

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 semiconducting nanostructures such as quantum dots (QDs) [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.

A basic requirement for a good qubit is that it can be initialized in a given state (say, spin up) and that the relaxation and decoherence times be much longer when compared to the switching times for single- and two-qubit operations. The spin of a HH localized in a quantum dot has been successfully initialized [4], and the relaxation time has been measured [4,5], and found to be on the order of 100 microseconds. The relaxation (T_1) and decoherence (T_2) times of a HH spin localized in a flat QD are, like for electrons, determined by the interaction of the HH 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) [6,7]. More importantly, it is of Ising type, making it ineffective for HH spins initialized along the growth direction [6], as typically done in experiments [4], thus implying very long dephasing times. This is in contrast to electrons, where the hyperfine interaction is isotropic and dominates the spin dynamics at low B fields [8–11].

Phonons couple to the HH spin through the spin-orbit interaction (SOI) [12]. The predicted values [12] for the one-phonon induced relaxation time T_1 agree quite well with data obtained in high B fields [5]. However, for low B fields ($B \sim 1.5$ – 3 T) and high temperatures ($T > 2$ K), a clear deviation from the one-phonon theory has been observed [5]. Furthermore, recent experiments on optical

pumping of HH spins in QDs showed saturation of T_1 for very low or even vanishing B field [4]. The relaxation time was found to be unusually long, $T_1 \approx 0.1$ – 1 ms, like previously observed in high B fields [5]. 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 [4,5]. The importance of such two-phonon processes was noticed a long time ago for electron spins in silicon-donors [13] and rare-earth ions [14], while for electrons in QDs it was shown that these processes are negligible compared to nuclear spin effects [15].

To describe a HH confined to a QD and interacting with the surrounding phonon bath, we start with the following Hamiltonian

$$H_h = H_0 + H_Z + H_{SO} + H_{h-ph} + H_{ph}, \quad (1)$$

where $H_0 = p^2/2m_h + V(\mathbf{r})$, is the dot Hamiltonian, $V(\mathbf{r}) \equiv m_h \omega_0^2 r^2/2$ is the confinement potential which is assumed to be harmonic, with m_h being the HH mass. The second term in Eq. (1) is the Zeeman energy of the HH (pseudo-) spin $H_Z = g\mu_B \mathbf{B} \cdot \boldsymbol{\sigma}/2$, with \mathbf{B} being the magnetic field and $\boldsymbol{\sigma}$ 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 [12]

$$H_{SO} = \beta p_- p_+ p_- \sigma_+ + \text{H.c.} \quad (2)$$

This Hamiltonian represents the effective Dresselhaus SOI (restricted to the HH subspace) due to bulk inversion asymmetry of the crystal [12], where $p_{\pm} = p_x \pm ip_y$, $\mathbf{p} = -i\hbar\nabla - e\mathbf{A}(\mathbf{r})$, $\mathbf{A}(\mathbf{r}) = (-y, x, 0)B/2$, 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 [12]. 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

$$M_{qj} = \frac{F(q_z)e^{iq_r}}{\sqrt{2\rho_c\omega_{qj}}} [e\beta_{qj} - i(\Xi_0\mathbf{q} \cdot \mathbf{d}_{qj} - \Xi_z q_z d_{qj}^z)], \quad (3)$$

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, $\omega_{qj} = c_j q$ the phonon energy, with c_j the speed of sound in the j th branch, \mathbf{d}_{qj} the polarization unit vector, ρ_c the sample density (per unit volume), and $e\beta_{qj}$ the piezoelectric electron-phonon coupling and $\Xi_{0,z}$ the deformation potential constants [12]. The form factor $F(q_z)$ in Eq. (3) 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. (1) describes the free phonon bath.

In the following, we analyze the effect of the phonons on the HH spin. The phonons do not couple directly to the spin, but the SOI plays the role of the mediator of an effective spin-phonon interaction. Let us define the dot Hamiltonian $H_d \equiv H_0 + H_Z + H_{SO}$. These eigenstates $|n\sigma\rangle$ of H_d are formally connected to the eigenstates $|n\rangle|\sigma\rangle$ of $H_0 + H_Z$ by an exact Schrieffer-Wolff (SW) transformation [16,17], i.e., $|n\sigma\rangle = e^S|n\rangle|\sigma\rangle$, where $S = -S^\dagger$ 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.).

