 0.91 \times 10^3$ and $J_D = 0.65 \times 10^2$.

Using Eq. (5) in Eq. (6), we obtain for the parallelepiped the rates involving the ripple coupling due to changes in the parameters:

$$\Gamma_{V} = \frac{2\pi}{\hbar\rho s^{2}} \left[\gamma_{c} \frac{\mu g B}{\Delta E^{2}} \frac{\Delta V}{a} \left(\frac{1}{a^{2}} - \frac{1}{c^{2}} \right) \right]^{2} \left(\frac{\mu g B}{\hbar s} \right)^{3} J_{1}, \quad (9)$$

$$\Gamma_{g} = \frac{2\pi}{\hbar\rho s^{2}} \left[\gamma_{c} \frac{\mu \Delta g B}{\Delta E} \frac{1}{a} \left(\frac{1}{a^{2}} - \frac{1}{c^{2}} \right) \right]^{2} \left(\frac{\mu g B}{\hbar s} \right)^{3} J_{2}, \quad (10)$$

$$\Gamma_m = \frac{2\pi}{\hbar\rho s^2} \left[\gamma_c \frac{\mu g B}{\Delta E^2} \frac{\hbar^2 \Delta m}{2m_1^2 a^2} \frac{1}{a} \left(\frac{1}{a^2} - \frac{1}{c^2} \right) \right]^2 \left(\frac{\mu g B}{\hbar s} \right)^3 J_3, \tag{11}$$

$$\Gamma_{\gamma_c} = \frac{2\pi}{\hbar\rho s^2} \left[\Delta \gamma_c \frac{1}{a^2} \left(\frac{1}{a^2} - \frac{1}{c^2} \right) \right]^2 \frac{\mu g B}{\hbar s} J_4, \qquad (12)$$

where $J_1 = 1.32 \times 10^4$, $J_2 = 5.76 \times 10^5$, $J_3 = 6.868 \times 10^5$, and $J_4 = 1.43 \times 10^4$. For nonzero T, the rates in Eqs. (7)–(12) are multiplied by (1 + N) for phonon emission and absorption where $N = 1/(e^{\mu g B/k_B T} - 1)$, which gives additional contributions linear in T at small T. Results for cylindrical dots have similar dependences on size and magnetic field and are not given explicitly here.

For a spherical QD the mixing of spin states into the first and second excited electronic functions in Eq. (8) is zero from the symmetry. Then the leading contribution comes from the third excited electronic state, and the relaxation rate is approximately 50 times smaller than those for less symmetrical shapes given here.

Results for the spin relaxation rates at B=1 T are given in Fig. 1 for cylindrical and flat parallelepiped dots of GaAs in Al_{0.3}Ga_{0.7}As (Ref. 15). The size dependences are obtained from $\Delta E \sim a^{-2}$ for all geometries. Then the rates from the piezophonons are $\Gamma_{piezo} \sim a^4$. The rates from the ripple mechanism, $\Gamma_{g,m} \sim a^{-2}$, and $\Gamma_{\gamma_c} \sim a^{-8}$, increase dramatically for decreasing size, and Γ_V is independent of size explicitly. The rates for parallelepiped dots of lateral size *a* and height a/2 are the similar to those for cylinders in Fig. 1(a). The ripple coupling at the surface becomes more important as size decreases. The total scattering rate has a minimum for dot diameters in the range of 10–30 nm depending on geometry. The *B* dependences are strong, with that $\Gamma_{piezo} \sim B^5$ and



FIG. 1. Scattering rates due to one-phonon processes at B = 1 T and T=0 K as functions of size for (a) cylinder with diameter d, (b) parallelepiped with height c=5 nm. The term Γ_V gives a contribution independent of size, and it is included in Γ_{tot} .

 $\Gamma_{V,g,m} \sim B^5$ and $\Gamma_{\gamma_c} \sim B$. Processes similar to those in Eqs. (7) and (8) control the rates for larger sizes and were studied earlier⁸ for large, flat QD's.

Spin relaxation for increasing T: For temperatures larger than a few degrees Kelvin,¹⁰ two-phonon processes begin to make the dominant contributions to spin relaxation in QD's. They sum over higher effective densities of phonons than do the one-phonon processes. A phonon is scattered from \mathbf{Q} to \mathbf{Q}' accompanied by a spin flip. Transition rates are calculated from the Fermi Golden Rule.

We have examined all two-phonon contributions, and we find that the dominant contribution comes from a spin-phonon interaction in which phonons modulate the carrier spin-orbit interaction. This interaction can be thought of as spin-orbit coupling arising from the potential caused by phonon scattering and is present in systems both with and without inversion symmetry. The form of the interaction is obtained from perturbation theory by including the mixing of the upper valence and the lowest conduction bands,³

$$H = \alpha \sigma \cdot (\mathbf{p} \times \nabla U_{\mathbf{0}}) \tag{13}$$

where $\alpha = \hbar^2 \eta (1 - 1/2 \eta)/3mE_g(1 - 1/3 \eta)$, and $\eta = \Delta/(\Delta + E_g)$ with E_g the gap energy and Δ the spin-orbit split-off band gap. U_Q is a potential that scatters the electron. For the QD's studied here $U_{\mathbf{Q}}$ comes either (i) from the piezoelectric interaction in Eq. (2) or (ii) from the ripple coupling in Eq. (3). In the case of ripple coupling, the difference in mass gives the largest contribution to Eq. (13), $H = -i\alpha\hbar^2\nabla \cdot \mathbf{u}_Q/2m_1^2\sigma \cdot [\mathbf{p} \times \mathbf{Q}]\nabla \cdot \nabla$. Formally, the interaction in Eq. (13) is similar to the "Elliott-Yafet mechanism"





FIG. 2. Representation of the amplitude for two-phonon spinflip processes.

in bulk semiconductors³ or "skew scattering" in bulk metals.¹⁶ In QD's, however, the electrons are in confined states, and the scattering involves inelastic phonon scattering.

