Light-hole exciton spin relaxation in quantum dots

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The phonon-induced flip of the exciton spin in single flat semiconductor quantum dots with a light-hole exciton ground state is studied. The corresponding quartet, split by the exchange interaction, consists of three bright states and a dark state located energetically below the bright exciton. The two in-plane polarized bright states contribute to single-phonon transitions to the dark state and also to the upper bright state polarized in the z growth direction of the dot. For these processes, the presented analytical results are calculated for the relaxation driven by the spin-orbit interaction in the conduction and the light-hole valence subbands. The estimated spin-relaxation times at low temperature are (at least) one order of magnitude lower than the bright exciton lifetime. Two other possible transitions, within the in-plane polarized doublet and between the z-polarized bright and dark states as well, proceed via intermediate states with a contribution from two acoustic phonons. These processes are strongly suppressed at low temperature, whereas they appear to be of the same intensity as single-phonon transitions at high enough temperatures.

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I. INTRODUCTION

Epitaxially grown structures of semiconductor self-assembled quantum dots (QDs), which are considered as candidate building blocks for quantum technologies, have attracted great interest during the last decade. In particular, epitaxial QDs can act as triggered sources of single [1] and entangled photons [2]. Extensive experimental studies have identified the main features of the exciton fine structure in self-organized QDs. It is commonly accepted that the ground states of the heavy-hole (hh) and light-hole (lh) excitons in typical QDs are well separated, as a consequence of (intrinsic) strain and confinement in the growth direction of a QD, and the hh exciton has the lower energy. Systems with a light-hole ground state, known recently, refer to nanostructures with a large height:base ratio resembling vertical nanorods [3–5]. Quite recently in Ref. [6] the creation of an excitonic ground state of the lh type by applying elastic stress to an initially unstrained QD with a hh exciton ground state was reported. The obtained self-assembled GaAs QDs are characterized by high optical quality and the corresponding microphotoluminescence spectra, which show three orthogonally polarized bright optical transitions, are fully consistent with the behavior of a lh exciton. Obviously, the spin effects in such systems can be of interest.

In this paper we discuss the spin relaxation within the fine structure of an excitonic ground state of the light-hole type in flat (with a small height:base ratio) QDs similar to those studied in Ref. [6]. To our knowledge, this problem has not been considered to date, in contrast to the conventional (unperturbed) QDs with the hh exciton ground state. Although both cases are similar, for the spin relaxation within the ground state of the lh exciton, one can expect some important peculiarities due to the different "spin" structure in comparison with the hh exciton. Our main aim here is to discuss these features. Note that, for the heavy-hole exciton, two main microscopic mechanisms of spin relaxation—a deformation-induced exchange interaction [7,8] and spin-orbit–phonon coupling [9,10]—have been recognized. The first transitions have been found to dominate in strongly confining QDs, while the second transitions are relevant for large QDs with closely spaced levels [11]. Evidently, the same processes can operate the spin-flip transitions within the ground state of the lh exciton. For the GaAs QDs from Ref. [6], which are characterized by large enough (in-plane) sizes, the spin-orbit–phonon coupling is probably most likely and below we restrict our model calculations of the lh exciton spin lifetimes to the spin-orbit (SO) interaction in the conduction and the light-hole valence subbands. Note that the SO-induced transitions between the exciton (integer) spin states require no external magnetic field, unlike the case of transitions between the (half-integer) spin states of a free electron or hole [13].