In order to derive the effective spin-phonon interaction, we perform another SW transformation of the total HH Hamiltonian \tilde{H}_h . We get an effective Hamiltonian $H_{\text{eff}} = e^T \tilde{H}_h e^{-T}$, where $T = -T^\dagger$ is chosen such that it diagonalizes \tilde{H}_{h-ph} in the eigenbasis of H_d . In lowest order in H_{h-ph} , we obtain $T \approx \tilde{L}_d^{-1} \tilde{H}_{h-ph}$, where the Liouvillian is defined as $\tilde{L}_d A = [\tilde{H}_d, A]$, $\forall A$, 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

$$H_{s-ph} = \boldsymbol{\sigma} \cdot \sum_{qj,q'j'} [\delta_{qj,q'j'} \mathbf{C}_{qj}^{(1)} X_{qj} + \mathbf{C}_{qj,q'j'}^{(2)} X_{qj} X_{q'j'} + \mathbf{C}_{qj,q'j'}^{(3)} (P_{qj} X_{q'j'} - P_{q'j'} X_{qj})], \quad (4)$$

with $\boldsymbol{\sigma} \cdot \mathbf{C}_{qj}^{(1)} = \langle 0 | \tilde{M}_{qj} | 0 \rangle$, $\boldsymbol{\sigma} \cdot \mathbf{C}_{qj,q'j'}^{(2)} = \langle 0 | [\tilde{L}_d^{-1} \tilde{M}_{qj}, \tilde{M}_{q'j'}] | 0 \rangle$, $\boldsymbol{\sigma} \cdot \mathbf{C}_{qj,q'j'}^{(3)} = \langle 0 | [\tilde{L}_d^{-1} \tilde{M}_{qj}, \tilde{L}_d^{-1} \tilde{M}_{q'j'}] | 0 \rangle$, $P_{qj} = i\sqrt{\hbar\omega_{qj}}(a_{-qj}^\dagger - a_{qj})$ is the phonon field momentum operator, and $|0\rangle$ is the orbital ground state. In Eq. (4) we have neglected 2nd order corrections in SOI to the energy levels. Note that for vanishing magnetic field $\mathbf{B} \rightarrow 0$ only the last term in H_{s-ph} is nonzero, since only this one preserves time-reversal invariance and thus gives rise to zero field relaxation (ZFR) [13–15].

We now assume the orbital energy $\hbar\omega_0$ is much larger than the SOI, i.e., $\|H_0\| \gg \|H_{SO}\|$, and treat the SOI to leading order in perturbation theory. We consider also the B field to be applied perpendicularly to the dot plane (as in Refs. [4,5]). The SW-generator S can be written as $S =$

$S_+ \sigma_- - \text{H.c.}$, and we then find

$$S_+ = A_1 p_+ p_- p_+ + A_2 [p_+ p_- p_+ - (p_+ p_- - p_+ p_-) p_+] + A_4 p_+ p_- p_+ + A_3 [(p_+ p_- - p_+ p_-) p_+ + p_+ p_- p_+]. \quad (5)$$

Here, $A_i \equiv A_i(\omega_Z, \omega_c)$ with $\omega_Z = g\mu_B B/\hbar$ and $\omega_c = eB/2c$. For $\omega_Z, \omega_c \ll \omega_0$, we obtain $A_1 \approx -(7\beta/9\hbar) \times [(\omega_Z + \omega_c)/\omega_0^2]$, $A_2 \approx -(\beta/3\hbar)(\omega_c/\omega_0^2)$, $A_3 \approx -(2\beta/9\hbar)[\omega_c^2(\omega_c + \omega_Z)/\omega_0^4]$, and $A_4 \approx (2\beta/3\hbar) \times (\omega_c^3/\omega_0^4)$, while $P_\pm = P_x \pm iP_y$ with $P_{x(y)} = -i\hbar\nabla_{x(y)} \pm (m_h\omega_0^2/\omega_c)y(x)$. After somewhat tedious calculations, we obtain analytic expressions for $\mathbf{C}^{(i)} = (C^{(i,x)}, C^{(i,y)}, 0)$ occurring in Eq. (4). We give below only the exact expression for $i = 3$, the rest being too lengthy to be displayed here:

$$C_{qj,q'j'}^{(3,x/y)} = \pm M_{qj}^{q'j'} \frac{m_h \lambda_d^2 \beta e^{-q^2 \lambda_d^2/4}}{3\hbar\omega_0^2} \mathcal{F}(\mathbf{q} \cdot \mathbf{q}') \times [q_y^2 q_x' - q_y^2 q_x \pm (q_x - q_x')(2q_y q_y' + 3q_x q_x')], \quad (6)$$

where

$$\mathcal{F}(\mathbf{q} \cdot \mathbf{q}') = \frac{1}{\lambda_d^2(\mathbf{q} \cdot \mathbf{q}')^2} (e^{-\lambda_d^2 \mathbf{q} \cdot \mathbf{q}'/2} - \lambda_d^2 \mathbf{q} \cdot \mathbf{q}'/2) [\gamma + \log(\lambda_d^2 \mathbf{q} \cdot \mathbf{q}'/2) + \Gamma(0, \lambda_d^2 \mathbf{q} \cdot \mathbf{q}'/2)]. \quad (7)$$

Here, $M_{qj}^{q'j'} = (F(q_z)F(q'_z)\hbar/2\rho_c\sqrt{\omega_{qj}\omega_{q'j'}})(\Xi_0\mathbf{q} \cdot \mathbf{d}_{qj} - \Xi_z q_z d_{qj}^z)(\Xi_0\mathbf{q}' \cdot \mathbf{d}_{q'j'} - \Xi_z q'_z d_{q'j'}^z)$, where λ_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. (4). We first mention that all terms in Eq. (4) can be cast in a general spin-boson type of Hamiltonian $H_{s-b}^p = (1/2)g\mu_B \delta\mathbf{B}^p(t) \cdot \boldsymbol{\sigma}$, $p = 1, 2, 3$, with the corresponding identification of the fluctuating magnetic field terms $\delta\mathbf{B}_j(t)$ from Eq. (4) (e.g., $\delta\mathbf{B}^1(t) \sim \mathbf{C}_{qj}^{(1)} X_{qj}$).

Within the Bloch-Redfield approach, the relaxation rate $\Gamma \equiv 1/T_1$ can be expressed as $\Gamma = \sum_{i=x,y} [J_{ii}(E_Z/\hbar) + J_{ii}(-E_Z/\hbar)]$. The correlation functions J_{ij} are defined by $J_{ij}(\omega) = (g\mu_B/2\hbar)^2 \int_0^\infty dt e^{-i\omega t} \langle \delta B_i(0) \delta B_j(t) \rangle$, where $\langle \dots \rangle$ denotes the average over the phonon bath, assumed to be in thermal equilibrium at temperature T . The relaxation time associated with the three types of spin-phonon processes in Eq. (4) is $\Gamma = \sum_{i=1,2,3} \Gamma^{(i)}$ with

$$\Gamma^{(1)} = \frac{4\pi}{\hbar} \sum_{qj} |\mathbf{C}_{qj}^{(1)}|^2 \left(n(\omega_{qj}) + \frac{1}{2} \right) \delta(E_Z - \hbar\omega_{qj}),$$

$$\Gamma^{(m)} \simeq \frac{8\pi}{\hbar} \sum_{qj,q'j'} |\mathbf{C}_{qj,q'j'}^{(m)}|^2 (\omega_{qj}\omega_{q'j'})^{m-2} n(\omega_{qj}) \times [n(\omega_{q'j'}) + 1] \delta(\hbar\omega_{qj} - \hbar\omega_{q'j'}), \quad (8)$$

