Relaxation of hole spins in quantum dots via two-phonon processes

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We investigate theoretically spin relaxation in heavy hole quantum dots in low external magnetic fields. We demonstrate that two-phonon processes and spin-orbit interaction are experimentally relevant and provide an explanation for the recently observed saturation of the spin relaxation rate in heavy hole quantum dots with vanishing magnetic fields. We propose further experiments to identify the relevant spin relaxation mechanisms in low magnetic fields.

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In the last decade, remarkable progress has been made in the manipulation and control of the spin of electrons confined in semiconductor nanostructures such as quantum dots (QD) [1]. These achievements pave the way toward quantum spin electronics and may lead to spin-based quantum computing [2]. In the past years, a new candidate for a qubit state has been attracting growing interest: the spin of a heavy hole (HH) confined in a flat QD. In a bulk semiconductor the HH \((J_z = \pm 3/2)\) and light hole (LH) \((J_z = \pm 1/2)\) bands are degenerate giving rise to strong mixing and thus to strong HH-spin relaxation. However, in a 2D system the HH and LH bands are split due to the strong confinement along the growth direction [3] implying a significant reduction of the HH spin relaxation via HH-LH mixing.

The relaxation \((T_1)\) and decoherence \((T_2)\) times of a HH-spin localized in a QD are, like for electrons, determined by the hole spin interacts with: the nuclear spin bath in the QD and the lattice vibrations (phonons). The former interaction is weaker for HHs than for electrons (due to the p-symmetry of the hole) [4, 5]. More importantly, it is of Ising type, making it ineffective for HH-spins initialized along the growth direction [4], as typically done in experiments [6]. This is in contrast to electrons, where the hyperfine interaction is isotropic and dominates the spin dynamics at low B-fields [7, 8, 9, 10].

The other relevant source of relaxation are phonons which couple to the hole spin through the spin-orbit interaction (SOI) [11]. The predicted values [11] for the one-phonon induced relaxation time \(T_1\) agree quite well with data obtained in high B-fields [12]. However, for low B-fields \((B \sim 1.5–3T)\) and high temperatures \((T > 2K)\), a clear deviation from the one-phonon theory has been observed [12]. Furthermore, recent experiments on optical pumping of HH-spins in QDs showed saturation of \(T_1\) for very low or even vanishing B-field [6]. The relaxation time was found to be unusually long, \(T_1 \approx 0.1 – 1\) ms, like previously observed in high B-fields [12]. Both observations suggest other sources of relaxation, and the question arises what are they and what are their observable consequences? The answer to this question is not only interesting by itself but also relevant for using HHs as qubits. In the following, we show that two-phonon processes are good candidates and even provide a quantitative explanation of the mentioned measurements at low B-fields [3, 12]. Indeed, as we will see, these processes show weak or no B-field dependence, whereas one-phonon relaxation rates vanish quickly with \(B \to 0\).

\[H_h = H_0 + H_Z + H_{SO} + H_{h-ph} + H_{ph},\]

(1)

where \(H_0 = p^2/2m^* + V(r)\), is the dot Hamiltonian, \(V(r) \equiv m^*\omega_0^2r^2/2\) is the confinement potential which is assumed to be harmonic, with \(m^*\) being the HH mass.

The second term in Eq. (1) is the Zeeman energy of the HH (pseudo-) spin \(H_Z = gj\mu_B B \cdot \sigma/2\), with \(B\) being the magnetic field and \(\sigma\) the Pauli matrices for the HH spin defined in the \(J_z = \pm 3/2\) subspace. The third term represents the spin-orbit Hamiltonian, which, for well separated HH-LH bands (flat dots), reads [11]

\[H_{SO} = \beta p_x + p_y - \sigma_+ + h.c.\]

(2)

This Hamiltonian represents the effective Dresselhaus SOI (restricted to the HH subspace) due to bulk inversion asymmetry of the crystal [11], where \(p_\pm = p_x \pm ip_y\), \(p = -i\hbar \nabla - eA(r)\), \(A(r) = (y, x, 0)B/2\), and \(\sigma_\pm = \sigma_x \pm i\sigma_y\). We note that in Eq. (2) we have neglected the Rashba SOI and other possibly linear-in-\(k\) but small SOI terms [11]. The fourth term in Eq. (1) represents the interaction of the HH charge with the phonon field, i.e.