The electron and the light hole have the same (z-projection) angular momentum $|j_z| = |s_z| = \frac{1}{2}$ and form therefore the pair states $|s_z, j_z\rangle$ of the total momentum $F_z = 0, \pm 1$. The resulting four basis states are mixed and, consequently, split by the electron-hole exchange interaction [14], as is shown in Fig. 1. Below in numerical calculations we treat the exchange-induced splittings as parameters with values taken from Ref. [6]. The upper state $|0^U\rangle = \frac{1}{\sqrt{2}}[|0^+\rangle + |0^-\rangle]$ ($|0^\pm\rangle = |\pm\frac{1}{2}, \mp\frac{1}{2}\rangle$) contributes to the optical transition, which is polarized in the z growth direction. Below it is located the $|\pm 1\rangle = |\pm\frac{1}{2}, \pm\frac{1}{2}\rangle$ optically active doublet, which is circularly (σ^+ and σ^-, respectively) polarized. The doublet states can be slightly split (due to the in-plane anisotropy of the confinement potential) into two linearly polarized states (labeled $|X\rangle$ and $|Y\rangle$) with dipole moments along the two nonequivalent in-plane QD axes. The lower state $|0^L\rangle = \frac{1}{\sqrt{2}}[|0^+\rangle - |0^-\rangle]$ is optically forbidden [15]. Hence for the lh exciton ground state, there are three bright recombination channels associated with the $|\pm 1\rangle$ (or $|X\rangle$ and $|Y\rangle$) states and the $|0^U\rangle$ state, respectively. These states are characterized by different radiative lifetimes $\tau_{r\perp} = 3\tau_r$ and $\tau_{r\parallel} = 3\tau_r/2$, respectively, where τ_r is the radiative lifetime of the hh exciton [16]. Phonon-assisted transitions, in which an independent spin flip of the exciton-bound electrons or light holes occurs, are possible between the states with $F_z = 0$ and $F_z = \pm 1$. Consequently, such processes are available between the $|0^U\rangle$ ($|0^L\rangle$) state and the $|\pm 1\rangle$ states—the $U \leftrightarrow 1$ and $1 \leftrightarrow L$ transitions shown in Fig. 1. Two other transitions in Fig. 1, the

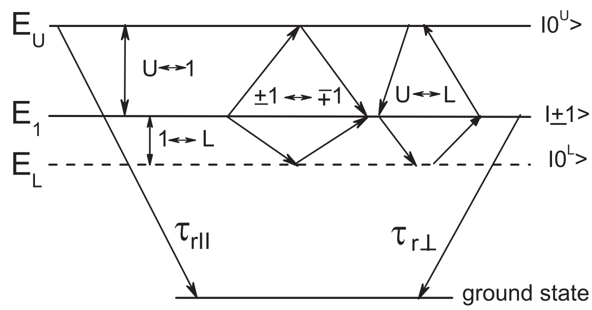

FIG. 1. The light-hole exciton spin states and the exciton-bound single-particle spin-flip transitions in a quantum dot with a ground state of the light-hole type; $\tau_{r\parallel}$ and $\tau_{r\perp}$ are the radiative lifetimes.

$U \leftrightarrow L$ and $\pm1 \leftrightarrow \mp1$ transitions, require the participation of two phonons and involve the intermediate states [17].

II. THE MODEL

As noted above, we concentrate on the lh exciton spin relaxation driven by the SO interaction in weakly confined QDs. For this case, a way to calculate the spin-flip transitions within the lh exciton fine structure is, formally, similar to that for the hh exciton developed in Ref. [9]. Here we formulate briefly some necessary statements and pay more attention to the features typical for the lh exciton. The main assumptions and definitions are the following. The spin-flip transitions are driven by the $\vec{k}$-linear part of the SO interaction (in the conduction and light-hole valence bands) and the piezoelectric nature of the carrier-phonon interaction. The confinement along the growth direction is assumed to be stronger than both the lateral quantum dot and the Coulomb potentials, so that the envelope for the electron-hole pair wave function $\Psi(\vec{r}_e,\vec{r}_h) = \psi(\vec{\rho}_e,\vec{\rho}_h)\phi_e(z_e)\phi_h(z_h)$ is factorized, 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 the 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, $\psi(\vec{\rho}_e,\vec{\rho}_h) = \phi(\vec{\rho})F(\vec{R})$, and only the exciton center-of-mass motion is affected by the lateral confinement. In what follows the lateral potential is assumed to be a harmonic potential $V(\vec{R}) = M\Omega^2R^2/2$, where $M = m_e + m_{lh\perp}$ is the translational mass of the lh exciton and $m_{lh\perp}$ is the light-hole mass in the (x,y) direction [18]. For electrons in the Γ_6 conduction band the SO interaction is given by [19]

$$H^e_{\rm SO} = \beta_e(\sigma_y k_y - \sigma_x k_x), \quad (1)$$

where $\vec{k}_\perp = \{k_x,k_y\}$ is the (in-plane) momentum operator, $\vec{\sigma}$ are the Pauli matrices, and the strength of the SO coupling β_e depends on the material and the height of the QD, $\beta_e = \gamma_c\langle k_z^2\rangle \sim l_z^{-2}$ with γ_c the Dresselhaus constant [20]. This term arises from the cubic-$\vec{k}$ term in the electron Hamiltonian describing the removal of the spin degeneracy of the conduction-band states in a bulk semiconductor without inversion symmetry [21]. It is usually called the bulk inversion asymmetry term, or sometimes the Dresselhaus term. For the light holes, we consider a $\vec{k}$-linear term similar to that for heavy holes, which in the light-hole basis has the form [9,22]

$$H^{\rm lin}_{\rm SO} = \beta_h(\sigma_x k_y + \sigma_y k_x), \quad (2)$$

where the strength β_h is of relativistic origin.