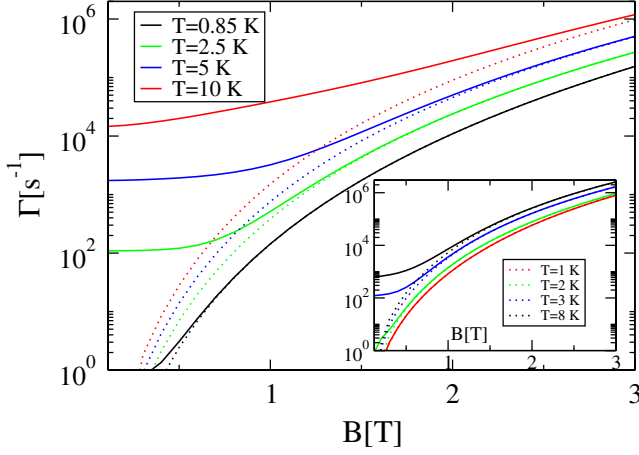


FIG. 1 (color online). The heavy-hole spin-relaxation rate Γ for InAs QDs (GaAs QDs in the inset) as a function of magnetic field B for different temperatures T . The solid lines represent the rate due to one- and two-phonon processes, i.e., $\Gamma = \sum_{i=1}^3 \Gamma^{(i)}$ as defined in Eq. (8) for different temperatures T , while the dotted lines represent the one-phonon rate $\Gamma^{(1)}$.

where $n(\omega) = 1/[\exp(\omega/k_B T) - 1]$ is the Bose factor and $m = 2, 3$ correspond to B -dependent and B -independent two-phonon rates, respectively. We remark that in Eq. (8) we have neglected some irrelevant processes in the limit of low- B field [18]. Also, for B fields perpendicular to the dot plane the decoherence time satisfies $T_2 = 2T_1$ for one- and two-phonon processes since the spin-phonon fluctuations $\delta \mathbf{B}_j \perp \mathbf{B}$ [12,17].

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 Γ as a function of the B field and of temperature, respec-

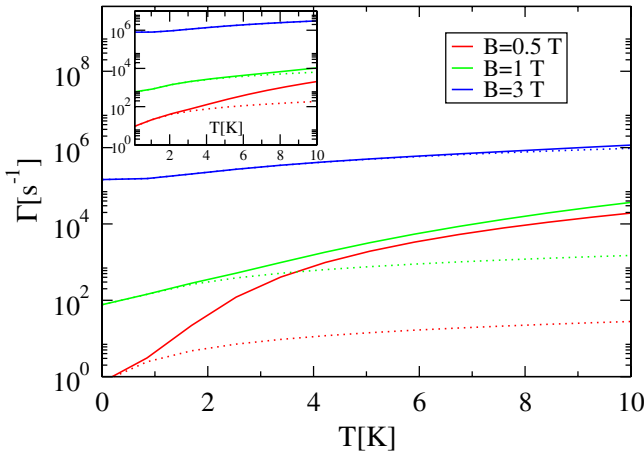


FIG. 2 (color online). The heavy-hole spin-relaxation rate Γ in Eq. (8) for InAs QDs (GaAs QDs in the inset) as a function of temperature T for different B -field values. For finite B field, Γ saturates at low temperatures due to one-phonon processes.

tively, for InAs and GaAs quantum dots. Figure 1 shows a clear saturation of Γ 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 [12].

For these plots, we used the following HH InAs QDs (labeled by A) [19,20] and GaAs QDs (labeled by B) parameters [12]: $\Xi_0 = 1.9$ eV, $\Xi_z = 2.7$ eV, $c_l^A = 2.64 \times 10^3$ m/s ($c_l^B = 3.35 \times 10^3$ m/s), $c_l^A = 3.83 \times 10^3$ m/s ($c_l^B = 4.73 \times 10^3$ m/s), $\rho_c^A = 5.68 \times 10^3$ kg/m³ ($\rho_c^B = 5.3 \times 10^3$ kg/m³), $m_h^A = 0.25m_e$ ($m_h^B = 0.14m_e$), $g^A = 1.4$ ($g^B = 2.5$), and we assume $\lambda_d = 3$ nm ($\hbar\omega_0^A = 35$ meV, $\hbar\omega_0^B = 60$ meV) and $d = 3$ nm (dot height). Also, $\beta_A \approx 4.2 \times 10^{18} \text{ m}^3/\text{eV}^2 \text{ s}^3$ and $\beta_B \approx 2 \times 10^{18} \text{ m}^3/\text{eV}^2 \text{ s}^3$. 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 due to one- and two-phonon processes are comparable to the ones recently measured in Refs. [4,5], 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

$$\Gamma^{(i)} = \delta_i \sum_{m=0}^{r_i} \frac{\omega_Z^{r_i-m} \omega_c^m}{\omega_0^{r_i}} F_i^{(m)}(t), \quad (9)$$

