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Spin–orbit-induced hole spin relaxation in InAs and GaAs quantum dots

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Abstract. We study the effect of valence band spin–orbit interactions (SOI) on the acoustic phonon-assisted spin relaxation of holes confined in quantum dots (QDs). Heavy hole–light hole (hh–lh) mixing and all the spin–orbit terms arising from zinc-blende bulk inversion asymmetry (BIA) are considered on equal footing in a fully three-dimensional Hamiltonian. We show that hh–lh mixing and BIA have comparable contributions to the hole spin relaxation in self-assembled QDs, but BIA becomes dominant in gated QDs. Simultaneously accounting for both mechanisms is necessary for quantitatively correct results in quasi-two-dimensional QDs. The dependence of the hole spin relaxation on the QD geometry and spin splitting energy is drastically different from that of electrons, with a non-monotonic behavior which results from the interplay between SOI terms. Our results reconcile contradictory predictions of previous theoretical works and are consistent with experiments.

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Over the last few years, the spin of holes confined in III–V semiconductor quantum dots (QDs) has emerged as a promising building block for spintronic and spin-based quantum information devices [1]. As compared to electrons, the p-like nature of the hole orbitals leads to weaker hyperfine interaction with the lattice nuclei, resulting in coherence times which hold promise for applications [2–7]. As a matter of fact, demonstrations of hole spin manipulation in QDs have been recently reported [8–10] and theoretical proposals of control mechanisms are being proposed [11–14]. In this context, the study of hole spin relaxation has become a subject of interest. Hole spin relaxation is also important for optical applications because it is believed to rule the exciton spin dynamics in both dark-to-bright exciton transitions [15–18], which affect exciton storage times [19–21], and transitions within the bright doublet [22], which affect light depolarization [17].

Experimental observations in self-assembled InAs QDs point at hole spin lifetimes ranging from $T_1^h \approx 10$ ps to 1 ms [2, 23–26]. The large dispersion is partly attributed to the different relaxation mechanisms involved in different studies. When the energy splitting between orthogonal spin states is small, hyperfine interaction is the dominant relaxation channel [16, 27]. In this case, the lifetime is strongly dependent on the degree of hh–lh mixing. If the hole state is a pure hh, as in the ground state of flat (quasi-two-dimensional (2D)) QDs, the hyperfine interaction takes an Ising form and spin relaxation is slow, but it rapidly increases in non-flat QDs due to hh–lh mixing [1, 7]. On the other hand, when the energy splitting exceeds the nuclear magnetic field, the valence band spin–orbit interaction (SOI) takes over as the main source of relaxation [16, 27]. Long hole spin lifetimes have then been observed, reaching up to $T_1^h \approx 0.25$ ms, which is only five to ten times shorter than electron spin lifetimes, $T_1^e$ [26]. This result is encouraging for the use of holes in quantum information and optical applications, but it is surprising because the valence band SOI is known to be much stronger than that of the conduction band [28].

The above paradox has prompted a number of theoretical works trying to understand which factors determine the relaxation dynamics of single holes under magnetic fields [29–33] and
that of holes forming excitons [15, 22, 34, 35] in quasi-2D InAs/GaAs QDs. For the relaxation to take place one needs a source of energy relaxation, which in these systems is provided by the acoustic phonon bath [23, 24], plus a source of spin admixture. Woods et al [29] and Lu et al [30] proposed that the latter is the coupling between hh and lh subbands. Other authors have suggested instead that the splitting between hh and lh subbands in flat QDs is large owing to confinement and strain, so that spin admixture must be due to other SOI mechanisms. It was then proposed that hole SOI should have an origin similar to that of conduction electrons, namely the bulk inversion asymmetry (BIA) of zinc-blende crystals, which gives rise to Dresselhaus SOI terms [28]. Bulaev and Loss assumed that the cubic-in-\textit{k} Dresselhaus term is dominant and showed that $T_{th}^1$ could then become comparable to $T_{te}^1$ in flat QDs [31]. Other studies followed this assumption and succeeded in explaining some experimental observations [15, 26, 32]. By contrast, Tsitsishvili et al [35] suggested that if the lateral confinement is weak, it is the linear-in-\textit{k} term that dominates the mixing. Last, Roszak et al [34] suggested that for holes forming excitons, it is the electron–hole (e–h) exchange interaction together with the strain that gives rise to hole spin admixture.

It is worth noting that all the previous works assumed a dominating SOI mechanism without actually comparing it with others. In addition, simplified models disregarding hh–lh mixing become highly parametric, and different parameters were needed to explain different experimental observations even in the same system [15, 26, 32]. The lack of a comprehensive study translates into many open questions which show that hole spin relaxation in QDs is still not fully understood. To name a few: (i) while Woods et al [29] predict that $T_{th}^1$ decreases with the QD diameter, Lu et al [30] predict exactly the opposite behavior; (ii) Bulaev and Loss [31] predicted $T_{th}^1 > T_{te}^1$ in the limit of 2D QDs, but experiments on self-assembled QDs have only shown $T_{th}^1/T_{te}^1 = 5 − 10$ [26], so that one wonders if any realistic QD structure would actually permit holes relaxing slower than electrons; (iii) in excitons, the role of e–h exchange energy is not clear: while experiments with self-assembled QDs have shown negligible dependence of $T_{th}^1$ [24], a strong dependence has been found in colloidal quantum rods [36].

In this work, we aim at covering the existing gap in the understanding of hole spin relaxation in QDs. We study the hole spin dynamics considering simultaneously the most relevant intrinsic SOI terms of III–V QDs, namely hh–lh mixing and all the different Dresselhaus SOI terms arising from the BIA of zinc-blende crystals, along with the hole–acoustic phonon coupling. All the terms are described within a four-band $k \cdot p$ formalism and three-dimensional (3D) Hamiltonians, which allows us to provide a general overview on the effect of the QD size and geometry dependence while relying on well-known bulk parameters only. In this way, we are able to establish the regime of validity of previous studies which assumed a single dominating SOI mechanism. Furthermore, we explore spheroidal QDs beyond the usual quasi-2D limit, thus providing theoretical assessment for developing experimental research with spherical and prolate QDs [18, 36].

We find that hh–lh is the main SOI channel in prolated or spherical QDs, but Dresselhaus SOI has a comparable contribution in oblated QDs (such as self-assembled dots), and it becomes dominant in quasi-2D QDs with very weak confinement (as in electrostatically confined dots). The competition between SOI coupling terms and the energy splitting between hh and lh leads to a non-monotonic dependence of $T_{th}^1$ with the QD geometry, in sharp contrast with the well-known case of electrons. This explains the opposite trends reported by different theoretical studies in the literature. The dependence of $T_{th}^1$ on the e–h exchange energy we predict is consistent with experiments on colloidal nanorods [36], but it suggests that...
two-phonon processes are relevant in self-assembled QDs. In prolate QDs, where the ground state is formed by lh, the spin relaxation is shown to take place in similar timescales as for transitions between hh states. However, the coupling to acoustic phonons is different, with deformation potential interaction being the main mechanism even for vanishing spin splitting energy.

