Exciton-spin relaxation in quantum dots due to spin-orbit interaction

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We present theoretical results for the spin relaxation of exciton-bound electrons and holes in weakly confining quantum dots. The relaxation is driven by the spin-orbit interaction in the conduction band and the linear in the momentum term in the valence band, respectively. The relaxation occurs between the optically active (bright) and inactive (dark) exciton states due to acoustic-phonon-assisted spin flips. The exchange splitting between the bright and dark states acts as a constant external magnetic field. A sequential flip of the (exciton-bound) electron and hole spins results in the spin-flip transition between the bright exciton states (i.e., an exciton-spin relaxation). We find that the spin relaxation time for an exciton-bound electron is several orders of magnitude faster than for a single electron. The resulting exciton spin-relaxation time is also several orders of magnitude faster than the one in small dots which is driven by the electron hole exchange interaction. We obtain the dependence of the exciton-spin relaxation time on dot size and temperature.

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In the last decade the theoretical study of spin relaxation in semiconductor quantum dots (QDs) has been intense, but the relevant mechanisms are still being debated.[1] It is believed that for electrons the dominant mechanism is a hyperfine-interaction process[2] which gives relaxation times on the order of 10^{-6} s. Phonon-assisted electron-spin flips (between Zeeman sublevels) mediated by the spin-orbit (SO) coupling[3] give relaxation times on the order of 10^{-4} s. For holes, the SO coupling-induced mechanism must be dominant, since the hyperfine interaction induced mechanism is here irrelevant. Hole-spin relaxation times on the order of 10^{-8} s (i.e., a few orders of magnitude faster than for the electrons) were recently calculated in Ref. 4. For excitons, a combined effect of the electron-hole exchange interaction and the short-range deformation potential interaction was recognized as a possible spin-flip mechanism.[5] Such a mechanism is relevant for InAs QDs and gives relaxation times on the order of 10^{-5} s.

The term "quantum dot" is actually used for a variety of different objects. InAs QDs formed in the Stransky-Krastanow growth mode (see, e.g., Ref. 6) show a strong carrier confinement in all three directions of space.[7] In contrast, interfacial QDs in GaAs/AlGaAs quantum wells or quantum islands in CdTe/ZnTe or CdSe/ZnSe quantum wells have a strong confinement perpendicular to the quantum well plane and typically only a weak lateral confinement. All these materials have a zinc-blend crystal structure without an inversion center and the electron and hole states approach the two-dimensional case. In such a scenario the SO interaction must come into play as an important source of spin flips. In the presence of the SO coupling, the projection of the total momentum is conserved, whereas the states of different angular momentum are mixed. This mixing increases, evidently, for large QDs with small lateral quantization energies.

In the case of excitons, the SO coupling could be responsible for the relaxation process in which an independent spin-flip of the exciton-bound electrons or holes occurs. Such a channel of exciton-spin relaxation was studied for quantum wells,[9] but to our knowledge has not been considered to date for QDs. But there is experimental evidence that such a mechanism might be important since a strong enhancement of the spin relaxation efficiency was observed for InGaAs quantum disks[10] and CdTe QDs (Ref. 11) when the lateral dot size was increased.

In this paper we consider phonon-assisted spin-flip processes of exciton-bound electrons and holes mediated by the SO coupling in flat QDs with weak lateral confinement. The exciton ground states in flat QDs are associated with the heavy-hole valence band and are split into the $|\pm 1\rangle$ bright (optical active) and the $|\pm 2\rangle$ dark (optical inactive) states by the short-range exchange interaction.[6,12] Typically, this (singlet-triplet) splitting is of the order of $\Delta_{st} \sim 100-200$ μeV.[13] When an exciton-bound electron or hole flips its spin, the transitions occur between the bright and dark exciton states, see Fig. 1. Such a transition between the exciton (integer) spin states requires no external magnetic field unlike the case of the SO-induced transitions between the (half-integer) spin states of a free electron or hole.[3] A sequential flip of the (exciton-bound) electron and hole spins results in the spin-flip transition between the $|\pm 1\rangle$ bright states, see Fig. 1. The spin relaxation rate for such an indirect channel (in the sense that transitions between the bright states involve an intermediate dark state) is determined by the slowest process of the sequence.[14]

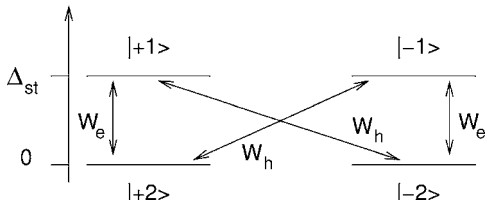

FIG. 1. The heavy-hole exciton spin states and exciton-bound single-particle spin-flip transitions with rates W_e and W_h for electrons and holes, respectively.

