Spin–orbit-induced hole spin relaxation in a quantum dot molecule: the effect of s-p coupling

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Abstract
We study the effect of the coupling between the hole s shell of one quantum dot and the p shell in the other dot forming a vertically stacked self-assembled quantum dot molecule on the spin relaxation between the sublevels of the hole s state. Using an effective model that captures the spin–orbit effects in the p shell irrespective of their origin, we show that the strong spin mixing in the p shell can be transferred to the s shell of the other dot, leading to enhanced spin relaxation in a certain energy range around the s-p resonance if the dots are misaligned and the magnetic field is tilted from the sample plane.

Keywords: quantum dot molecule, spin–orbit coupling, spin relaxation, phonons

(Some figures may appear in colour only in the online journal)
by the presence of Dresselhaus SO interaction, with the spin life times around a $\mu$s at the magnetic field of 2 T, roughly constant across the resonance. In a subsequent study [40] the spin-flip rate was also studied for an electron, using a more realistic model including strain in a QDM built of two nominally identical QDs.

The SO coupling that underlies the hole spin relaxation manifests itself in the band structure of a semiconductor in a rather complicated way that is captured e.g. by various inter-band terms of the $k \cdot p$ model [41, 42]. For a carrier confined in a QD the SO coupling is particularly pronounced in the hole $p$ shell, where it dominates over the Zeeman terms, leading to reordering of the $p$-shell hole states [43]. It turns out that the results of exact $k \cdot p$ calculations are reproduced by an effective model with a few fitting parameters determined by comparison to $k \cdot p$ results or to experiment [38, 43, 44]. When supplemented with a simple model of carrier-phonon couplings, such a model yields a simple, yet reasonably accurate, description of charge and spin relaxation in a QD, accounting for all the SO effects, including those induced by strain [28]. Similar effective models were used in the description of tunneling between QDs in a QDM [45].

In this paper we study hole spin relaxation between Zeeman sublevels of the ground ($s$-shell) state in one of the QDs forming a self-assembled QDM, resulting from tunnel coupling to the $p$-shell states of the other QD. We use the effective model for SO couplings in the valence band extended by tunneling terms along with the standard model of carrier-phonon coupling. We show that the strong SO coupling in the hole $p$ shell can be transferred via tunnel coupling to the $s$-shell of the other QD in an axially misaligned QDM leading to considerably enhanced spin relaxation in a tilted magnetic field. The hole spin flip rate becomes particularly large in the vicinity of the $s$-$p$ resonance but the effect persists also away from the resonance, where the states are nearly completely localized in one of the QDs and nearly ideally spin-polarized.

The organization of this paper is as follows. In section 2 we describe the system and introduce the effective Hamiltonian. In section 3 we present the results for the relaxation rates as a function of electric and magnetic field. Section 4 concludes the paper.

2. Model

2.1. Description of the system

The structure consists of two self-assembled lens-shaped QDs grown in the [001] direction. The dots are stacked vertically with the inter-dot distance $D_z$. The cylindrical symmetry of the system is broken by a small relative in-plane displacement $D_x$ (see figure 1).

The wave function of the hole confined in a QD is the solution to the 2-dimensional quantum harmonic oscillator problem, i.e. the Fock–Darwin state [46, 47], with the principal quantum number $n$ and the angular quantum number $\ell$ representing the envelope angular momentum. The wave functions are localized in the plane on a distance $l_{\perp}$ and in the growth direction on a distance $l_{\parallel}$. The model includes the $s$-shell states ($n = 0, m = 0$) in the QD1, and the $p$-shell states ($n = 0, m = \pm 1$) in QD2. We take into account the two values of the band angular momentum (‘spin’) of a heavy hole, represented by $\sigma = \uparrow, \downarrow$. The basis states are then $\{|m\sigma⟩\} = \{|0 \uparrow⟩, |0 \downarrow⟩, |1 \uparrow⟩, |1 \downarrow⟩, |−1 \uparrow⟩, |−1 \downarrow⟩\}$. The off-axis misalignment leads to inter-dot $s$-$p$ coupling because of the symmetry breaking [48]. The system is placed in an arbitrarily oriented magnetic field and interacts with phonons.