where $\delta_1 \approx 2\pi(\hbar^4 e h_{14}^2 \beta^2 / \kappa^2 m_h \lambda_d^6 \rho_c c_l^5)$, $\delta_2 \approx \pi(m_h^4 \beta^2 \Xi_0^4 / \hbar^2 \lambda_d^5 \rho_c^2 c_l^3)$, $\delta_3 \approx \pi(m_h^6 \beta^2 \Xi_0^4 / \hbar^4 c_l \lambda_d^3 \rho_c^2)$, $r_1 = 5$, $r_2 = 2$, $r_3 = 0$, and $t = k_B T / E_{\text{ph}}$ with $E_{\text{ph}} \equiv \hbar c_l / \lambda_d$.

The functions $F_i^{(m)}(t)$ depend on the ratios $t = k_B T / E_{\text{ph}}$, d/λ , and c_l/c_t . In Table I we list the asymptotic (scaling) expressions for $F_i^{(m)}(t)$ in low B fields $\omega_{c,Z} \ll \omega_0$ for low ($t \ll 1$) and high ($t \gg 1$) temperatures. We note that $F_1^{(1)}(t) \approx F_1^{(2)}(t)$ in both regimes, and $F_1^{(3,4,5)} \equiv 0$.

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

$$\begin{aligned} \Gamma^{(2)} &= \delta_2 \begin{cases} 10^7 (10 \frac{\omega_Z^2}{\omega_0^2} + \frac{\omega_Z \omega_c}{\omega_0^2} + 0.5 \frac{\omega_c^2}{\omega_0^2}) t^{13}, & t \ll 1 \\ 10^2 (\frac{\omega_Z^2}{\omega_0^2} + \frac{\omega_Z \omega_c}{\omega_0^2} + 0.3 \frac{\omega_c^2}{\omega_0^2}) t^2, & t \gg 1 \end{cases} \\ \Gamma^{(3)} &= \delta_3 \begin{cases} 10^9 t^{15}, & t \ll 1 \\ 0.3 t^2, & t \gg 1 \end{cases}. \end{aligned} \quad (10)$$

TABLE I. The asymptotic values for $F_i^{(m)}(t)$.

	$F_1^{(0)}$	$F_1^{(1)}$	$F_2^{(0)}(t)$	$F_2^{(1)}(t)$	$F_2^{(2)}(t)$	$F_3(t)$
$t \ll 1$	0.004	0.015	$10^8 t^{13}$	$10^7 t^{13}$	$5 \times 10^6 t^{13}$	$10^9 t^{15}$
$t \gg 1$	$0.08 \frac{t}{\omega_Z}$	$0.03 \frac{t}{\omega_Z}$	$10^2 t^2$	$10^2 t^2$	$30 t^2$	$0.3 t^2$

From Eq. (10) we find that for $T < 2$ K and for $B > 0.5$ T the one-phonon processes dominate the relaxation rate Γ . On the other hand, for low B fields (0.1 T $< B < 1$ T) and finite temperatures ($T > 2$ K) the two-phonon processes will give the main contribution to Γ , see Fig. 2. The main phonon processes could be identified experimentally by analyzing the temperature dependence of Γ , scaling as $\Gamma \sim T$ for one-phonon processes and as $\Gamma \sim T^2$ for two-phonon processes. Also, the saturation of Γ 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. Even more, the coupling of the phonon field to the HH spin is qualitatively different compared to electrons (in-plane coupling vs perpendicular-to-the-plane coupling) allowing for a clear distinction between linear (electrons) and cubic (holes) in momentum SOI via two-phonon relaxation processes.

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 $T/E_{\text{ph}} > 0.1$ and $\omega_Z, \omega_c \ll \omega_0$. 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 $\Gamma^{(3)} \sim \lambda_d^{-1}$, while the other two rates decrease with decreasing the dot size as $\Gamma^{(1)} \sim \lambda_d^4$ and $\Gamma^{(2)} \sim \lambda_d$. This behavior strongly differs from the electronic case where the ZFR mechanism is efficient for rather large dots [15].

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 $\omega_{c,Z} \rightarrow \omega_{c,Z} \cos \theta$ in above results, with θ 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 [4]. 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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