For flat QDs grown along the crystal cubic axis [001](z), the SO interaction for electrons (in a Γ_6 conduction band) is given by[15]

$$H_{so}^{e} = \beta_e(\sigma_y k_y - \sigma_x k_x), \tag{1}$$

where $\vec{k}_\perp = \{k_x, k_y\}$ is the (in-plane) momentum operator, $\vec{\sigma}$ are the Pauli matrices, and the strength of a SO coupling β_e depends on the material and the height of the QD, $\beta_e \sim l_z^{-2}$. Equation (1) arises from the so called k^3 spin-orbit term in the Γ_6 conduction band of semiconductor compounds without an inversion center that was first obtained by Dresselhaus.[16]

For holes (in the Γ_8 valence band), the $\vec{k}$-linear term is of a relativistic origin and leads to the bulk Hamiltonian (the notation follows the one by Bir and Pikus)[17]

$$H_v^{lin} = \frac{4}{\sqrt{3}} k_0 \sum_{i=x,y,z} k_i V_i, \tag{2}$$

where k_0 is a material constant, $V_i = J_i(J_{i+1}^2 - J_{i+2}^2)$, and $\vec{J}$ is the operator of the (hole) total angular momentum ($J=3/2$). In the angular momentum basis $|\frac{3}{2},\frac{3}{2}\rangle$, $|\frac{3}{2},\frac{1}{2}\rangle$, $|\frac{3}{2},-\frac{1}{2}\rangle$, and $|\frac{3}{2},-\frac{3}{2}\rangle$, the matrix of the Hamiltonian Eq. (2) is given by

$$H_v^{lin} = k_0 \begin{pmatrix} 0 & k_- & -2k_z & \sqrt{3}k_+ \\ k_+ & 0 & -\sqrt{3}k_- & 2k_z \\ -2k_z & -\sqrt{3}k_+ & 0 & k_- \\ \sqrt{3}k_- & 2k_z & k_+ & 0 \end{pmatrix}, \tag{3}$$

where $k_\pm = k_y \pm i k_x$. For flat QDs with a large splitting between the heavy-hole (hh) and the light-hole (lh) bands, the hh-lh mixing can be neglected.[18] Equation (3) reduces, hence, to two equivalent matrices in the heavy-hole and the light-hole basis, respectively. As a result, the $\vec{k}$ linear interaction for the heavy-holes is given by

$$H_{hh} = \beta_h(\sigma_x k_y - \sigma_y k_x), \tag{4}$$

where $\beta_h = \sqrt{3}k_0$. This interaction was considered as a possible source of the hole spin relaxation in quantum wells.[19]

We concentrate now on the spin relaxation in flat QDs which are symmetrical in the lateral plane. Usually the bright exciton states lie above the dark states, so that the spin relaxation from the bright states to the dark ones leads to a thermal equilibration and is accompanied by an emission of acoustic phonons. An opposite transition from a dark state to a bright state requires phonon absorption and is suppressed at temperatures with $kT < \Delta_{st}$ (typically less than a few degrees Kelvin).

We calculate the (exciton-bound) carrier-phonon scattering rate from Fermi's golden rule. For transitions from the upper (bright) states to the lower (dark) states the relaxation rate is given by

$$\frac{1}{\tau_{1\to 2}^{(e,h)}} = \frac{2\pi}{\hbar} \sum_{\vec{q}} |M_{\vec{q}}^{e,h}|^2 [N_{\vec{q}} + 1] \delta(\hbar\omega_{\vec{q}} - \Delta_{st}), \tag{5}$$

where $\tau_{1\to 2}^{e}$ ($\tau_{1\to 2}^{h}$) determines the relaxation due to the spin-flip of the electron (hole) and the notation 1 (2) stands for the bright (dark) state. $N_{\vec{q}}$ is the phonon occupation factor and $M_{\vec{q}}^{e,h} = \langle \Psi_b(\vec{r}_e, \vec{r}_h) | H_{\vec{q}}^{e,h} | \Psi_d(\vec{r}_e, \vec{r}_h) \rangle$ is the corresponding matrix element with $H_{\vec{q}}^{e,h}$ the electron (hole)–phonon interaction, and $|\Psi_{b,d}(\vec{r}_e, \vec{r}_h)\rangle$ is the initial (final) exciton state. Below we consider the piezoelectric carrier-phonon interaction which is known to be most effective in polar crystals for small energy transfers.[20] For the $(2 \to 1)$ transitions, the phonon occupation factor $(N_{\vec{q}}+1)$ in Eq. (5) must be replaced by $N_{\vec{q}}$.