The system is described by an effective Hamiltonian similar to that introduced in [43] but generalized to the QDM structure,

$$H = H_0 + H_{\text{int}},$$

where $H_0$ is the effective Hamiltonian of the holes and $H_{\text{int}}$ stands for hole interaction with the phonon bath.

The hole Hamiltonian $H_0$ contains various components accounting for the QD energy levels, external fields, tunneling and SO coupling [43],

$$H_0 = \sum_{\alpha=1}^{7} H_0^{(\alpha)}.$$  

These components of $H_0$ can be written in the form of a tensor product corresponding to the decomposition $|m⟩ \otimes |\sigma⟩$, where the first component refers to the spatial degrees of freedom and the second one to the spin. The first part,

$$H_{0}^{(1)} = \text{diag}(\Delta E_l + eFzD_z, 0, 0) \otimes \mathbb{I}_2$$

describes the bare energy $\Delta E_l$ of the $s$-shell states in the QD1 (defined with respect to the $p$-shell energy in the QD2), as well as the effect of the electric field $F_z$ applied along the stacking direction. Here $\mathbb{I}_2$ is the unit operator on the spin subsystem. Next,

$$H_{0}^{(2)} = \sum_{i=x,y,z} \frac{1}{2} g_i |m\mu_B B| |0⟩⟨0| \otimes \sigma_i.$$
and
\[ H_0^{(3)} = \sum_{i=x,y,z} \frac{1}{2} \delta_{p,i} \mu B_i (|+1\rangle\langle+1| + |-1\rangle\langle-1|) \otimes \sigma_i \]
account for Zeeman splittings in the s- and p-shells, respectively. Anisotropic Landé factors for s- and p-shell states are denoted by \( g_s \) and \( g_p \). \( \sigma_i \) stand for Pauli matrices, \( B_i \) are the components of the magnetic field and \( \mu_B \) is the Bohr magneton. The next term,
\[ H_0^{(4)} = V_s (|+1\rangle\langle-1| + |-1\rangle\langle+1|) \otimes I_2, \]
describes the influence of an anisotropic QD elongation. Next,
\[ H_0^{(5)} = t_p \langle 0|\langle+1| + |0\rangle\langle-1| + \text{h.c.} \rangle \otimes I_2 \]
is the inter-dot s-p coupling of strength \( t_p \). The part
\[ H_0^{(6)} = V_{SO} (|+1\rangle\langle+1| - |-1\rangle\langle-1|) \otimes \sigma_z \]
describes the SO coupling in the p shell. Finally,
\[ H_0^{(7)} = WB_s (|+1\rangle\langle-1| - |-1\rangle\langle+1|) \otimes \mathbb{I}_2 \]
accounts for the magnetic effect on the envelope angular momentum.

In our model we neglect the SO coupling between the s shells of the two QDs. Such a coupling, induced by SO interaction, is in principle non-zero in a misaligned structure and leads to spin-flip admixture from the s state of QD2 to the s state of QD1. However, we expect the resulting corrections to be very small since the s shell of QD2 is separated from the energy sector of interest by several meVs, which suppresses perturbative admixtures due to the weak SO couplings. We neglect also the effect of the vector potential on the exact shape of the wave functions and energies of the state, assuming that the essential physical properties of the system are contained in the constant fitting parameters. This approximation is justified in a self-assembled system with strongly confined states, as confirmed by the agreement between the effective Hamiltonian and k p results [43].

The hole-phonon interaction Hamiltonian has the form
\[ H_{int} = \sum_{m,m'} |m\rangle\langle m'| \sum_{\lambda=0} \mathcal{F}_{mm'}^{(\lambda)} (\mathbf{k}) \left( b^\dagger_{\mathbf{k}} + b_{-\mathbf{k}}^\dagger \right) \otimes \mathbb{I}_2. \]  
(2)

