Oscillatory spin relaxation rates in quantum dots

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Phonon-induced spin relaxation rates in quantum dots are studied as function of in-plane and perpendicular magnetic fields, temperature and electric field, for different dot sizes. We consider Rashba and Dresselhaus spin-orbit mixing in different materials, and show how Zeeman sublevels can relax via piezoelectric and deformation potential coupling to acoustic phonons. We find that strong lateral and vertical confinements may induce minima in the rates at particular values of the magnetic field, where spin relaxation times can reach even seconds. We obtain good agreement with experimental findings in GaAs quantum dots.

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Since the proposal of a qubit based on the electron spin of quantum dots (QDs), much work has been done to understand the processes that may cause their relaxation, since long coherence times are required. One of those processes is related to the phonon-induced spin-flip rates of Zeeman sublevels in QDs in magnetic fields, where the spin purity of the levels is broken by the spin-orbit (SO) interaction. A recent experiment has shown that spin relaxation time has a lower bound of 50 µs at an in-plane field of 7.5 T in a GaAs QD defined in a 2DEG, while theoretical work has given 5 µs at the same QD parameters. In general, SO effects have been considered via perturbation theory, although exact treatments have also been presented. The perturbative approach, which includes only a few states, has been called into question when the QD vertical width is narrow, as the complex interplay between the physical scales playing a role in the system: Zeeman and SO energies, as well as QD lateral and vertical sizes.

The QD is defined by an in-plane parabolic confinement, \( V(r) = m\omega_0^2r^2/2 \), where \( \omega_0 = E_0/h \) is the electronic effective mass (confinement frequency); the QD lateral length is \( l_0 = \sqrt{\hbar/(m\omega_0)} \). The vertical confinement \( V(z) \) is strong enough so that only the state in the first quantum well subband is relevant, and its function is \( \phi_z(z) = \sqrt{2z_0} \sin(\pi z/z_0) \) if a hard wall is assumed, \( z_0 \) being the QD vertical well thickness. In a magnetic field \( B \), the unperturbed Hamiltonian, \( H_0 = \hbar^2k^2/2m + V(r) + H_z \), has the well-known Fock-Darwin (FD) solution \( |\psi_{E, \ell} \rangle \), where \( H_{BIA} = g_0\mu_B B \cdot \sigma/2 \) is the Zeeman term. We include all SO terms in 2D zincblende QDs, namely Rashba and Dresselhaus interactions. The former is due to surface inversion asymmetry (SIA) induced by the 2D confinement, while the latter is caused by the bulk inversion asymmetry (BIA) present in zincblende structures. The SIA Hamiltonian is \( H_{SIA} = \alpha \sigma \cdot \nabla V(r, z) \cdot k \), while the BIA is \( H_{BIA} = \gamma(\sigma_xk_x(k_z^2 - k_y^2) + \sigma_yk_y(k_z^2 - k_x^2) + \sigma_zk_z(k_x^2 - k_y^2)) \), with coupling constants \( \alpha \) and \( \gamma \). The z-confinement yields the electric field \( dV/dz \) in the SIA Hamiltonian as well as the momentum average \( \langle k_z^2 \rangle = (\pi/\zeta_0)^2 \) in the BIA terms. The full QD Hamiltonian is then \( H = H_0 + H_{SIA} + H_{BIA} \), which is diagonalized in a basis containing 110 FD states. Details about the derivation of terms in \( H \), as well as their selection rules for (anti)crossings in the QD energy spectrum, are found elsewhere.