There are no phonon-assisted spin-flip transitions between exciton states in the absence of the SO interaction, since the electron (hole)–phonon interaction is independent of the carrier spin. The SO coupling, however, mixes the states of different spin and angular momentum leading to a nonzero value of $M_{\vec{q}}^{e,h}$. The SO-induced splitting $\sim \beta_{e,h}/a$ (a is the size of the QD) is, typically, smaller than the (lateral) quantization energy,[3,18] so that the SO interaction can be treated as a perturbation in the calculation of the exciton states. As a result, for transitions between the $|+1\rangle$ ($|-1\rangle$) and $|+2\rangle$ ($|-2\rangle$) exciton states (flip of the electron spin), one obtains in first order of the SO-coupling strength β_e,

$$M_{\vec{q}}^{e} = \sum_k \frac{\langle \Psi_b | H_{\vec{q}}^{e} | \Psi_k \rangle \langle \Psi_k | H_{so}^{e} | \Psi_d \rangle}{E_b - \Delta_{st} - E_k} + \sum_k \frac{\langle \Psi_b | H_{so}^{e} | \Psi_k \rangle \langle \Psi_k | H_{\vec{q}} | \Psi_d \rangle}{E_b - E_k}. \tag{6}$$

In Eq. (6) the summation occurs over the unperturbed excited exciton states and $E_b(E_k)$ is the exciton (unperturbed) energy for a ground (excited) state. For transitions between the $|-1\rangle$ ($|+1\rangle$) and $|+2\rangle$ ($|-2\rangle$) exciton states (flip of the hole spin), the SO interaction H_{so}^{e} in Eq. (6) must be replaced by H_{hh} and $H_{\vec{q}}^{e}$ by $H_{\vec{q}}^{h}$.

Assuming that the confinement along the growth (z) direction is stronger than both the lateral quantum dot and the Coulomb potentials, the envelope for the electron-hole pair wave function can be written approximately as[21]

$$\Psi(\vec{r}_e, \vec{r}_h) = \psi(\vec{\rho}_e, \vec{\rho}_h) \phi_e(z_e) \phi_h(z_h), \tag{7}$$

where $\phi_{e,h}(z_{e,h})$ are the electron and hole envelope functions in the z direction, and $\psi(\vec{\rho}_e, \vec{\rho}_h)$ is an in-plane wave function of the exciton in a lateral confinement potential. For QDs in a weak confinement regime, the relative electron-hole motion ($\vec{\rho} = \vec{\rho}_e - \vec{\rho}_h$) and the motion of the exciton center of mass [$\vec{R} = (X, Y)$] are separated, $\psi(\vec{\rho}_e, \vec{\rho}_h) = \phi(\vec{\rho}) F(\vec{R})$. Only the motion of the exciton center of mass is affected by the lateral potential which is assumed to be a harmonic potential $V(\vec{R}) = M\Omega^2 R^2/2$, where $M = m_e + m_{hh\perp}$ is the translational mass of the hh exciton.

For small splittings Δ_{st} (i.e., $\Delta_{st} < \hbar s/a$, s is the sound velocity and a is the lateral size of a QD), long-wave phonons contribute mainly to the relaxation rate in Eq. (5), so that one obtains

$$\frac{1}{\tau_{1\to 2}^{(e,h)}} \approx w_0 \left(\frac{m_{e,hh\perp}}{M} \right)^2 \beta_{e,h}^2 (N+1), \tag{8}$$

$$w_0 = \frac{(eh_{14})^2}{35\pi\rho\hbar^3 s^5}\frac{\Delta_{st}}{\hbar}\left(\frac{\Delta_{st}}{\hbar\Omega}\right)^2\left(\frac{\Delta_{st}}{\hbar\Omega+\Delta_{st}}\right)^2, \tag{9}$$