\[H_{h-ph} = \sum_{qj} M_{qj} X_{qj}\]

with

\[M_{qj} = \frac{F(q_j)e^{i\mathbf{q} \cdot \mathbf{r}}}{\sqrt{2\hbar \omega_{qj}}} [\epsilon\mathbf{\beta}_{qj} - \mathbf{\left(\Xi_0 \mathbf{q} \cdot \mathbf{d}_{qj} - \Xi_z q_z \mathbf{d}_{qj}^2\right)}],\]

(3)

and

\[X_{qj} = \sqrt{\hbar/\omega_{qj}}(a_{-qj}^\dagger + a_{qj}),\]

where \(\mathbf{q}\) is the phonon wave-vector, with \(j\) denoting the acoustic branch, \(\omega_{qj} = \gamma_1 |q|/\hbar\).
$c_j q$ the phonon energy, with $c_j$ the speed of sound in the $j$-th branch, $d_{ij}$ the polarization unit vector, $\rho_s$ the sample density (per unit volume), and $\varepsilon_{ij}$ the piezoelectric electron-phonon coupling and $\Xi_{i,j}$ the deformation potential constants \cite{11}. The form factor $F(q_i)$ in Eq. \ref{eq:4} equals unity for $|q_z| \ll d^{-1}$ and zero for $|q_z| \gg d^{-1}$, with $d$ being the dot size in the (transverse) $z$-direction. The last term in Eq. \ref{eq:1} describes the free phonon bath.

In the following, we elaborate the effect of the phonons on the HH spin, both in the low and high temperature regimes. The phonons do not couple directly to the spin, but the SOI plays the role of the mediator of an effective spin-phonon interaction. Under the realistic assumption that the level splitting in the dot is much larger than the HH-phonon interaction, we can treat $H_{h_{-}ph}$ in perturbation theory.

Let us define the dot Hamiltonian $H_d \equiv H_0 + H_Z + H_{SO}$ and denote by $|n\sigma\rangle$ the eigenstates of $H_0$ (where $n$ labels the orbital and $\sigma$ the spin states). The product states $|n\sigma\rangle$ are then the eigenstates of $H_d$ in the absence of SOI (i.e., for $H_{SO} = 0$). These states are formally connected to $|n\sigma\rangle$ by an exact Schrieffer-Wolff (SW) transformation \cite{10,17}, i.e., $|n\sigma\rangle = e^{S} \langle n|\sigma\rangle$, where $S = -S^i$ is the SW generator and can be found in perturbation theory in SOI. After this transformation, any operator $A$ in the old basis transforms as $A \rightarrow \tilde{A} = e^{-S} A e^{S}$ in the new basis (e.g., $H_d \rightarrow \tilde{H}_d$, $H_{h_{-}ph} \rightarrow \tilde{H}_{h_{-}ph}$, etc.).

Let us now derive the effective spin-phonon interaction under the above assumptions. To do so, we perform another SW transformation of the total HH Hamiltonian $H_b$ to obtain an effective Hamiltonian $H_{eff} = e^\beta H_b e^{-\beta}$, where $\beta = -1/T$ is chosen such that it diagonalizes the HH-phonon Hamiltonian $H_{h_{-}ph}$ in the eigenbasis of $H_d$. In lowest order in $H_{h_{-}ph}$, we obtain $T \approx T_1^{-1} \tilde{H}_{h_{-}ph}$, where the Liouvillean is defined as $\tilde{L}_A = [H_d, A]$, $\forall A$, and diagonal terms of $H_{h_{-}ph}$ are to be excluded. In 2nd order in $H_{h_{-}ph}$, we obtain then the effective spin-phonon Hamiltonian

$$
H_{s_{-}ph} = \sigma \cdot \sum_{q_j, q_{j'}} [\delta_{q_j, q_{j'}} C^{(1)}_{q} X_{q_j} + C^{(2)}_{q, q_{j}} X_{q_j} X_{q_{j'}} + C^{(3)}_{q, q_{j}, q_{j'}} (P_{q_j} X_{q_{j'}} - P_{q_{j'}} X_{q_j})],
$$