2. Theoretical model

We study the spin relaxation of holes confined in zinc-blende QDs grown along the [001] direction. The hole spin states are considered split energetically, for example by the e–h exchange interaction in excitons or any other source that can be viewed as an effective axial magnetic field. Thus, similar results can be expected for transitions between Zeeman sublevels under moderate external magnetic fields.

2.1. Hamiltonian

The system Hamiltonian reads

\[ H = H_h + H_{ph} + H_{h-ph}. \]  

In equation (1), \( H_h \) is the hole Hamiltonian

\[ H_h = H_L + H_{BIA} + V_{QD} I + H_Z, \]  

where \( H_L \) is the four-band Luttinger Hamiltonian describing the coupled hh–lh bands [37]. It includes quadratic terms in \( k \) only:

\[ H_L = \frac{1}{m_0} \left[ \left( \gamma_1 + \frac{5}{2} \gamma_2 \right) \frac{k^2}{2} - \gamma_2 (k_x^2 J_x^2 + k_y^2 J_y^2 + k_z^2 J_z^2) \right. \]

\[ \left. -2 \gamma_3 (\{k_x, k_y\} \{J_x, J_y\} + \{k_y, k_z\} \{J_y, J_z\} + \{k_z, k_x\} \{J_z, J_x\}) \right], \]

where \( m_0 \) is the free electron mass, \( \gamma_i \) are the Luttinger parameters, \( k_j = -i \hbar \partial_j \) the \( j \) component of the linear momentum, \( \{A, B\} = \frac{1}{2} (AB + BA) \) and \( J_i \) is the \( i \)th component of the angular momentum corresponding to the quantum number \( J = 3/2 \). To obtain the matrix representation of this Hamiltonian we multiply the first term of equation (3) by the 4 × 4 unit matrix and employ the standard matrix representation of the \( J = 3/2 \) components of the angular momentum [38]. We finally obtain

\[
H_L = \begin{pmatrix}
P + Q & -S & R & 0 \\
-S^\dagger & P - Q & 0 & R \\
R^\dagger & 0 & P - Q & S \\
0 & R^\dagger & S^\dagger & P + Q \\
\end{pmatrix}
\]

with

\[
P = \frac{1}{2 m_0} \gamma_1 \left( k_x^2 + k_y^2 + k_z^2 \right),
\]

\[
Q = \frac{1}{2 m_0} \gamma_3 \left( k_x^2 + k_y^2 - 2k_z^2 \right).
\]
Figure 1. Geometry of QDs with varying lateral (a) and vertical (b) confinement frequency.

\[ R = \frac{1}{2m_0} \left[ -\sqrt{3} \gamma_2 (k_x^2 - k_y^2) + 2i \sqrt{3} \gamma_3 k_x k_y \right], \quad (7) \]

\[ S = \frac{1}{2m_0} 2 \sqrt{3} \gamma_3 (k_x - i k_y) k_z. \quad (8) \]

\[ H_{\text{BIA}} \] includes the linear and the Dresselhaus SOI third order in \( k \) terms \([28]\):

\[
H_{\text{BIA}} = \frac{2}{\sqrt{3}} C_k [k_x \{ J_x, J_x^2 - J_z^2 \} + \text{cp}] + b_{41} (\{ k_x, k_y^2 - k_z^2 \} J_x + \text{cp}) + b_{42} (\{ k_x, k_y^2 - k_z^2 \} J_x^3 + \text{cp}) \\
+ b_{51} (\{ k_x, k_y^2 + k_z^2 \} \{ J_x, J_y^2 - J_z^2 \} + \text{cp}) + b_{52} (k_z^3 \{ J_x, J_y^2 - J_z^2 \} + \text{cp}), \quad (9)\]

where \( C_k, b_{41}, b_{42}, b_{51} \) and \( b_{52} \) are material-dependent coefficients and \( \text{cp} \) stands for cyclic permutations of the preceding terms. The matrix form of the Hamiltonian terms above is given in the appendix. One can note that all BIA terms provide direct mixing between hh spin-up and spin-down (\( J_z = \pm 3/2 \) and \( \pm -3/2 \)) components except for \( b_{41} \), which requires the participation of the lh (\( J_z = \pm 1/2 \) and \( \pm -1/2 \)) components. Rashba SOI is neglected in this study because it is an extrinsic effect, which depends on the details of the electric field felt by the system. Besides, for holes it couples energetically distant states so that, under moderate external fields, it is less efficient than Dresselhaus SOI \([31]\). \( V_{QD} \) describes the confining potential of the QD. We model QDs with parabolic confinement in the \( x, y \) and \( z \) directions:

\[ V_{QD} = -\frac{1}{2} \chi_\perp (x^2 + y^2) - \frac{1}{2} \chi_\parallel z^2, \quad (10) \]

where \( \chi_\perp \) and \( \chi_\parallel \) are the force constants perpendicular and parallel to the growth direction, respectively. Equation (10) allows us to simulate 3D spheroidal QDs with different aspect ratios, from flat (quasi-2D) to spherical or elongated (quasi-one-dimensional (1D)) structures, see figure 1. \( H_Z \) is the Hamiltonian modeling the splitting of the hole states by an effective axial magnetic field, three times larger for heavy (\( |J_z| = 3/2 \)) than for light holes (\( |J_z| = 1/2 \)). This field could originate, e.g. from the e–h exchange interaction \([34]\) or a spin Zeeman effect. Then, we assume that

\[
H_Z = \frac{1}{2} \begin{pmatrix}
\Delta & 0 & 0 & 0 \\
0 & \frac{1}{3} \Delta & 0 & 0 \\
0 & 0 & -\frac{1}{3} \Delta & 0 \\
0 & 0 & 0 & -\Delta
\end{pmatrix}. \quad (11)
\]
To calculate the hole states from $H_h$, we note that the diagonal terms correspond to harmonic oscillator Hamiltonians:

$$P + Q + V_{QD} = T_{hh, \perp} - \frac{1}{2} \chi_\perp (x^2 + y^2) + T_{hh, z} - \frac{1}{2} \chi_z z^2$$ (12a)

and

$$P - Q + V_{QD} = T_{hh, \perp} - \frac{1}{2} \chi_\perp (x^2 + y^2) + T_{hh, z} - \frac{1}{2} \chi_z z^2,$$ (12b)

where $T_{i,j} = \frac{q^2}{2m_i} k_j^2$, with $i = (hh, lh)$, $j = (\perp, z)$, $k_\perp = (k_x^2 + k_y^2)^{1/2}$, $m_{z,h}^h = m_0/(\gamma_1 + \gamma_2)$, $m_{z,l}^h = m_0/(\gamma_1 - \gamma_2)$ and $m_{z,l}^l = m_0/(\gamma_1 + 2\gamma_2)$. This suggests rewriting all derivatives and coordinates of $H_h$ in terms of harmonic oscillator ladder operators and then projecting it onto a basis formed by oscillator eigenstates. The problem is that equation (12a) has hh masses while equation (12b) has lh masses, and hence they have different oscillator frequency, $\omega_i = (\chi_i/m_i)^{1/2}$. Because the off-diagonal terms of $H_L$ couple hh and lh components, it is convenient to use a single kind of oscillator state, e.g. hh state. This can be done by rewriting equation (12b) in terms of the hh harmonic oscillator Hamiltonians:

$$P - Q + V_{QD} = \frac{\gamma_1 - \gamma_2}{\gamma_1 + \gamma_2} H_{hh, \perp} - \frac{\gamma_2}{\gamma_1 + \gamma_2} \chi_\perp (x^2 + y^2) + \frac{\gamma_1 + 2\gamma_2}{\gamma_1 - 2\gamma_2} H_{hh, z} + \frac{2\gamma_2}{\gamma_1 - 2\gamma_2} \chi_z z^2,$$ (13)

where $H_{hh, \perp} = T_{hh, \perp} - \chi_\perp (x^2 + y^2)/2$ and $H_{hh, z} = T_{hh, z} - \chi_z (z^2)/2$. The resulting hole states are Luttinger spinors of the form

$$|\Psi_m^h\rangle = \sum_{r,J_z} c_{r,J_z}^m |v_x, v_y, v_z\rangle |3/2, J_z\rangle,$$ (14)

where $v_j = 0, 1, 2\ldots$ is the quantum number of the 1D hh harmonic oscillator along the $j$ direction, $r$ is the combined orbital quantum number, $r = (v_x, v_y, v_z)$ and $|3/2, J_z\rangle$ the Bloch function.

$H_{ph}$ in equation (1) is the Hamiltonian of acoustic phonons, given by $H_{ph} = \sum_{\lambda} \hbar \omega_{q,\lambda} a_{q,\lambda}^\dagger a_{q,\lambda}$, with $\omega_{q,\lambda}$ standing for the phonon energy spectrum of branch $\lambda$ ($\lambda = l, t_1, t_2$ for longitudinal and the two transversal phonon modes) and momentum $q$. We restrict to low phonon energies, where the linear dispersion regime applies, $\omega_{q,\lambda} = c_\lambda q$, with $c_\lambda$ as the phonon velocity.

$H_{h-ph}$ is the hole–phonon interaction

$$H_{h-ph} = e \phi_{pz} E + H_{dp},$$ (15)

where $e$ is the hole charge, $\phi_{pz}$ the piezoelectric potential and $H_{dp}$ the deformation potential term. These are the two relevant scattering mechanisms at low temperatures [30]. The piezoelectric potential is given by [39]

$$\phi_{pz} = \sum_\lambda \phi_{pz}(\lambda) = -\sum_{\lambda,q} \frac{4\pi i}{\epsilon_s q^2} h_{14} \left(q_x \epsilon_{yz}^\lambda + q_y \epsilon_{zx}^\lambda + q_z \epsilon_{xy}^\lambda\right),$$ (16)

where $\epsilon_s$ is the relative dielectric constant, $h_{14}$ is the piezoelectric constant and $\epsilon_{ij}$ is the strain tensor component. On the other hand, the deformation potential term is given by the Bir–Pikus
In turn, the deformation potential operators become

\[
\mathcal{H}_{dp} = \sum_{\lambda} \begin{pmatrix}
    p^\lambda + q^\lambda & -s^\lambda & r^\lambda & 0 \\
    -(s^\lambda)^\dagger & p^\lambda - q^\lambda & 0 & r^\lambda \\
    (r^\lambda)^\dagger & 0 & p^\lambda - q^\lambda & s \\
    0 & (r^\lambda)^\dagger & (s^\lambda)^\dagger & p^\lambda + q^\lambda
\end{pmatrix},
\]

where

\[
p^\lambda = -a (\epsilon_{xx}^\lambda + \epsilon_{yy}^\lambda + \epsilon_{zz}^\lambda),
\]

\[
q^\lambda = -\frac{b}{2} (\epsilon_{xx}^\lambda + \epsilon_{yy}^\lambda - 2\epsilon_{zz}^\lambda),
\]

\[
r^\lambda = \frac{\sqrt{3}}{2} b (\epsilon_{xx}^\lambda - \epsilon_{yy}^\lambda) - i d \epsilon_{xy}^\lambda,
\]

\[
s^\lambda = -d (\epsilon_{zx}^\lambda - i \epsilon_{yz}^\lambda).
\]

Here \(a, b\) and \(d\) are the valence band deformation potential constants.

The components of the strain tensor are rewritten using normal-modes coordinates \[29\]

\[
\varepsilon_{ij}^\lambda = -\frac{i}{2} \sum_{q} U^\lambda(q) (\eta_i^\lambda(q) q_j + \eta_j^\lambda(q) q_i) F(q, r),
\]

where \(F(q, r) = (e^{-iqr} a_q^* + e^{iqr} a_q)\) and \(U^\lambda(q) = \sqrt{\hbar/2 \rho V \omega_q \lambda}\), with \(\rho\) and \(V\) standing for the crystal density and volume. \(\eta_i(q)\) are the phonon polarization vectors: \(\eta_i(q) = (q_x, q_y, q_z)/q\), \(\eta_{11}(q) = (q_x, q_z, -q_y)/q \) and \(\eta_{12}(q) = (q_y, -q_x, 0)/q \), with \(q = \sqrt{q_x^2 + q_y^2 + q_z^2}\). The piezoelectric potential now reads

\[
\phi_{pw}^{11} = -\frac{12 \pi \hbar q_{y}}{e_t} U^1(q) \sum_{q} \frac{q_x q_y q_z}{q^3} F(q, r),
\]

\[
\phi_{pw}^{11} = -\frac{4 \pi \hbar q_{y}}{e_t} U^1(q) \sum_{q} \frac{q_x q_y (2q_z^2 - q_y^2)}{q^3 q^4} F(q, r),
\]

\[
\phi_{pw}^{12} = -\frac{4 \pi \hbar q_{y}}{e_t} U^1(q) \sum_{q} \frac{q_z (q_y^2 - q_z^2)}{q^3 q^4} F(q, r).
\]