Equation (13) gives nonzero contributions only to transitions between different electronic states. The form for the amplitude of these processes is shown in Fig. 2 where V_1 gives a spin flip process and V_2 a process without a spin flip. We find that the two largest contributions come from (i) a process in which V_1 is from Eq. (13) using the piezophonon coupling for $\nabla U_{\mathbf{Q}}$ and V_2 is from the piezophonon coupling in Eq. (2) (*piezo*) and (ii) a process in which V_1 is from Eq. (13) using ripple coupling for $\nabla U_{\mathbf{Q}}$ and V_2 is from ripple coupling from Eq. (3) (m). For the parallelepiped the scattering rates are

$$\Gamma_{piezo}^{2-ph} = \frac{2\pi}{\rho^2 s^3} \left[\frac{\alpha}{\Delta E} a^2 h_{14}^2 \right]^2 \left(\frac{k_B T}{\hbar s} \right)^{11} I, \tag{14}$$

$$\Gamma_m^{2-ph} = \frac{2\pi}{\rho^2 s^3} \left\{ \frac{\alpha}{\Delta E} \frac{1}{a^2} \left[\frac{\hbar^2 \Delta m}{2m_1^2} \left(\frac{2}{a^2} + \frac{1}{c^2} \right) \right]^2 \right\}^2 \left(\frac{k_B T}{\hbar s} \right)^7 \tilde{I},$$
(15)

where I = 1.7 and $\tilde{I} = 0.54 \times 10^7$. Similar results are obtained for the other dot shapes.

These two contributions to the rates are shown in Fig. 3 for T=4 K. They increase rapidly with temperature, going as $\Gamma_{piezo} \sim T^{11}$ and $\Gamma_m \sim T^7$. Processes from ripple coupling



FIG. 3. Two-phonon scattering rates for a parallelepiped with lateral size d and height c = d/4 at T = 4 K and B = 1 T.

dominate at small size, and those from piezophonons dominate at large size. The total size dependence at any *T* is given by the sum of the one-phonon processes and the two-phonon processes. From Figs. 1 and 3, we see that for the parallelepiped for temperatures increasing from zero up to a few degrees, the minimum moves from ~ 20 nm diameter to smaller sizes. The two-phonon contributions do not depend on B and thus will be present even for B=0.

A two-phonon process was suggested earlier by Khaetskii and Nazarov⁸ in which the spin flip transition V_1 in Fig. 2 was given by the piezoelectric coupling in Eq. (2) mediated by the band mixing from the Dresselhaus term, and the transition V_2 was given by the piezoelectric coupling in Eq. (3). This contribution is given by Γ_{KN} in Fig. 3, and for most temperatures it is orders of magnitude smaller than those discussed here.

Here we have given a complete treatment of spin relaxation due to spin-flip scattering in quantum dots. We have introduced a novel process due to interface motion that determines the rates for small dots and we have given a new coupling mechanism that determines the rates for increasing temperatures. Our intent has been to establish the phononassisted mechanisms for low-temperature spin relaxation in quantum dots and their dependences size, magnetic field, and temperature.

These rates are relatively low at small *T*, which is encouraging in the search for qu-bits using quantum dots. The rates,

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- ⁴Processes involving electronic spin flips accompanied by nuclear spins flips⁵ or involving phonon-assisted nuclear spin flips^{5,6} also have been suggested. For a single dot in B, the difficulty of satisfying energy conservation with the widely disparate electronic and nuclear Zeeman energies and the suppression of dipole-dipole interactions among nuclei by the field cause the rate of electronic spin decay into the nuclear system to be slow even in the presence of spin-orbit interaction.⁷ Estimates of the spin relaxation times from phonon-assisted nuclear spin relaxation give⁶ longer spin relaxation times than those obtained here.
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however, increase strongly with *B* and *T*. They also depend on material. For example, the one-phonon rates $\Gamma_{piezo,V} \sim m^3$, $\Gamma_{g,m} \sim m$, $\Gamma_{\gamma_c} \sim m^{-1}$ where *m* is the carrier mass. The two-phonon rate Γ_{piezo}^{2-ph} does not depend on mass explicitly, and Γ_m^{2-ph} goes as m^{-4} . Thus for many II-VI QD's, with their heavier conduction masses, the mass dependent rates will be orders of magnitude larger than those to GaAs. For the same reason, the rates for holes also would be higher. We find that the relaxation rates depend only weakly on dot shape.

The rates are strong functions of size, and they exhibit minima for diameters ~20 nm for the GaAs/Al_xGa_{1-x}As systems at B=1 T and low T. Similar results are obtained for quantum dots from related III-V materials. We suggest that III-V self-organized QD's¹⁷ grown by MBE, which typically have diameters of ~15–25 nm, are near the size for minimum relaxation. On the other hand, QD's formed from monolayer fluctuations in MBE growth of quantum wells¹⁸ have varying lateral sizes on the order of 40 nm and shapes similar to that in Fig. 1(c). We suggest that their spin relaxation times should be shorter than those for self-organized dots.

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- ¹⁴In general, the spin-orbit coupling in Eq. (1) will produce an additional spin-dependent term in the Hamiltonian at the boundary from boundary conditions [P. Pfeffer and W. Zawadzki, Phys. Rev. B **52**, R14 332 (1995).] This results in an additional mixing between spin states, for which phonon matrix elements are $\sim \Delta \gamma$. For the systems here, however, $\Delta \gamma$ is about an order smaller than γ_{12} , and therefore the standard boundary conditions are used here.
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