with $\sigma \cdot C^{(1)}_{q} = (\tilde{L}_d^{-1} M_{q_j})|0\rangle$, $\sigma \cdot C^{(2)}_{q, q_{j}} = (i\sqrt{h \omega_{q}})(a_{q_j} - a_{q_{j}'})$ is the phonon field momentum operator, and $|0\rangle$ is the orbital ground state. In Eq. \ref{eq:4} we have neglected spin-orbit corrections to the energy levels, being 2nd order in SOI. Note that for vanishing magnetic field $B \rightarrow 0$ the only term which survives in $H_{s_{-}ph}$ is the last one since this is the only one which preserves time-reversal invariance and thus gives rise to zero field relaxation (ZFR) \cite{13,14,15}. Quite remarkably, this term is 1st order in SOI, whereas for electrons it is only 2nd order\cite{13}. This is one of the main reasons why eventually two-phonon processes are much more effective for HHs than for electrons.

We now assume the orbital confinement energy $h \omega_0$ much larger than the SOI, i.e., $||H_0|| \gg ||H_{SO}||$, so that we can treat the SOI in perturbation theory. We consider also the $B$-field to be applied perpendicularly to the dot plane (as in Refs. \cite{8,12}). We limit our description to first order effects in SOI. The SW-generator $S$ can be written as $S = S_+ \sigma_+ - h.c.$, and we then find

$$
S_+ = A_1 p_+ p_+ - p_+ (p_+ - p_+ p_+) + A_2 p_+ p_+ - p_+ (p_+ - p_+ p_+) p_+ + A_3 (p_+ p_+ - p_+ p_+) + A_4 (p_+ - p_+ p_+). \tag{5}
$$

Here, $A_\pm \equiv A_{\mp}(\omega_Z, \omega_c)$ with $\omega_Z = g B / \hbar$ and $\omega_c = e B / 2 c$. For $\omega_Z, \omega_c \ll \omega_0$, we obtain $A_1 \approx -(7/9)(\omega_Z + \omega_c) / \omega_0^5$, $A_2 \approx -(\omega_c / \omega_0^3)$, $A_3 \approx -(2/3)(\omega_c / \omega_0^3)$, and $A_4 \approx (2/3)(\omega_c / \omega_0^3)$, while $p_\pm = p_\pm \pm i p_0$ with $p_{q_j} = -i \hbar \nabla_{q_j} \pm (m \omega_0^2 / \omega_c) q_j(x)$. After somewhat tedious calculations, we obtain analytic expressions for $C^{(i)} = (C^{(i,x)} + C^{(i,y)}, 0)$ occurring in Eq. \ref{eq:4}. We give below only the exact expression for $i = 3$, the rest being too lengthy to be displayed here:

$$
C^{(3,x/y)}_{q, q_{j}, q_{j'}} = \pm \frac{M_{q_j} M_{q_{j'}}}{3 \lambda_3 h \omega_0^5} F(b \cdot b') \times \left( b_{x}^2 b_{z} - b_{y}^2 b_{x} \pm (b_{x} - b_{z}) (2 b_{y} b_{y} + 3 b_{y} b_{z}) \right), \tag{6}
$$

where

$$
F(b \cdot b') = \frac{1}{(b \cdot b')^2} \left( e^{-b \cdot b'/2} - b \cdot b'/2 \right) \times \left( \gamma + \log(b \cdot b'/2) + \Gamma(0, b \cdot b'/2) \right). \tag{7}
$$

Here, $M_{q_j} M_{q_{j'}} = (F(q_j) F(q_{j'}) / 2 \rho_s \sqrt{\Xi_{q, q_j} - \Xi_{q, q_{j'}}}) (\Xi_{q_j, q_{j'}} - \Xi_{q_j, q_{j'}}) d_{q_j} d_{q_{j'}}$ and $b = q d$ is the dot-diameter. We have also introduced $\gamma \approx 2.17$ the Euler constant and $\Gamma(s, x)$ the incomplete gamma function. We note that $C^{(1,2)} \propto B$, so that these two terms vanish with vanishing $B$-field.