where $N=(e^{\Delta_{st}/kT}-1)^{-1}$ is the phonon occupation factor, ρ is the crystal mass density, $s^{-5}=s_l^{-5}+4s_t^{-5}/3$, and s_l, s_t are the (longitudinal and transverse) sound velocities. The quantity h_{14} determines the component of the piezotensor β_{ijk} which for crystals of interest without an inversion center (class T_d) has only one independent component $\beta_{ijk}=h_{14}$, $i\neq j\neq k$. The energy $\hbar\Omega$ is the lateral quantization energy, which can be determined from the experiment or estimated from the size of the wave function $F(\vec{R})$ using $\hbar\Omega=\hbar^2/2Ma^2$ (see Appendix A).

Equations (8) and (9) determine the SO-driven spin-relaxation rates of the exciton-bound electron and hole in QDs. Results (8) and (9) are valid under the following conditions: (i) The QD's height l_z is smaller than the lateral size a and the exciton Bohr radius a_B ($l_z \ll \{a, a_B\}$), i.e., there is a large hh-lh splitting and the excitons can be considered to be two dimensional; (ii) the exciton quantization energy $\hbar\Omega$ is larger than the SO-induced splittings β_e/a and β_h/a in the conduction and valence band, respectively ($\hbar\Omega > \beta_{e,h}/a$), i.e., the SO coupling acts on the exciton states as a perturbation; (iii) the QD's lateral size a is larger than the exciton Bohr radius a_B ($a > a_B$), i.e., there is only weak confinement in the lateral plane of the QD.[7,22]

The SO-driven spin-relaxation, as follows from Eqs. (8) and (9), dominates in large QDs (small lateral quantization energy $\hbar\Omega$) consisting of materials with significant but not too large exchange interaction (splitting Δ_{st}) and a strong SO coupling (large parameters β_e and β_h). The spin-relaxation rate increases with temperature due to the phonon occupation factor N.

Note also that according to Eq. (8), the spin-relaxation times for the (exciton-bound) electron and hole are related via the masses and the spin-orbit parameters,

$$\tau^{(e)} = \tau^{(h)}\left(\frac{m_{hh\perp}}{m_e}\right)^2\left(\frac{\beta_h}{\beta_e}\right)^2. \tag{10}$$

In semiconductor compounds of interest the (in-plane) hole-mass and the electron-mass are, usually of the same order, $m_{hh\perp}\sim m_e$.[23] On the other hand, the parameters β_h and γ_c ($\beta_e=\gamma_c\langle k_z^2\rangle$ with γ_c the strength of the Dresselhaus coupling) vary strongly in different materials,[21,24] besides the parameter β_e depends on l_z. Hence it depends on the material whether the electron or the hole spin relaxation is strongest. For GaAs QDs, for example, the (exciton-bound) electron-spin relaxation dominates since $\tau^{(e)}\sim 0.2\tau^{(h)}$ (at $l_z=4$ nm).[25] A *single carrier* spin relaxation between the Zeeman sublevels in GaAs-based QDs, on the contrary, is much less efficient for the electrons than for the holes due to a large Zeeman splitting in a valence band.[4]

In order to estimate the relaxation rate resulting from the mechanism proposed here, we choose a set of parameters which are relevant for InGaAs QDs. Namely, we use $|\beta_h|=11$ meV Å,[24,25] $\Delta_{st}=0.28$ meV,[26] and $s=2.6\times10^5$ cm/sec, $\rho=5$ g/cm^3, $M=0.15m_0$, and $eh_{14}=1.2\times10^7$ eV/cm (Ref. 23). We obtain a hole spin relaxation time $\tau_{1\to2}^{(h)}\simeq 1.2$ ns for a lateral dot diameter $a=30$ nm and a temperature $T=4$ K.

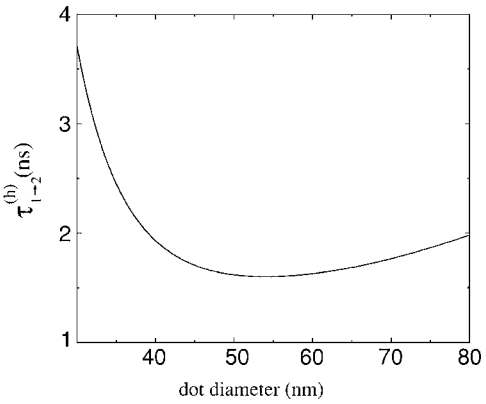

FIG. 2. The lateral size variation of the exciton-bound hole-spin relaxation time in flat InGaAs QDs at 4 K. For the parameters see the text.