In turn, the deformation potential operators become

\[
p^1 = i a U^1(q) \sum_{q} q F(q, r),
\]

\[
q^1 = \frac{b}{2} U^1(q) \sum_{q} \left( q - 3 \frac{q_z^2}{q} \right) F(q, r),
\]

\[
r^1 = -i U^1(q) \sum_{q} \left( \frac{\sqrt{3}}{2} b \frac{q_z^2 - q_y^2}{q} - i d \frac{q_x q_y}{q} \right) F(q, r),
\]

\[
s^1 = i d U^1(q) \sum_{q} \frac{q_z (q_x - i q_y)}{q} F(q, r).
\]
for longitudinal phonons,

\[ p^{11} = 0, \]

\[ q^{11} = \frac{i}{2} b U''(q) \sum_q \left( \frac{3 q_z q_\perp}{q} \right) F(q, r), \]

\[ r^{11} = -i U''(q) \sum_q \left( \frac{\sqrt{3} b}{2} \frac{q_z}{q_\perp} - i \frac{d}{2} \frac{q_x^2 - q_y^2}{q_\perp} \right) F(q, r), \]

\[ s^{11} = i \frac{d}{2} U''(q) \sum_q \left( \frac{q_z^2 - q_\perp^2}{q_\perp} \right) (q_x - i q_y) F(q, r) \tag{25} \]

for transversal t1 phonons and

\[ p^{12} = 0, \]

\[ q^{12} = 0, \]

\[ r^{12} = -i U''(q) \sum_q \left( \frac{\sqrt{3} b}{2} \frac{q_x}{q_\perp} - i \frac{d}{2} \frac{q_x^2 - q_\perp^2}{q_\perp} \right) F(q, r), \]

\[ s^{12} = -\frac{d}{2} U''(q) \sum_q \frac{q_z (q_x - i q_y)}{q_\perp} F(q, r) \tag{26} \]

for transversal t2 phonons.

### 2.2. Relaxation rate

We calculate the spin relaxation from an initial hole state |\( \Psi_1^h \rangle \), with energy \( E_1^h \), to a final hole state |\( \Psi_1^f \rangle \), with energy \( E_1^f \). The relaxation rate is estimated with a Fermi golden rule. We consider zero temperature, so that there is no phonon absorption. After integrating over phonon degrees of freedom, the rate is given by

\[ \tau_{1-1}^{-1} = \frac{2 \pi}{\hbar} \sum_{\lambda, \mathbf{q}} \left| \langle \Psi_1^f \mid \mathcal{H}_{\text{h-ph}}^{\lambda q} \mid \Psi_1^h \rangle \right|^2 \delta(\Delta E_1 + \hbar c \mathbf{q}). \tag{27} \]

Here \( \mathcal{H}_{\text{h-ph}}^{\lambda q} \) is the hole–phonon interaction Hamiltonian, equation (15), but for a fixed phonon branch \( \lambda \) and momentum \( \mathbf{q} \), and \( \Delta E_1 = E_1^f - E_1^h \). It can be seen from equations (23)–(26) that all the terms of \( \mathcal{H}_{\text{h-ph}}^{\lambda q} \) contain a factor which depends on \( \mathbf{q} \) only and \( F(q, r) \), which depends on spatial coordinates as well. Thus, when expanded, the matrix element \( \langle \Psi_1^f \mid \mathcal{H}_{\text{h-ph}}^{\lambda q} \mid \Psi_1^h \rangle \) takes the form

\[ \langle \Psi_1^h \mid \mathcal{H}_{\text{h-ph}}^{\lambda q} \mid \Psi_1^f \rangle = \sum_{J_z, J_z'} (c^\dagger_{r, J_z})^* c^\dagger_{r, J_z'} M^{\lambda}_{J_z, J_z'}(\mathbf{q}) G_{r', r}(\mathbf{q}), \tag{28} \]

where \( G_{r, r'}(\mathbf{q}) = \langle r' \rangle e^{-i\mathbf{q}\mathbf{r}} |r \rangle \) and \( M^{\lambda}_{J_z, J_z'}(\mathbf{q}) \) gathers the \( \mathbf{q} \)-dependent factor of the \( \mathcal{H}_{\text{h-ph}} \) term coupling \( J_z \) and \( J'_z \). \( G_{r, r'}(\mathbf{q}) \) is calculated analytically using iterative procedures as described in [40]. The sum over \( \mathbf{q} \) in equation (27) is then carried out using numerical integration. To this
Table 1. Parameters used in the numerical calculations for InAs (left column) and GaAs (right column) QDs. GaAs parameters are used for the matrix in both cases. $e^-$, $h^+$ and ph stand for electron, hole and phonon.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>InAs</th>
<th>GaAs</th>
</tr>
</thead>
<tbody>
<tr>
<td>$e^-$ mass ($m_0$)</td>
<td>$m_e$</td>
<td>0.026</td>
<td>0.067</td>
</tr>
<tr>
<td>$h^+$ Luttinger param.</td>
<td>$\gamma_1$</td>
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<td>6.98</td>
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<tr>
<td>$h^+$ Luttinger param.</td>
<td>$\gamma_2$</td>
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<td>2.06</td>
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<tr>
<td>$h^+$ Luttinger param.</td>
<td>$\gamma_3$</td>
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<td>2.93</td>
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<td>$e^-$ deformation pot. (eV)</td>
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<td>$-7.17$</td>
</tr>
<tr>
<td>$h^+$ deformation pot. (eV)</td>
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<td>1.16</td>
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<tr>
<td>$h^+$ deformation pot. (eV)</td>
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<td>$-2.0$</td>
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<td>$h^+$ deformation pot. (eV)</td>
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<td>$-0.0034$</td>
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<td>4720</td>
</tr>
<tr>
<td>Transversal ph speed (m s$^{-1}$)</td>
<td>$c_t$</td>
<td>3340</td>
<td>3340</td>
</tr>
<tr>
<td>Crystal density (kg m$^{-3}$)</td>
<td>$\rho$</td>
<td>5310</td>
<td>5310</td>
</tr>
<tr>
<td>Piezoelectric coeff. (V cm$^{-1}$)</td>
<td>$h_{14}$</td>
<td>$3.5 \times 10^6$</td>
<td>$1.45 \times 10^7$</td>
</tr>
</tbody>
</table>

end, it is convenient to use spherical coordinates, as the modulus $q$ is fixed by the resonance condition and we are left with a 2D integral.

Calculations are carried out for InAs QDs embedded in a GaAs matrix. When differences are expected, we also calculate GaAs QD embedded in an Al$_x$Ga$_{1-x}$As matrix. Table 1 summarizes the parameters we use. The parameters correspond to the QD material, except for the crystal density and velocity of sound, which correspond to the matrix material because we assume bulk phonons (for simplicity, for Al$_x$Ga$_{1-x}$As we assume $x \to 0$ and use GaAs phonon parameters). For the dielectric constant, an average value of $\epsilon_r = 12.9$ is taken all over the structure. The basis used to solve Hamiltonian (1) contains all the hh oscillator eigenstates with the quantum numbers $v_x, v_y < 13$ and $v_z < 9$.

3. Numerical results and discussion

We shall start this section by describing the dependence of the hole spin lifetime on the QD geometry and the spin splitting magnitude (section 3.1). The influence of each parameter can be understood by analyzing the spin admixture mechanisms, as we show in section 3.2. Next, in section 3.3, we study the effect of the ground state changing from mainly hh character, which is the case addressed in most previous studies, to mainly lh character. This transition is observed in QDs with large aspect ratio [42–45]. Last, in section 3.4, we compare the role of deformation potential and piezoelectric potential interactions in determining the efficiency of hole–phonon coupling.