Here the phonon annihilation and creation operators for wave vector \( \mathbf{k} \) and polarization \( \lambda \) are denoted by \( b^\dagger_{\mathbf{k}} \) and \( b_{-\mathbf{k}}^\dagger \). \( \mathcal{F}_{mm'}^{(\lambda)} (\mathbf{k}) \) are hole-phonon coupling constants,
\[ \mathcal{F}_{mm'}^{(\lambda)} (\mathbf{k}) = -i \int d^3r \psi_m^\dagger (\mathbf{r}) \mathcal{E}^\lambda (\mathbf{r}, \mathbf{k}) \psi_{m'} (\mathbf{r}), \]  
(3)
where \( \rho \) is the crystal density, \( c_\lambda \) is the sound velocity, \( d_p \) stands for piezoelectric constant, \( \varepsilon_r \) denotes relative electric permittivity, and \( \mathcal{E}_{\mathbf{k}p} \) is the form-factor
\[ \mathcal{E}_{\mathbf{k}p} (\mathbf{k}) = \int d^3r \psi_p^\dagger (\mathbf{r}) e^{i \mathbf{p} \cdot \mathbf{r}} \psi_p (\mathbf{r}). \]  
(4)

\[ \begin{array}{|c|c|c|c|}
\hline
\text{Property} & \varepsilon_s & 13.2 \\
\hline
\text{Piezoelectric constant} & d & 0.16 \text{ C m}^{-2} \\
\text{Longitudinal sound speed} & c_l & 5600 \text{ m s}^{-1} \\
\text{Transverse sound speed} & c_t & 2800 \text{ m s}^{-1} \\
\text{Density} & \rho_c & 5360 \text{ kg m}^{-3} \\
\text{Hole wave function widths} & \text{in-plane} & l_L & 5.0 \text{ nm} \\
\text{z direction} & l_z & 1.5 \text{ nm} \\
\text{Dots stacking} & \text{vertical} & D_v & 7 \text{ nm} \\
\text{horizontal} & D_h & 1 \text{ nm} \\
\text{Landé g-factor} & \text{For s-shell in-plane} & g_s^{(s)} & -0.1 \\
& \text{z direction} & g_s^{(z)} & -5.51 \\
& \text{For p-shell in-plane} & g_p^{(s)} & 0.05 \\
& \text{z direction} & g_p^{(z)} & 2.62 \\
\text{Anisotropy factor} & V_4 & 1.54 \text{ meV} \\
\text{SO coupling} & V_{SO} & -9.79 \text{ meV} \\
\text{Envelope angular} & \text{momentum coefficient} & W & -0.532 \text{ meV T}^{-1} \\
\text{momentum} & \text{Inter-dot s-p coupling} & t_p & 0.1 \text{ meV} \\
& \text{Intra-dot s-p energy} & \Delta E_2 & -40 \text{ meV} \\
& \text{separation} & \Delta E_1 & -7 \text{ meV} \\
\hline
\end{array} \]

Table 1. Parameters.

with \( \Psi_i (\mathbf{r}) \) denoting the envelope wave function of the hole. In the QDM system, neglecting the overlap between wave functions of basis states located in different QDS, the form-factors have the form
\[ \mathcal{F}_{00} (\mathbf{k}) = e^{-\frac{1}{2}(\mathbf{k}_x^2 + \mathbf{k}_y^2)} e^{i \mathbf{k}_z \mathbf{D}_z + i \mathbf{k}_x \mathbf{D}_x}. \]
and
\[ \mathcal{F}_{mm'} (\mathbf{k}) = f_{mm'} (k_L \mathbf{L}) e^{-\frac{1}{2}(\mathbf{k}_x^2 + \mathbf{k}_y^2)} e^{i (m - m') \varphi}, \]
for \( m, m' = \pm 1 \), where \( k_L \) is the in-plane component of the wave vector and \( f_{-1,-1} (x) = f_{+1,+1} (x) = 1 + f_{+1,-1} (x) = 1 + f_{-1,+1} (x) = 1 - x^2 \).

The geometric factor \( M_\lambda \) in equation (3) is defined as
\[ M_\lambda (\tilde{k}) = \sum \left| k_{\tilde{k} \tilde{\lambda}} + k_{\tilde{\lambda} \tilde{k}} \right|^2 \]
where \( \tilde{k} \) is the unit polarization vector for the mode \((\lambda, \tilde{k})\). Note that our model does not include the usual SO terms responsible for spin mixing and spin relaxation within the s shell, so that it allows us to single out the effect of s-p coupling and p-shell SO structure.