Let us now analyze the relaxation of the spin induced by all the phonon processes in the spin-phonon Hamiltonian in Eq. \ref{eq:4}. We first mention that all terms in Eq. \ref{eq:4} can be cast in a general spin-boson type of Hamiltonian $H_{p_{-}ph} = (1/2) g B \sqrt{B^3} \langle t | \sigma, p = 1, 2, 3, \text{the corresponding identification of the fluctuating magnetic field terms } \delta B_j(t) \text{ from Eq. } \ref{eq:4} \text{ (e.g. } \delta B^3(t) \sim C_{q_j}^{(1)} X_{q_j}). \text{ Within the Bloch-Redfield approach, the relaxation rate } \Gamma \equiv 1 / T_1 \text{ can be expressed as } \Gamma = \sum_{x, y, z} J_{ij}(Z E / h) + J_{ij}(-E Z / h). \text{ The correlation functions } J_{ij} \text{ are defined by } J_{ij}(\omega) = \langle \gamma_{ij, q_j} / 2 \rangle \int_0^\infty dt e^{-i \omega t} < \delta B_j(t) \delta B_j(t) >, \text{ where } < \ldots \ldots > \text{ denotes the average over the phonon bath, assumed to be in thermal equilibrium at temperature } T. \text{ The relaxation time associated with the three types of spin-phonon processes in Eq. } \ref{eq:4} \text{ is }
where two-phonon rates, \(\Gamma = \Gamma_A\) \([19, 20]\) and GaAs QDs (labeled by B) come from the effective spin-phonon coupling itself.

Note that for two-phonon processes the single phonon-energies do not need to match the Zeeman energy separately (as opposed to one-phonon processes), so that there is only a weak dependence on the B-field left which comes from the effective spin-phonon coupling itself.

In Figs. 1 and 2 we plot the phonon spin-relaxation rate \(\Gamma\) as a function of the B-field and of temperature, resp., for InAs and GaAs quantum dots. Fig. 1 shows a clear saturation of \(\Gamma\) at low magnetic fields which is due to two-phonon processes, while Fig. 2 shows the known saturation at low temperatures due to one-phonon processes \([11]\).

For these plots, we used the following HH InAs QDs (labeled by A) \([11, 19, 20]\) and GaAs QDs (labeled by B) parameters \([11]\): \(\Delta_z = 1.9 eV, \Delta_x = 2.7 eV, c^A_t = 2.64 \cdot 10^3 m/s (c^B_t = 3.35 \cdot 10^3 m/s), c^A_i = 3.83 \cdot 10^3 m/s (c^B_i = 4.73 \cdot 10^3 m/s), \rho^A_c = 5.68 \cdot 10^3 kg/m^3 (\rho^B_c = 5.3 \cdot 10^3 kg/m^3), m^A_x = 0.25 m_e (m^B_x = 0.14 m_e), g^A = 1.4 (g^B = 2.5)\), and we assume \(\lambda_t = 3 nm (\hbar \omega_{0_B} = 35 meV, \hbar \omega_{0_B} = 60 meV)\) and \(d = 3 nm\) (dot height). Also, \(\beta_A \approx 2.1 \cdot 10^5 m/s\) and \(\beta_B \approx 4.6 \cdot 10^4 m/s\). From Fig. 1 we can infer that the two-phonon processes become dominant for magnetic fields \(B < 2 T\) \((B < 0.5 T)\) and for temperatures \(T > 2 K\) \((T > 3 K)\) for InAs (GaAs) QDs. These estimates for the relaxation rates depend on one- and two-phonon processes are comparable to the ones recently measured in Refs. 6, 12, thus providing a reasonable explanation for these measurements. Note that, in contrast to the HH case, the relaxation time for electrons shows no deviation from the one-phonon time (or saturation) with decreasing B-field \([21]\).

Next, we provide explicit expressions of the relaxation rates for low and high temperature limits. The rates \(\Gamma^{(i)}\) can be written as

\[
\Gamma^{(i)} = \delta_i \sum_{m=0}^{m} \frac{\omega_{0_A}^{2m} \omega_{0_B}^{2m}}{\omega_{r}^{2m}} F_i^{(m)}(t),
\]

where \(\delta_i \approx 2\pi (h^4 e^2 \epsilon c^A_i / \hbar^2 m_{e_B} \lambda^5_0 \rho^A_c c^A_i), \delta_2 \approx \pi (m^A_{h} \beta^2 \Delta_0 / \hbar^2 \lambda^5_0 \rho^A_c c^A_i), \delta_3 \approx \pi (m^B_{h} \beta^2 \Delta_0 / \hbar^2 \lambda^5_0 \rho^A_c c^A_i),\)

ratios \(t = k_B T/E_{ph}\), \(d/\lambda\), and \(c_1/c_\lambda\). In Table II we list the asymptotic (scaling) expressions for \(F_i^{(m)}(t)\) in low B-fields \(\omega_{0A} \ll \omega_0\) for low \((t \ll 1)\) and high \((t \gg 1)\) temperatures. We note that \(F_i^{(1)}(t) \approx F_i^{(2)}(t)\) in both regimes, and \(F_i^{(3,4,5)} = 0\).