In Fig. 2 we show the dependence of the hole-spin relaxation time $\tau_{1\to2}^{(h)}$ on the lateral QD size at 4 K as calculated from the detailed evaluation of Eqs. (5) and (6) (see Appendix B). The dependence on the lateral size comes from $\hbar\Omega \sim a^{-2}$. From Eq. (8) we see that the relaxation rate is then $W_h\sim a^8$. For large $a > \hbar s/\Delta_{st}$, however, the contribution of the short-wave phonons becomes increasingly important. The corresponding envelope integrals result in a decrease of the relaxation rate in large dots with $a \geqslant 50$ nm (see Fig. 2).

The temperature dependence of the spin relaxation is related to the phonon occupation factor N in Eq. (8). In Fig. 3 is shown that the relaxation time $\tau_{1\to2}$ is almost temperature independent up to $T\sim 1$ K and decreases as $\sim 1/T$ for higher T. The $(2\to1)$ transitions are suppressed up to $T\sim 2$ K and $\tau_{1\to2}^{(h)}$ is larger than $\tau_{2\to1}^{(e)}$ for higher temperatures. Hence, the spin flip transitions between bright exciton states, as well as transitions from dark states to bright states, are suppressed at low temperatures $T \leqslant 2$ K.

The values and trends obtained here for the spin relaxation time are in reasonable agreement with experimental results. A spin relaxation time of 0.9 ns was measured in

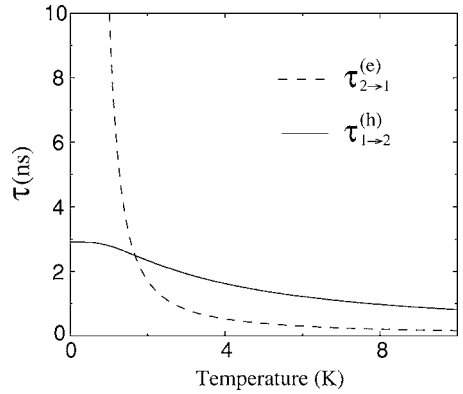

FIG. 3. The relaxation times in a flat InGaAs quantum dot with a diameter of 50 nm as a function of temperature. For the parameters see the text.

InGaAs quantum disks with about 30 nm lateral diameter at 4 K and an enhancement of the relaxation efficiency was found in larger disks and at higher temperatures.[10] We expect therefore that the SO coupling-induced relaxation considered here is responsible for the observed indirect exciton spin relaxation in weakly confining (In)GaAs QDs. Further, a strong enhancement of the spin-relaxation efficiency with an increase of the QD size was observed in experiments on CdTe QDs.[11] Finally, a fast (about 200 ps) transfer from dark exciton states to bright states was observed recently in InP QDs by two-photon absorption experiments for temperatures $T \geqslant 10$ K.[27] Such a transfer takes place by a spin-flip process of the exciton-bound electron or hole, as discussed above. For InP dots with a diameter of about 20 nm, our estimates[28] give a spin-flip time of about 500 ps (at $T=20$ K).[29]

The presented results are obtained for the piezoelectric type of the carrier-phonon interaction. The relative magnitudes of the relaxation times τ_{def} and τ_{piezo} due to the deformation potential and the piezoelectric type of the carrier-phonon interaction, respectively, are given by[20]

$$\tau_{def}^{(e,h)} \sim \tau_{piezo}^{(e,h)} \left(\frac{eh_{14}}{D^{(e,h)}} \right)^2 \left(\frac{\hbar s}{\Delta_{st}} \right)^2, \tag{11}$$

where D^e (D^h) is the deformation potential constant in a conduction (valence) band. Using typical parameters: $D \simeq 5$ eV, $s=2.4\times10^5$ cm/sec, (Ref. 23), and $\Delta_{st}=0.2$ meV (Ref. 6), from Eq. (11) we obtain $\tau_{def} \sim 3\tau_{piezo}$. Hence for realistic parameters, the piezoelectric type of the carrier-phonon interaction is indeed dominant.