We calculate spin relaxation rates between the two lowest QD Zeeman sublevels caused by piezoelectric and deformation acoustic phonons via Fermi’s Golden Rule: \( \Gamma_{fi} = 2\pi/\hbar \sum_j Q_j |\gamma_{fi}(q)|^2 |Z(q_j)|^2 |M_j(Q)|^2 (n_Q + 1)\delta(\Delta E + \hbar c j Q) \), where the sum is over the emitted phonon modes \( j \) (\( j = LA, TA1, TA2 \)) with momentum \( Q = (q, q_z) \). The term \( Z(q_z) = \langle \phi_z | e^{iq_z} | \phi_z \rangle \) \( \langle \gamma_{fi}(q) | f | e^{iQ} | i \rangle \) is the form factor perpendicular (parallel) to the 2D-plane (position is \( R = (r, z) \)), while \( n_Q \) is the phonon distribution with energy \( \hbar c j Q \); energies
$\Delta E = \varepsilon f - \varepsilon_i$ and states $|i\rangle$, $|f\rangle$ are obtained via diagonalization of the total $H$, so that the SO mixing is fully taken into account. The element $M_j^z(Q) = \Lambda_j(Q) + i \Xi_j(Q)$ includes both piezoelectric $\Lambda_j$ and deformation $\Xi_j$ potentials; in zincblende structures and in cylindrical coordinates, they become $\Xi_{LA}(Q) = \Xi_0 A_{LA} \sqrt{Q}$ (only $LA$ is present for $\Xi_j$), $\Lambda_{LA}(Q) = 3 \Lambda_0 A_{LA} \sin(2\theta) q_z^2 / 2 Q^{7/2}$, $\Lambda_{T A 1}(Q) = \Lambda_0 A_{T A 1} \cos(2\theta) q_z^2 / Q^{7/2}$, and $\Lambda_{T A 2}(Q) = \Lambda_0 A_{T A 2} \sin(2\theta) (2 q_z^2 - 1) q_z^2 / 2 Q^{7/2}$ (both $T A 1$ and $T A 2$ modes are compacted as a single $TA$ mode for $\Lambda_j$), where $A_j = \sqrt{h(2 N_0 \varepsilon_{Cj})^{-1}}$ and $\Lambda_0 = 4 \pi \hbar c_{114} / \kappa$. The bulk phonon constants are $\Xi_0$ and $\hbar c_{14}$, $c_j$ are the sound velocities ($c_{T A 1} = c_{T A 2} = c_{T A} \neq c_{LA}$), $\kappa$ is the dielectric constant, and $N_0$ is the electron density.

The triple space integration $\langle \nu, \phi, z | \rangle$ yields an analytical solution and we were able to do two $(|\phi_q, q_z|)$ out of the momentum integration, leaving a numerical integral only in $q$. For a later use, the only $z_0$-dependence in this remaining integral in $\Gamma_f$, reads $F_j(z_0) = (d_j z_0 - (d_j z_0)^3 / \pi^2)^2 - 2 \sin^2(d_j z_0)$, where $d_j = \sqrt{(\Delta E / \hbar c_j)^2 - q_z^2 / 2}$; $q$ runs from 0 to $\Delta E / \hbar c_j$, while $F_j(z_0)$ is multiplied by polynomials and exponentials in $q$ in the total $\Gamma_f$. No approximation is needed in our derivation of $\Gamma_f$, so that the 3D nature of the phonon is taken into account and the full form factor $e^{Q \mathbf{R}}$ is used.

Panel A of Fig. 1 shows the phonon-induced spin-flip rates as a function of in-plane field, $B_{||}$, for different values of $E_0$ (solid lines) for GaAs QDs. Larger $E_0$ (smaller QDs) present smaller spin-flip rates, i.e., longer relaxation times. This happens because as $E_0$ increases, the orbital levels become more separated and then the SO coupling becomes relatively less important. A strong dependence of the rates with $B_{||}$ and $E_0$ is clear. For example, at $B_{||} = 2$ T, one gets $2.5 \times 10^2$ s$^{-1}$ at $E_0 = 0.7$ meV and $2.5 \times 10^{-1}$ s$^{-1}$ at $E_0 = 5.0$ meV; at $B_{||} = 15$ T the rate goes from $5.2 \times 10^6$ s$^{-1}$ to $6.9 \times 10^4$ s$^{-1}$ for those same values of $E_0$. Finite temperature (dashed-dotted line for $T = 12$ K) enhances the rates by about one order of magnitude due to the enhancement of the phonon population. Larger wells (dotted line for $z_0 = 100$ Å) have rates about one order of magnitude smaller because the BIA term ($\langle k_z^2 \rangle \approx 1 / 3 z_0^2$) decreases. The same $T$ and $z_0$ behavior is found for any $E_0$. Larger electric fields (dashed line for $dV / dz = -38$ meV/Å) have little effect on the rates for large $E_0$, but at smaller $E_0$ the rates decrease by one order of magnitude; this means that by increasing the SIA term so that it equals the BIA coupling, the rates can decrease even though the spin mixing gets stronger. For GaAs QDs under in-plane fields, we find that the TA piezoelectric coupling dominates at low fields ($\leq 14$ T for $E_0 = 5$ meV), while at high fields the deformation potential takes over (symbols in Fig. 1A).