Using Eq. (9) and Table II we can write for the two-phonon rates, say, for InAs QDs

\[
\Gamma^{(2)} = \delta_2 \left\{ 10^7 \left( \frac{\omega_0^2}{\omega_{0_B}^2} + \frac{\omega_0^2}{\omega_{0_A}^2} + 0.5 \frac{\omega_0^2}{\omega_{0_B}^2} \right) T^{13}, T \ll E_{ph} \right\} + 10^2 \left( \frac{\omega_0^2}{\omega_{0_B}^2} + \frac{\omega_0^2}{\omega_{0_A}^2} + 0.3 \frac{\omega_0^2}{\omega_{0_B}^2} \right) T^2, T \gg E_{ph} \right\}.
\]

\[
\Gamma^{(3)} = \delta_3 \left\{ 10^9 T^{15}, T \ll E_{ph} \right\} + 0.3 T^2, T \gg E_{ph}. \]

From Eqs. (10) we find that for \(T < 2K\) and for \(B > 0.5T\) the one-phonon processes dominate the relaxation rate \(\Gamma\). On the other hand, for low B-fields \((0.1T < B < 1T)\) and finite temperatures \((T > 2K)\) the two-phonon processes will give the main contribution to \(\Gamma\), see Fig. 2. The main phonon processes could be identified.
experimentally by analyzing the temperature dependence of $\Gamma$, scaling as $\Gamma \sim T$ for one-phonon processes and as $\Gamma \sim T^2$ for two-phonon processes. Also, the saturation of $\Gamma$ in vanishing $B$-field is a clear indication of two-phonon processes. Note that the strong enhancement of the two-phonon HH spin relaxation arises because (i) the rate is 2nd order in SOI (whereas for electrons it is 4th order) and (ii) the effective mass for HHs is much larger than that for electrons.

In order to compute $\Gamma^{(2,3)}$, we took into account only the contribution from the deformation potential since this dominates the two-phonon relaxation for $T/E_{ph} > 0.1$ and $\omega_\perp, \omega_\parallel \ll \omega_0$. For the evaluation of $\Gamma^{(1)}$ instead, we considered both the piezoelectric and deformation potential contributions, both of them being important for $B$ and $T$ considered here. Surprisingly, we found that the ZFR rate $\Gamma^{(3)}$ increases when decreasing the dot size as $\Gamma^{(3)} \sim \lambda_\perp^{-1}$, while the other two rates decrease with decreasing the dot size as $\Gamma^{(1)} \sim \lambda_\parallel$ and $\Gamma^{(2)} \sim \lambda_\parallel$. This behavior strongly differs from the electronic case where the ZFR mechanism is efficient for rather large dots.

Interestingly, the present results do not change much if the $B$-field is tilted with respect to the QD plane. The $g$-factor for HHs is strongly anisotropic with $g_\parallel \ll g_\perp$ so that one can neglect the in-plane Zeeman splitting. This implies performing the substitution $\omega_\perp, \omega_\parallel \rightarrow \omega_\perp \cos \theta$ in above results, with $\theta$ being the angle between the $B$-field and the $z$-direction. This will lead to a reduction of the $B$-dependent rates ($\Gamma^{(1,2)}$), while the ZFR ($\Gamma^{(3)}$) being independent of $B$ remains the same.

In conclusion, we have shown that two-phonon processes give rise to a strong relaxation of the HH spin in a flat quantum dot. This time is predicted to be in the millisecond range, comparable to the one measured in recent experiments on optical pumping of a HH spin in QDs.

Though other sources of relaxation are not excluded, a careful scaling analysis of the measured relaxation time with the magnetic field and/or the temperature should allow one to identify the two-phonon process as the leading relaxation mechanism for the heavy-hole spin localized in small QDs.

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