The values of the parameters can be found in table 1. The value of the hole s-p tunnel coupling, which is a crucial parameter in the presented calculations, is assumed to be one order of magnitude lower than the analogous value calculated for the electron [49]. This seems to be a safe estimate in view of

\[ \cdot \cdot \cdot \]
of the $k\cdot p$ results that yield similar orders of magnitude for the electron [50] and hole [48] s-shell tunnel couplings.

### 2.2. Spin relaxation rate

We find the spin-flip rate between the s-shell Zeeman sublevels of the ground hole states in the QD1 using the Fermi golden rule. We diagonalize effective Hamiltonian to find the eigenstates $|\alpha\rangle = \sum_j c_{\alpha j}|j\rangle$ (6) and the transition frequencies $\omega_{\alpha\beta} = (E_i - E_f)/\hbar$, where initial and final states are the nominally spin-up and spin-down s-shell states in the top QD. We define the spectral densities (in terms of the basis states)

$$ R_{\alpha\beta\gamma\delta}(\omega) = \sum_{j,j',m'\gamma} \int d\Omega_{j} \frac{1}{c_{\lambda}} M_{\lambda}^{j}(\hat{k}) F_{j\gamma'}(\frac{\omega}{c_{\lambda}}, \hat{k}) F_{m'\delta}(\frac{\omega}{c_{\lambda}}, \hat{k}), $$

where $l_1, l_2$ denote the longitudinal and transverse phonon branches. Using the Fermi golden rule and writing the system eigenstates as a superposition of the basis states according to equation (6), we obtain the formula for the spin relaxation rate, which is a combination of the spectral densities taken at the transition frequency

$$ \gamma_{\alpha\rightarrow\alpha'} = \sum_{j,j',m'} c_{\alpha j'}^* c_{\alpha j} c_{\alpha' j} c_{\alpha' j'} R_{j\gamma j' m'}(\omega_{\gamma}). $$

### 3. Results

The rate of phonon-induced spin relaxation in the QD1 caused by the coupling to the p-shell of the QD2 was obtained by numerical implementation of the model discussed in section 2. Figure 2(a) shows the diagram of energy levels as a function of electric field for the range of field magnitudes around the resonances between the s shell of QD1 and the p shell of QD2. The results were obtained for the magnetic field of 5 T, tilted by...
15° off the plane. The states localized in QD1, distinguishable by their evolution in the electric field, are marked with colors. The two branches of the $s$ states are the Zeeman sublevels with respect to the spin quantization axis determined by the field orientation. The two $p$-shell states are the lower half of the four $p$-shell states split by the SO interaction [43], corresponding to anti-parallel orientation of orbital and band angular momenta (the upper two states lie above the axis range).

Figure 2(b) shows the transition rate between the $s$-shell Zeeman sublevels of QD1 (orange to green in figure 2(a)) calculated from equation (7). The rate peaks at the resonances with the $p$-shell states, where the life time of the upper state is reduced to about 100 ps. In order to interpret this, we present the spin polarization of the two states in figure 2(c) (with color coding referring to figure 2(a)) and their localization in figure 2(d) (the former is defined as the average value of $I_z \otimes \sigma_z$ and the latter as the average value of $\langle |0\rangle |0\rangle - |1\rangle |1\rangle + |+1\rangle |-1\rangle - |-1\rangle |+1\rangle \otimes I_z$). At the resonance, one of the Zeeman branches in QD1 mixes with a state with the opposite spin from the $p$ shell of QD2. This results in a reduction of the spin polarization of this state from $\pm 1$ to 0, as can be seen in figure 2(c) and delocalization of the state between the QDs, visible in figure 2(d). As a result, the spin-flip time becomes comparable to the charge relaxation time (tens of picoseconds).

This resonant enhancement is, however, constrained to a very narrow parameter range, since the resonance between states with opposite spins is very narrow (see the insets in figure 2(a)). Away from the resonance the rate decreases, as expected for an effect that relies on the admixture of $p$ states. Still, in a range of a few meV from the resonance, the spin-flip rates remain relatively high and correspond to spin life times in the $\mu$s range, which are comparable to or shorter than those observed for intrinsic spin relaxation in single QDs [23, 32]. This shows that in a certain range of energies around the resonance with the $p$ shell of the QD2 the hole spin flip in the ground state of the QD1 is dominated by the inter-QD $s$-$p$ coupling and $p$-shell SO effects.