Figure 2. Hole (red solid line) and electron (blue dotted line) spin lifetime in InAs QDs, as a function of the lateral confinement (a), vertical confinement (b)–(c) and spin splitting energy (d). The inset in (b) compares $T_{1h}$ for InAs and GaAs. (a) $\hbar \omega_z = 50 \text{meV}$, $\hbar \omega_z = 179 \text{meV}$, $\Delta = 0.4 \text{meV}$. (b) $\hbar \omega_z = 20 \text{meV}$, $\hbar \omega_z = 23.2 \text{meV}$, $\Delta = 0.4 \text{meV}$. (c) $\hbar \omega_z = 5 \text{meV}$, $\hbar \omega_z = 5.8 \text{meV}$, $\Delta = 0.4 \text{meV}$. (d) $\hbar \omega_z = 20 \text{meV}$ and $\hbar \omega_z = 50 \text{meV}$ (red solid line); $\hbar \omega_z = 23.2 \text{meV}$ and $\hbar \omega_z = 179 \text{meV}$ (blue dotted line); $\hbar \omega_z = 40 \text{meV}$ and $\hbar \omega_z = 5 \text{meV}$ (red dashed line).

3.1. Geometry and spin splitting dependence

Solid lines in figure 2 show the hole spin lifetime as a function of the QD geometry and the spin splitting energy of InAs QDs. For comparison, we also plot the electron spin lifetimes (dotted lines). The latter have been calculated using the same formalism as for holes but adapted for single-band conduction electrons [40]. Both electrons and holes are assumed to be confined in the same QD, hence they share the same force constants but have different confinement frequencies. One can see immediately in the figure that the behavior of holes differs drastically from the well-known case of electrons. Below we summarize the influence of each parameter.

Figure 2(a) shows the spin lifetime dependence on the lateral confinement in QDs with strong vertical confinement. $T_{1h}$ increases monotonically with $\omega_z$. This is because, as we approach the spherical confinement regime ($\omega_z = \omega_z$), the Dresselhaus SOI of electrons is gradually suppressed [40]. No such trend is however observed for holes, as $H_{BIA}$ does not cancel out even if the confinement is isotropic. As a matter of fact, $T_{1h}$ shows an evident non-monotonic behavior, with a minimum at $\omega_z = 28 \text{meV}$ and increasing away from it. It is worth noting that a previous study by Woods et al [29] predicted $T_{1h}$ to decrease with the QD diameter, while a similar study by Lu et al [30] for somewhat larger QDs predicted the opposite trend. Figure 2(a)
shows that both predictions are conciliable, corresponding to the right and left sides of the $T^h_1$ minimum, respectively. The origin of the different trends will be discussed in section 3.2.

Figures 2(b) and (c) show the spin lifetime dependence on the vertical confinement in QDs with moderately strong and weak lateral confinement, respectively. These confinement strengths roughly correspond to self-assembled (panel (b)) [46, 47] and electrostatic (panel (c)) [48] QDs. As can be seen in figure 2(b), electrons and holes have opposite behaviors. $T^s_1$ now decreases with $\omega^e_\perp$ because the structure is becoming flatter (less isotropic). Instead, the behavior of $T^h_1$ is similar to that observed for varying lateral confinement, with a shallow minimum at $\omega^h_{\perp} = 14$ meV. Previous studies have shown that $T^h_1$ increases with the vertical confinement [30]. This is consistent with the right side of the $T^h_1$ minimum in figure 2(b), but here we show that the opposite trend is also possible if the QD aspect ratio is large enough (left side of the minimum).

Figure 2(c) illustrates the case in which the lateral confinement is weak. The minimum of $T^h_1$ is now shifted toward very small $h\omega^h_{\perp}$ values so that only the right side behavior is seen in the range under study. Interestingly, here $T^h_1$ shows a clear saturation with increasing vertical confinement ($h\omega^h_{\perp} > 40$ meV), which has not been noticed before [30]. As we show in section 3.2, this saturation reflects the fact that $H_{BIA}$ has replaced hh–lh mixing as the main source of SOI.

The inset in figure 2(b) compares $T^h_1$ in InAs and GaAs QDs with moderate lateral confinement. As can be observed, the spin lifetime in InAs QDs is longer than that in GaAs QDs when $\omega^h_{\perp} > \omega^h_{\perp}$, which is, e.g. the case of self-assembled QDs. This is an unexpected result because in bulk the valence band SOI of InAs is stronger than that of GaAs (compare the split-off band splittings [41] or the $\gamma_2$ and $\gamma_3$ coefficients appearing in $R$ and $S$ terms of equation (4), which couple hh to lh). The underlying reason is that in confined structures, the cubic Dresselhaus SOI becomes important and it is stronger for GaAs (see $b_{\text{III}}$ in table 1).

Figure 2(d) shows the spin-flip lifetime of electrons and holes in a self-assembled-like QD as a function of the spin splitting energy. For electrons, $T^s_1$ is largely determined by the efficiency of the carrier–phonon coupling [40]. It is short when the phonon wavelength is of the same order as the carrier wavefunction extension, but it increases for large (small) $\Delta$ values because the phonon wavelength becomes too short (long), as $q \approx \Delta / \hbar c$. The same happens for holes (notice that $G_{r,r}(q)$ in equation (28) vanishes in the limits of large and small phonon wavevector). However, figure 2(d) shows that $T^h_1$ is only sensitive to $\Delta$ for small splittings, but then it reaches a plateau where $T^h_1 \sim \mu s$. The different behavior of holes and electrons is due to the different effective mass along the growth direction, $m_{\perp}$. For $h\omega_\perp = 50$ meV, the characteristic length of the oscillator states in the growth direction, $l_{\perp} = \sqrt{\hbar / m_{\perp} \omega_\perp}$, is $l_{\perp} = 4.5$ nm for electrons and $l_{\perp} = 2.4$ nm for holes, i.e. the hole confinement is stronger. As a result, larger values of $\Delta$ than those used in figure 2(d) are required for $T^h_1$ to increase.

Experiments with excitons in self-assembled InGaAs QDs have revealed a negligible influence of e–h interactions on $T^h_1$ [24]. Our results would be consistent with such an observation in the regime of large $\Delta$. For self-assembled QDs, however, $\Delta \leq 0.5$ meV. Thus, the insensitivity noted in the experiment is inconsistent with the single-phonon processes we consider in figure 2. This suggests that two-phonon processes dominate in these systems [15, 32]. On the other hand, experiments with colloidal nanorods have shown a sizable increase of $T^h_1$ when changing from type-I to type-II confinement, which modulates the e–h exchange energy from typical colloidal structure values (few meV) to type-II system values (fractions of meV) [36]. We have run simulations for a nanorod-like geometry (red dashed line in figure 2(d))
and find that the weak vertical confinement renders $T^h_1$ sensitive to $\Delta$ for all the range under study, in agreement with the experiment. This indicates that the weak vertical confinement of rods renders single-phonon processes efficient.