Panels B and C of Fig. 1 respectively show the spin expectation value $\langle \sigma_z \rangle$ of the Zeeman sublevels and their energy splitting $\Delta E$. Smaller $E_0$ values show higher SO-induced spin mixing, so that $\langle \sigma_z \rangle$ deviates from the pure +1 (−1) value for the ground (first excited) state, and $\Delta E$ deviates from the pure $g_0 \mu B B$ value of a QD without SO coupling. Notice at the 3.5 meV QD how a larger well (stronger field) decreases (increases) the SO-induced spin mixing by reducing (enhancing) the importance of the BIA (SIA) coupling. For comparison, the experimental lower bound for the spin-flip time is 50 μs at $B_{||} = 7.5$ T for a 1.1 meV QD while from panel A we find 5 μs. As discussed above, this time is increased by one order of magnitude in a larger $z$-well, which appears to be a better match for the experimental conditions. A 5 μs value has also been found from a perturbative formulation and our results also agree with a different model for the vertical confinement. One can then affirm that the perturbative approach is indeed enough for GaAs in in-plane fields at any QD sizes, and that the form of the $z$-confinement has little effect on the rates.

In Fig. 2 we do the same analysis for GaAs QDs in a perpendicular field, $B_{\perp}$. The most striking feature is the appearance of minima in the rates shown in panel A. The $E_0$-dependent minima ($12 \leq B_{\perp} \leq 16$ T) are due to the vanishing $\Delta E$, as shown in panel C. At such values of $B_{\perp}$, a level crossing of Zeeman sublevels results in a sudden spin-flip, as verified by the vertical lines of panel B for the 0.7 and 3.5 meV QDs. To understand the minima at low fields (6 ≤ $B_{\perp}$ ≤ 8 T) we have to be mindful of the sine argument in $F_j(z_0)$, which may induce a minimum in the rate at a particular value of the field, due to the interplay of energy and length scales in the problem. This behavior does not produce minima for in-plane field on GaAs QDs (Fig. 1). We find that those three low-field minima in Fig. 2A occur at $B_{\perp}$ values where a magnetic field-induced cancellation of the SO influence is produced on the respective $\Delta E$ values, as indicated by the arrows in panel C: below (above) such $E_0$-dependent values of $B_{\perp}$, where $\Delta E$ recovers its Zeeman value of $g_0 \mu B B$ (SO coupling increases (decreases) the Zeeman splitting as compared to the QD without SO).
Notice that the two smallest confinements do not show the low-field rate minima in Fig. 2A. This is due to the lowest sublevels acquiring the same spin (between 3 and 7 T at \( E_0 = 0.7 \) meV in panel B), so that the rate decreases monotonically at that field-range. These features have strong influence on QD effective \( g \)-factors. Like in the in-plane field problem, higher temperatures (\( T = 12 \) K) enhance the rates by one order of magnitude, and the dominating phonon mechanism (symbols) is the TA piezoelectric coupling, but now at any field. To confirm the intricate interplay between all QD energy scales, a larger well (\( z_0 = 100 \) Å) or a stronger field (\( dV/dz = -38 \) meV/Å) removes the rate minima, as shown in panel A for the 3.5 meV QD. Our results agree with available calculations at small fields (\( B_\perp \leq 1 \) T), so that we can confirm that the perturbative approach is not adequate when dealing with SO effects in QDs in a perpendicular field, even if the material is GaAs, if the vertical confinement is strong enough. From Figs. 1 and 2 the rates at small fields (\( \lesssim 2 \) T) have the same behavior under \( B_\parallel \) and \( B_\perp \), namely, the smaller \( E_0 \) the larger the rate; however, they are much larger under \( B_\perp \). The rate maximum (at 2.5 T for the 1.1 meV QD) is due to an anticrossing involving the first and second QD excited states.

Figure 3 shows results for InSb QDs in in-plane fields. In contrast to GaAs (Fig. 1), minima in the rates are visible under \( B_\parallel \) for the highest \( E_0 \) values. As shown for the 20 meV QD, the LA deformation potential is the dominant phonon mechanism for InSb at any field (and \( E_0 \)); this seems to be the case for any narrow-gap material. It is worth mentioning that, contrary to GaAs in Fig. 2A where a unique low-field minimum is present for a given \( E_0 \), the number of rate minima in InSb increases with \( E_0 \). Also, notice that both LA modes have the same number of minima at the same \( B_\parallel \) values (dashed arrows in panel A), while a larger number of minima in the TA mode occurs at different fields; this happens since \( c_{LA} > c_{TA} \), so that the sine argument in \( F_j (z_0) \) is smaller in the LA mode. Panel B shows that Zeeman sublevels have normal spin character, where ground (excited) state is predominantly spin-up (spin-down) at low fields. Finite temperature (\( T = 12 \) K) has no appreciable effect on the rates, a reflection of the much larger energy scales. The energy scale interplay results at higher field (\( dV/dz = -2 \) meV/Å) in slightly smaller (larger) \( \Delta E \) (rates), and rate minimum shifts to higher fields. On the other hand, a larger well (\( z_0 = 100 \) Å) produces a higher \( \Delta E \), so that the spin-flip rate is suppressed by orders of magnitude and its number of minima increases; notice in panel B that the two lowest levels acquire the same spin at \( \approx 8 \) T for such \( z_0 \), so that the oscillatory rate is well defined until that field. Previous calculations in InSb did not include the deformation potential, and yielded unrealistically low rates.