A remarkable feature in the field dependence of the relaxation rate shown in figure 2(b) is the pronounced minimum located nearly exactly at the point where the resonant part of the $s$-$p$ spectrum (excluding the upper part of the $p$ shell) is symmetric. This suppression of spin relaxation results from destructive interference of spin-flip admixtures to the $s$-shell states at this spectral symmetry point.

An additional effect, that is not included in our model, is the dependence of the hole $g$ factor on the wave function symmetry. Out of the two states in each resonance, one is symmetric and the other one is anti-symmetric, leading to different contribution of the barrier $g$ factor, which typically differs considerably from that of the QDs [38]. This effect would affect the widths and the exact positions of the resonances and might perturb the symmetry of the electric field dependence shown in figure 2(b), slightly shifting the location of the minimum of the relaxation rate. We do not expect this effect to considerably modify our results.

In figure 3 we show the system spectrum and relaxation rates as a function of magnetic field for a fixed electric field of 0.12 mV nm$^{-1}$ (marked by a vertical line in figure 2). The rates in figure 3(b) are plotted for three different tilt angles (marked by vertical lines in figure 4) and the spectrum in figure 3(a) is shown for one angle $\pi/12$, corresponding to the green line in figure 3(b). The rates show a peak at the resonance, the origin of which is the same as in the previous discussion. Away from the resonances, at lower field magnitudes, the rate grows as $B^5$.

In figure 4 the eigenstate diagram and the spin relaxation rates are presented as a function of the magnetic field orientation for different magnetic field magnitudes. The rates in figure 4(b) are plotted for various magnitudes of the magnetic field and the spectrum in figure 4(a) corresponds to $B = 5$ T (yellow line in figure 4(b)). Apart from the resonant enhancements, one can see considerable reduction of the rate at angles 0 and $\pi/2$ (exact Voigt and Faraday geometry, respectively). The former is due to the strong anisotropy of the hole $g$ factor and nearly vanishing $s$-shell Zeeman splitting in the Voigt geometry, which suppresses the phonon-assisted relaxation due to low phonon spectral density at low frequencies. In the Faraday geometry the $z$ projection of the spin is strictly conserved in our model, as follows directly from the form of the Hamiltonian, precluding spin relaxation at this orientation of the field.
4. Conclusions

We have studied phonon-assisted hole-spin relaxation rate between Zeeman sublevels in a QD forming part of a QDM. We have calculated the energy states using an effective Hamiltonian that accounts for the spin–orbit coupling of the other QD in the QDM and tunnel coupling between the s and p shells of the two dots. We have shown that the inter-QD s–p coupling induced by QD misalignment combined with strong SO effects in the p shell leads to enhanced spin relaxation in the vicinity of the s–p resonance, with the exception of exact Voigt and Faraday geometries, when the rates becomes small or vanish, respectively. We have also predicted suppression of the spin relaxation due the s–p coupling near the point where the spectrum of resonant states becomes symmetric.

Typical spin relaxation times resulting from the s–p coupling mechanism are on the order of µs even away from the resonance, when the states are nearly completely localized and spin-polarized. The mechanism studied here can therefore dominate over other known spin-flip processes in a certain energy interval around the s–p resonance. Such relaxation rates would limit the feasibility of spin control via electric dipole spin resonance (EDSR) [51–53]. Indeed, the predicted EDSR spin rotation times are on the order of 10 ns in various weakly confined structures [54, 55] and may be expected to be 2–3 orders of magnitude longer for holes in self-assembled structures due to stronger confinement and higher hole effective mass [54], which may be partly compensated by applying stronger fields. On the other hand, optical schemes that are available in self-assembled structures [7–10] allow one to control spins on the time scale of a single optical pulse, which is in picosecond range. The spin relaxation times discussed in this paper are still 5–6 orders of magnitude longer than these control times, except at exact resonance.

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Figure 4. The eigenstate diagram (a) and the hole spin relaxation rate (b) as a function of the orientation of magnetic field (angle with respect to the in-plane direction) for magnetic field magnitudes as shown. The electric field has the magnitude of 0.12 mV nm⁻¹.

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