For QDs with an asymmetric confining potential in the growth direction, the Rashba spin-orbit coupling presents an additional source for spin relaxation.[32] For the case of the exciton-bound electron, the spin-orbit constant β_e in Eqs. (8) and (9) is replaced then by $(\beta_e+\beta_R)$ (with β_R the Rashba coupling parameter in a conduction band). The Rashba coupling in the valence band Γ_8, however, is cubic in the hole momentum and contributes to the hh splitting only when the hh-lh mixing is taken into account.[33,34]

In summary, we have presented a microscopic theory of the spin-orbit-driven exciton-bound electron and hole-spin relaxation due to piezoelectric carrier-phonon interaction in flat quantum dots with weak lateral confinement. For such a process, no external magnetic field is required. The efficiency of the spin relaxation increases with a lateral size of the QDs as well as with temperature. We find that the relaxation time for the exciton-bound electron and hole spins is on the order of 10^{-9} s for dots $\gtrsim$40 nm. This is several orders of magnitude faster than the spin-relaxation time for a single electron,[3] and comparable to the relaxation time of a hole spin[4] in similar dots with an applied moderate magnetic field. This is also much faster than the relaxation of an exciton spin in strongly confining dots, which is controlled by the exchange interaction.[5] We believe that the theory presented here is able to explain the experimental data on the exciton-spin relaxation, e.g., in InGaAs quantum disks[10] and CdTe QDs.[11]

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APPENDIX A: EXCITON QUANTIZATION ENERGY

For QDs confined by a harmonic potential, the QD's size can be estimated by the (squared) coordinate expectation value, so that the lateral size of a (symmetrical) QD is given by

$$a^2 \approx \langle X^2 \rangle = \langle Y^2 \rangle = \int_{-\infty}^{+\infty} x^2 F(x) dx, \tag{A1}$$

where X and Y are the coordinates of the exciton center of mass. With the (ground state) wave function

$$F(x) = \left(\frac{M\Omega}{\pi\hbar} \right)^{1/4} e^{-(M\Omega/2\hbar)x^2} \tag{A2}$$

one obtains

$$a^2 \approx \frac{\hbar}{2M\Omega}, \tag{A3}$$

so that the quantization energy is $\hbar\Omega \approx \hbar^2/2Ma^2$, as used in the main text.

APPENDIX B: CONTRIBUTION OF THE SHORT-WAVE PHONONS

Equations (8) and (9) determine the spin-relaxation rates of the exciton-bound electron and hole for relaxation processes which are assisted by long-wave phonons. The short-wave phonons contribution results in an additional factor which depends, in particular, on the QD's size. For the case of $l_z \ll a$ one obtains

$$\frac{1}{\tau_{1\to2}^{(e,h)}} = w_0 \left(\frac{m_{e,hh\perp}}{M} \right)^2 \beta_{e,h}^2 (N+1) I_{e,h}, \tag{B1}$$

where w_0 is given by Eq. (9) and

$$I_{e,h} = \frac{35}{32} \sum_{i=l,t} \left(\frac{s}{s_i} \right)^5 \int_0^\pi e^{-a^2\Delta_{st}^2 \sin^2\vartheta/\hbar^2 s_i^2} f_{e,h}^i(\vartheta) g_i(\vartheta) \sin^3\vartheta \, d\vartheta, \tag{B2}$$

$$f_{e,h}^i(\vartheta) = \left[1 + \left(\frac{m_{e,hh\perp}}{M} \right)^2 a_B^2 \frac{(\Delta_{st}/\hbar s_i)^2}{16} \sin^2\vartheta \right]^{-3}, \tag{B3}$$

$$g_t = 8 \left[\cos^2\vartheta \sin^2\vartheta + \frac{1}{8} \sin^4\vartheta (1 - 9\cos^2\vartheta) \right],$$

$$g_l = 9 \cos^2\vartheta \sin^4\vartheta. \tag{B4}$$

In Eq. (B2) the contributions of the longitudinal and transverse phonon modes are taken into account. For small exchange splitting $\Delta_{st} \ll \hbar s_i/a$ it follows that $I_{e,h} \sim 1$ and Eq. (B1) reduces to Eq. (8).

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[29] In Ref. 27 a saturation of T dependence of the spin-flip time was found which is not typical for the spin-relaxation processes in QDs. Note that a specific feature of the InP QDs is a shallow potential well for the holes (Ref. 30). If a hole relaxes its spin in the wetting layer, the saturation of the T dependence of the spin relaxation can be explained in the framework of the (two-dimensional) D'yakonov-Perel'-like mechanism (Ref. 31).

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