To close this section, we notice that previous theoretical studies with simpler models had predicted that hole spin lifetimes can exceed those of electrons in very flat QDs $^{31}$. Figure 2 confirms that this could actually be achieved in gated structures, where lateral confinement is very weak (see the crossing between $T^e_1$ and $T^h_1$ in panel (c)). However, for typical self-assembled InAs QDs, $T^e_1$ is about one order of magnitude longer than $T^h_1$ (see panel (b) for large $\hbar \omega_z$). This is consistent with experimental measurements by Heiss et al $^{26}$.

### 3.2. Mechanism of spin admixture

Spin admixture is a requirement for spin-flip transitions to take place $^{50}$. In this section, we compare the spin admixture resulting from all the SOI terms affecting the hole ground state. As we shall see, once the dominant SOI mechanism is determined, one can rationalize the geometry dependence of $T^h_1$ described in the previous section. For convenience of the analysis, in what follows we plot and discuss relaxation rates, $1/T^h_1$. Furthermore, we shall often drop the hh superscript when referring to the confinement frequency, $\omega^h \perp$ or $\omega^h z$.

Figure 3(a) shows the relaxation rate for the InAs QDs of figure 2(a), but now obtained by calculating hole states with the diagonal terms of $H_\text{L}$ plus different SOI terms: off-diagonal $H_\text{L}$ terms (hereafter hh–lh coupling), full Dresselhaus Hamiltonian ($H_{\text{BIA}}$), hh–lh coupling plus linear-in-$k$ term ($H_\text{L} + H_{C_k}$) and hh–lh coupling plus the dominant cubic-in-$k$ Dresselhaus term

2 The same conclusions, albeit with somewhat lower $T^e_1$, are obtained using larger electron masses to account, e.g. for Ga diffusion into the InAs QD.
shows that most of the relaxation rate coming from $H_L$. This is inspite of the fact that it is a source of direct admixture between hh states with opposite spin, with no participation of lh states. For this reason, it had been proposed as the dominant SOI term in flat QDs with weak lateral confinement \cite{35}. Instead, figure 3 shows that most of the $H_{BIA}$ contribution comes from the cubic-in-$k$ $b_{41}$ term (see green dotted line).

This term relies on intermediate lh states in order to mix hh $J_z = +3/2$ and $-3/2$ components (see equation (A.3) in the appendix), which implies that a simultaneous description of hh and lh states is necessary for realistic modeling.

By comparing the total relaxation rate coming from $H_L + H_{BIA}$ with that coming from $H_L$ and $H_{BIA}$, it is clear that the total rate is not just the sum of the two independent mechanisms. For example, at $\hbar \omega_{\perp} = 30$ meV, adding $H_{BIA}$ to $H_L$ enhances $1/T_1^h$ about an order of magnitude, even though the contribution coming from $H_{BIA}$ alone is about 100 times smaller than that coming from $H_L$. This can be understood by means of a perturbative reasoning: neither $H_L$, equation (4), nor $H_{b_{41}}$—the most relevant term of $H_{BIA}$—, contribute to hh–lh mixing at first order. $H_L$ contributes at second order, due to terms involving non-zero products like $H_L(1, 2) H_L(2, 4)$, while $H_{b_{41}}$ does not. It contributes at third order, due to non-zero products like $H_{b_{41}}(1, 2) H_{b_{41}}(2, 3) H_{b_{41}}(3, 4)$. However, the sum of the two Hamiltonians allows $H_{b_{41}}$ to contribute at second order. Thus, the effect of $H_{b_{41}}$ is clearly non-additive because it is enhanced by $H_L$. Simultaneously accounting for both SOI terms is then required for quantitative estimates.

For a qualitative understanding of the geometry dependence of $1/T_1^h$, we rewrite the hole states, equation (14), as $|\Psi_m^h\rangle = \sum J_z c_{J_z}^m |\psi_{J_z}^m\rangle |3/2, J_z\rangle$, where $|\psi_{J_z}^m\rangle = \sum r e^{i m r} r \rangle$ is the envelope function associated with the periodic Bloch function $|3/2, J_z\rangle$. If we restrict to the diagonal components of $H_{h-\text{ph}}$, the matrix element determining the relaxation rate becomes

$$
\langle \Psi_m^h | H_{h-\text{ph}}^{eq} | \Psi_n^h \rangle \approx \sum J_z \langle c_{J_z}^m | H_{h-\text{ph}}^{eq} | c_{J_z}^n \rangle \langle \phi_{J_z}^m | H_{h-\text{ph}}^{eq} | \phi_{J_z}^n \rangle.
$$

When the QD is oblated or spherical, the low-energy states are essentially hh states. Thus, the initial (final) state of the spin transition is mostly a pure spin-up (spin-down) hh state. Considering that $|c_{3/2}^f| \gg |c_{3/2}^i| (|c_{-3/2}^f| \gg |c_{-3/2}^i|)$, one can obtain an approximate expression

$$
\frac{1}{T_1^h} \propto \left| \langle \Psi_i^h | H_{h-\text{ph}}^{eq} | \Psi_f^h \rangle \right|^2 \propto |c_{3/2}^f|^2 \left| \langle \phi_{3/2}^f | H_{h-\text{ph}}^{eq} | \phi_{3/2}^i \rangle \right|^2.
$$

In other words, the relaxation rate is proportional to the spin admixture of the ground state through the squared coefficient of the minor hh component (here spin-up, $J_z = 3/2$), and proportional to the efficiency of the hole–phonon coupling through the rightmost matrix element.

The geometry dependence of $1/T_1^h$ for a given SOI mechanism simply reflects the spin admixture variation. This can be seen in figure 3(b), which shows that, for $H_L$ and $H_{BIA}$, $|c_{3/2}^f|^2$ has the same qualitative dependence on the geometry as the corresponding $1/T_1^h$ values in figure 3(a). This allows us to interpret the observed maximum as a function of $\omega_{\perp}$. 

Figure 4. Hole spin relaxation as a function of the vertical confinement. Different SOI terms are considered. (a) Strong lateral confinement, $\hbar \omega_{\perp} = 20$ meV. (b) Weak lateral confinement, $\hbar \omega_{\perp} = 5$ meV. The inset in (a) is the corresponding result for GaAs. The system is the same as that of figures 2(b) and (c).

For $H_L$, the spin admixture between the hh spin-up and spin-down components takes place necessarily through the intermediate lh components (see equation (4)). The weight of the minor hh component is then related to the strength of the off-diagonal terms of $H_L$ ($R$ and $S$) and to the energy splitting between the hh and the lh states. For small $\omega_{\perp}$ values, the main effect of increasing the lateral confinement is to enhance the coupling terms, which are proportional to $k_x$ and $k_y$ (see equations (7) and (8)). As a result, the weight of the minor hh component $|c_{3/2}^f|$ increases, hence $1/T_{1h}$ increases. For larger $\omega_{\perp}$ values, however, when the lateral and vertical confinements become comparable, the main effect of increasing the lateral confinement is to bring lh states closer to hh ones [44, 45]. When this happens, the lh states stop acting as intermediate states for the admixture between hh components and start participating in the admixture themselves. This is at the expense of reducing the weight of the minor hh component, hence $1/T_{1h}$ decreases.