For InSb QDs in perpendicular fields, we see from panel B in Fig. 2 that all spin-flip rates in panel A are meaningful only for \( B_\perp < 8 \) T, a field where both Zeeman sublevels acquire the same spin. This happens earlier for small \( E_0 \) as shown by the 3.0 meV QD, whose rates are well defined only for \( B_\perp < 2 \) T; the vertical line around 1 T in panel B for this \( E_0 \) indicates a spin-flip, accompanied by the vanishing \( \Delta E \) in panel C, the rate minimum in panel A, and a sign change of the QD \( g \)-factor. Larger values of \( E_0 \) present normal spin behavior. Spin-flip rates in panel A do not show the monotonic behavior seen for small fields in Fig. 3. Like in the \( B_\parallel \) case, a finite temperature (\( T = 12 \) K) does not show visible influence on the rates under \( B_\perp \), and the deformation potential dominates at any field. A higher electric field (\( dV/dz = -2 \) meV/Å) slightly increases the rate; a larger well (\( z_0 = 100 \) Å) introduces an oscillatory rate order of magnitude smaller. We emphasize that the perturbative approach finds no use in InSb QDs because of the inherent higher SO coupling. It is worth mentioning that oscillatory rates have also been found in GaAs QDs under \( B_\perp \) by considering momentum relaxation from an excited orbital level to the ground state as well as in coupled GaAs QDs.

Even though both QD materials show oscillatory spin-flip rates – with \( B_\perp \) for GaAs and with both \( B_\parallel \) and \( B_\perp \) for InSb – their origin is slightly different. Minima come from the nature of the \( z \)-confinement, and the field where they occur depend on the lateral size \( l_0 \). The SO coupling mixes spins and alters splitting of sublevels in distinct ways according to field-direction and QD material, so that spin relaxation can be induced by piezoelectric (wide-gap) and deformation (narrow-gap) phonons. Our calculations reveal the rich interplay between all relevant energy scales in QDs. The external field opens channels (at the rate minima) where long spin relaxation times (\( \approx 1 \) s) may be reached, so that the spin coherence required for quantum computing could be improved.

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15. In GaAs: $m = 0.067 m_0$, $g_0 = -0.44$, $\kappa = 12.4$, $\alpha = 4.4$ Å, $\gamma = 26$ eV Å$^3$, $\Xi_0 = 7$ eV, $ch_{14} = 0.140$ eV/Å, $N_0 = 5.32 \times 10^{-27}$ Kg/Å$^3$, $c_{LA} = 4.73 \times 10^{13}$ Å/s, and $c_{TA} = 3.35 \times 10^{13}$ Å/s; also, $g_0 = 0$, $dV/dz = -0.5$ meV/Å, and $T = 0$ K if no other numbers are specified.

16. $\langle k_z^2 \rangle = (\pi/z_0)^2$ for an infinite well. If a Gaussian well is considered, one has $\langle k_z^2 \rangle = 1/z_0^2$, which yields a BIA term a factor of 10 smaller than used in this work.

17. In InSb: $m = 0.014 m_0$, $g_0 = -51$, $\kappa = 16.5$, $\alpha = 500$ Å$^2$, $\gamma = 160$ eV Å$^3$, $\Xi_0 = 7$ eV, $ch_{14} = 0.061$ eV/Å, $N_0 = 5.77 \times 10^{-27}$ Kg/Å$^3$, $c_{LA} = 3.40 \times 10^{13}$ Å/s, and $c_{TA} = 2.29 \times 10^{13}$ Å/s; also, $g_0 = 40$ Å, $dV/dz = -0.5$ meV/Å, and $T = 0$ K if no other numbers are specified.

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