Next, we analyze the mechanisms involved in the spin relaxation with varying vertical confinement. Figure 4 shows $1/T_{1h}$ for the same systems as in figures 2(b) and (c), but calculating the hole states with the diagonal terms of $H_L$ plus hh–lh coupling (red solid line) or full $H_{BIA}$ Hamiltonian (blue dashed line). Panel (a) corresponds to strong lateral confinement. The total rate has a maximum at $\hbar \omega_z = 14$ meV, whose origin is analogous to that described above for varying lateral confinement. Beyond the maximum, the total rate ($H_L + H_{BIA}$) decreases with the vertical confinement strength, in agreement with previous studies [30]. However, we also find that the decrease eventually saturates. This is evident for
InAs QDs with weak lateral confinement, as shown in figure 4(b), or GaAs QDs with strong lateral confinement, as in figure 4(a) inset. The origin of this saturation is the contribution of $H_{BIA}$, which provides a lower bound to $1/T_1^h$. In particular, $H_{b41}$ introduces off-diagonal coupling terms which are quadratic in $k_z$ (see operator $L_{41}$ in the appendix), instead of the linear $k_z$ terms of $H_L$ (see $S$ operator in equation (4)). Since the uncoupled hh and lh energies are also quadratic in $k_z$, a perturbational analysis easily shows that the two contributions compensate each other. For strong $\omega_z$, when lateral confinement is negligible, the cancelation is exact and the relaxation rate does not depend on the vertical confinement.

The magnitude of the spin splitting also influences the dominant mechanism of spin admixture. This is illustrated in figure 5, which considers a self-assembled InAs QD with varying spin splitting energy. For large $\Delta$, $H_L$ has a dominant contribution to the relaxation rate, but $H_{BIA}$ becomes equally important for small enough $\Delta$. The relative enhancement of the role of $H_{BIA}$ originates in its zero-field spin splitting, which leads to larger energy difference between the Kramers pair ($\Delta E_{fi}$) than with $\Delta$ alone, as shown in the figure inset. When $\Delta \rightarrow 0$ and the phonon wavelength increases beyond the QD size, the extra energy coming from the zero-field spin splitting of $H_{BIA}$ provides a significant contribution to preserve the hole–phonon coupling efficiency.

3.3. Light hole spin relaxation

When the aspect ratio increases and the QD shape becomes prolate, the hole ground state evolves from the eminent hh character discussed so far to lh character, as noted, e.g. in colloidal nanorods [42–45]. Here we investigate how the change of the ground state affects the relaxation between the two highest hole states. Figure 6(a) shows the hole energy levels in a QD with $\hbar\omega_\perp = 40$ meV as a function of $\hbar\omega_z$. In the limit of strong and weak vertical confinement, the two highest states are essentially hh and lh doublets, $J_z = \pm 3/2$ and $\pm 1/2$, respectively. In the intermediate regime, $\hbar\omega_z = 9–17$ meV, the two doublets cross. This gives rise to pronounced changes in the relaxation rate, as shown in figure 6(b).
Figure 6. Hole energy levels (a) and spin relaxation rate (b) around the hh–lh crossing region. $\hbar \omega_\perp = 40$ meV and $\Delta = 2$ meV. In (a), solid and dashed lines are used for states with dominant lh and hh character, respectively. The dotted line gives the energy splitting. Shades are used to distinguish the regions with different kinds of states involved in the transition.

The changes can be understood as follows. In region I of the figure, the two highest states are the hh doublet. The relaxation is between states with opposite spin and roughly constant energy splitting (see $\Delta E_{fi}$, dotted line in figure 6(a)). At $\hbar \omega_\perp = 17$ meV, when we enter region II, the excited hh state crosses with the first lh state. Now the relaxation is between an lh ($J_z = -1/2$) and an hh ($J_z = -3/2$). Since lh have mixed spin-up and spin-down projections, there is no need for spin flip. Then, the $s^4$ terms of the strain Hamiltonian, $H_{dp}$, provide direct coupling with hh and the resulting transition is much faster. This explains the abrupt increase of $1/T_1^h$. However, the energy splitting between the hh and lh becomes smaller with decreasing $\omega_\perp$ because of their different masses. As a result, at $\hbar \omega_\perp = 11$ meV, the lh replaces the hh as the ground state. Near the degeneracy point, $\hbar \omega_\perp = 11.2$ meV, $\Delta E_{fi}$ is so small that hole–phonon coupling becomes inefficient and the relaxation is strongly suppressed, but it increases again in region III for the same reasons as in region II. Finally, at $\hbar \omega_\perp = 9$ meV, the excited lh state crosses with the highest hh state. We thus enter region IV, where the transition takes place between two lh states with orthogonal Bloch functions, $|3/2, \pm 1/2 \rangle$. $J_z$ admixture mechanisms are necessary and the relaxation becomes slow. As a matter of fact, the spin relaxation timescale for transitions between lh states is similar to that between hh states, in spite of the fact that their Bloch functions contain mixed spins.

3.4. Mechanism of hole–phonon coupling

Electron–acoustic phonon coupling in QDs is known to occur mainly through deformation potential interaction when the energy splitting is large ($\Delta E_{fi} > 0.1$ meV) and through
Figure 7. Hole spin relaxation as a function of the spin splitting energy. (a) Transition between hh states in a QD with $\hbar\omega_\perp = 20$ and $\hbar\omega_z = 50$ meV. (b) Transition between lh states in a QD with $\hbar\omega_\perp = 40$ and $\hbar\omega_z = 5$ meV.

piezoelectric potential when it is small [51]. In principle, for holes, the situation may differ because the deformation potential Hamiltonian, equation (17), is formally different from that of electrons. To investigate this point, in this section we compare the role of the two kinds of carrier–phonon coupling mechanism for holes subject to varying effective magnetic fields.

Figure 7(a) shows the spin relaxation rate in an oblate (quasi-2D) QD, where the highest states are hh, while figure 7(b) shows that in a prolate (quasi-1D) QD, where the highest states are lh. For the spin transition within the hh doublet, panel (a), the behavior is analogous to that of electrons. Deformation potential interaction (dotted line) provides the largest contribution to $1/T_1^h$ except for very small $\Delta$, when piezoelectric interaction (dashed-dotted line) takes over. This is because all the terms of $\mathcal{H}_{dp}$ are proportional to the phonon momentum $\sqrt{q}$ (see equations (24)–(26)) while the piezoelectric potential is proportional to $1/\sqrt{q}$ (see equation (23)). With decreasing $\Delta$, both mechanisms become inefficient, because for long phonon wavelength $e^{iqr} \approx 1$. Then, in equation (28), the matrix element $G_{r,r'}(q) \approx (r'|r)$, i.e. it tends to the overlap between the envelope components of the initial and final states. For hh, these components have different symmetries, so the coupling vanishes. For example, in axially symmetric structures, the $J_z = +3/2$ component of the initial state has a well-defined azimuthal angular momentum $m_z = 0$, which couples through the $s^z$ operator of $\mathcal{H}_{dp}$ with the $J_z = +1/2$ component of the final state, for which $m_z = -2$.

The situation for lh is quite different. As shown in figure 7(b), now deformation potential interaction is the dominant relaxation channel even for small $\Delta$. The underlying reason is that, in contrast to the hh case, the off-diagonal terms of $\mathcal{H}_{dp}$ couple envelope components with the same symmetry. For example, the $J_z = +3/2$ and 1/2 components of the initial and final state

3 In axially symmetric systems, the envelope functions of the Luttinger spinor have well-defined $z$-component of the orbital angular momentum, $m_z = F_z - J_z$. In oblate QDs, the highest doublet has $F_z = \pm 3/2$, and in prolate QDs it has $F_z = \pm 1/2$. See e.g. [45].

now have both $m_z = -1$, and hence are not orthogonal. As a consequence, $G_{r,r'}(q)$ does not vanish when $q \to 0$.

4. Summary

We have investigated hole spin relaxation in InAs and GaAs QDs using 3D four-band $k \cdot p$ Hamiltonians. We have shown that the hole spin lifetime has a non-monotonic dependence on the lateral and vertical confinement strength. This is due to the interplay between the energy splitting of hh and lh, which is set by their different masses, and the anisotropic hh–lh coupling terms. The resulting geometry dependence of hole spin relaxation is qualitatively different from that of electrons.

hh–lh coupling and Dresselhaus SOI have been found to have comparable contributions to the spin admixture of hole states in self-assembled QDs, with the former becoming dominant for prolate structures, such as nanorods, and the latter for strongly oblate ones, such as gated QDs. The cubic-in-$k$ Dresselhaus term leads to an upper bound of $T^h_{1}$ with increasing vertical confinement, which is missed when considering hh–lh coupling only.

We have also investigated the spin relaxation involving states with dominant lh character. Transitions between lh and hh states are very fast because lh have strong spin admixture. Instead, transitions between lh states with different Bloch angular momentum $J_z$ are as slow as the transitions between hh states with opposite spin. There is, however, a difference in the dominating hole–phonon interaction mechanism. At small energy splittings, the relaxation is mainly due to deformation potential interaction, unlike for hh transitions, where it is due to piezoelectric interaction.

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Appendix. Dresselhaus Hamiltonian for holes

In this appendix, we write the explicit matrix forms of the different $H_{\text{BIA}}$ terms in Cartesian coordinates. Separating the different invariants in equation (9) we obtain

$$H_{\text{BIA}} = H_{C_k} + H_{b_{41}} + H_{b_{42}} + H_{b_{51}} + H_{b_{52}},$$

(A.1)

where

$$H_{C_k} = C_k \begin{pmatrix} 0 & -\frac{k_z}{2} & k_z & -\frac{\sqrt{3}}{2} k_z \\ -\frac{k_z}{2} & 0 & \frac{\sqrt{3}}{2} k_z & -k_z \\ k_z & \frac{\sqrt{3}}{2} k_z & 0 & -\frac{k_z}{2} \\ -\frac{\sqrt{3}}{2} k_z & -k_z & -\frac{k_z}{2} & 0 \end{pmatrix}$$

(A.2)
with $k_\pm = k_x \pm i k_y$:

$$H_{b41} = b_{41} \begin{bmatrix} \frac{3}{2} P_{41} & \frac{\sqrt{3}}{2} L_{41} & 0 & 0 \\ \frac{\sqrt{3}}{2} L_{41} & \frac{3}{2} P_{41} & \frac{3}{2} L_{41} & 0 \\ 0 & \frac{3}{2} L_{41} & \frac{1}{2} P_{41} & \frac{\sqrt{3}}{2} L_{41} \\ 0 & 0 & \frac{\sqrt{3}}{2} L_{41} & -\frac{3}{2} P_{41} \end{bmatrix},$$  \hspace{1cm} (A.3)

where $P_{41} = (k_x^2 - k_y^2) k_z$ and $L_{41} = i k_- k_x k_y - k_+ k_z^2$.

$$H_{b42} = b_{42} \begin{bmatrix} \frac{27}{8} P_{41} & \frac{7\sqrt{3}}{8} L_{41} & 0 & -\frac{3}{4} L_{42} \\ \frac{7\sqrt{3}}{8} L_{41} & \frac{3}{8} P_{41} & \frac{3}{2} L_{41} & 0 \\ 0 & \frac{3}{2} L_{41} & -\frac{1}{8} P_{41} & \frac{7\sqrt{3}}{8} L_{41} \\ -\frac{3}{4} L_{42} & 0 & \frac{7\sqrt{3}}{8} L_{41} & -\frac{27}{8} P_{41} \end{bmatrix},$$  \hspace{1cm} (A.4)

where $L_{42} = i k_+ k_x k_y + k_- k_z^2$.

$$H_{b51} = b_{51} \begin{bmatrix} 0 & -\frac{\sqrt{3}}{4} K_+ & \frac{\sqrt{3}}{4} K_z & -\frac{3}{4} K_- \\ -\frac{\sqrt{3}}{4} K_+ & 0 & \frac{3}{4} K_+ & -\frac{\sqrt{3}}{4} K_z \\ \frac{3}{4} K_z & \frac{3}{4} K_- & 0 & -\frac{\sqrt{3}}{4} K_+ \\ -\frac{3}{4} K_+ & -\frac{\sqrt{3}}{4} K_z & -\frac{\sqrt{3}}{4} K_- & 0 \end{bmatrix},$$  \hspace{1cm} (A.5)

where $K_+ = K_x + i K_y$, $K_- = K_x - i K_y$, $K_x = k_x (k_x^2 + k_z^2)$, $K_y = k_y (k_y^2 + k_z^2)$ and $K_z = k_z (k_x^2 + k_y^2)$.

$$H_{b52} = b_{52} \begin{bmatrix} 0 & -\frac{\sqrt{3}}{4} M_+ & \frac{\sqrt{3}}{2} k_z^3 & -\frac{3}{4} M_- \\ -\frac{\sqrt{3}}{4} M_- & 0 & \frac{3}{4} M_+ & -\frac{\sqrt{3}}{2} k_z^3 \\ \frac{\sqrt{3}}{2} k_z^3 & \frac{3}{4} M_- & 0 & -\frac{\sqrt{3}}{4} M_+ \\ -\frac{3}{4} M_+ & -\frac{\sqrt{3}}{2} k_z^3 & -\frac{\sqrt{3}}{4} M_- & 0 \end{bmatrix},$$  \hspace{1cm} (A.6)

where $M_+ = k_x^3 + i k_y^3$ and $M_- = k_x^3 - i k_y^3$.

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