III. DECAY RATES, RESULTS AND DISCUSSION

A. One-phonon processes

For a spin-flip transition accompanied by emission of a single phonon, the relaxation rate calculated from Fermi's golden rule is given by

$$\frac{1}{\tau^{(e,h)}_{i\to j}} = \frac{2\pi}{\hbar}\sum_{\vec{q}}\left|M^{e,h}_{\vec{q}}\right|^2[N_{\vec{q}}+1]\delta(E_i - E_j - \hbar\omega_{\vec{q}}), \quad (3)$$

where $\tau^{(e)}_{i\to j}$ $(\tau^{(h)}_{i\to j})$ determines the relaxation due to the spin flip of the (exciton-bound) electron (hole) and the notation i (j) stands for the initial (final) state with the energy E_i (E_j). The matrix element $M^{e,h}_{\vec{q}} = \langle\Psi_i|H^{e,h}_{\vec{q}}|\Psi_j\rangle$, where the electron-(hole-) phonon interaction $H^{e,h}_{\vec{q}}$ is calculated to first order of the SO-coupling strength β_e (β_h), and $N_{\vec{q}}$ is the thermal (acoustic) phonon distribution function. Considering the (in-plane) symmetrical QD, the resulting transition rates have the forms

$$\frac{1}{\tau^{(e,h)}_{U\to1}} = \frac{1}{\tau^{(e,h)}_{|0^U\rangle\to|+1\rangle}} = \frac{1}{\tau^{(e,h)}_{|0^U\rangle\to|-1\rangle}} = w_{U1}\left(\frac{m_{e,lh\perp}}{M}\right)^2\beta^2_{e,h}(N_{U1}+1), \quad (4)$$

$$\frac{1}{\tau^{(e,h)}_{1\to L}} = \frac{1}{\tau^{(e,h)}_{|+1\rangle\to|0^L\rangle}} = \frac{1}{\tau^{(e,h)}_{|-1\rangle\to|0^L\rangle}} = w_{1L}\left(\frac{m_{e,lh\perp}}{M}\right)^2\beta^2_{e,h}(N_{1L}+1), \quad (5)$$

$$w_{nm} = \frac{(eh_{14})^2}{70\pi\rho\hbar^3s^5}\frac{E_n - E_m}{\hbar}\left(\frac{E_n - E_m}{\hbar\Omega}\right)^2 \times\left(\frac{E_n - E_m}{\hbar\Omega + E_n - E_m}\right)^2 I(E_n - E_m), \quad (6)$$

$$I(E_n - E_m) = \frac{35}{32}\sum_{k=l,t}\left(\frac{s}{s_k}\right)^5\int_0^\pi e^{-a^2(E_n-E_m)^2\sin^2\vartheta/\hbar^2 s_k^2} \times g_k(\vartheta)\sin^3\vartheta\, d\vartheta, \quad (7)$$

with $g_t = 8\cos^2\vartheta\sin^2\vartheta + \sin^4\vartheta - 9\sin^4\vartheta\cos^2\vartheta$ and $g_l = 9\cos^2\vartheta\sin^4\vartheta$ [23]. In Eqs. (4)–(7) the occupation factor $N_{nm} = (e^{(E_n-E_m)/k_BT} - 1)^{-1}$, $\hbar\Omega \approx \hbar^2/2Ma^2$ is the lateral quantization energy with a the dot diameter [9], and the summation is taken over the longitudinal (l) and transversal (t) acoustic phonon branches. The first factor in Eq. (6) contains the piezotensor component h_{14}, the crystal mass density ρ, and the sound velocity $s^{-5} = s_l^{-5} + 4s_t^{-5}/3$. For the spin-flip transition accompanied by absorption of a single phonon, the corresponding relaxation rate $\tau^{-1}_{j\to i} = \tau^{-1}_{i\to j}\, e^{-(E_i-E_j)/kT}$.

According to Eqs. (4) and (5), the relative contribution of the (exciton-bound) electron and hole to the spin-relaxation times depends on the masses, the spin-orbit parameters, and the height of the QD (we recall that the SO strength $\beta_e = \gamma_c \langle k_z^2 \rangle \sim l_z^{-2}$), $\tau^{(e)} = \tau^{(h)}(m_{lh\perp}/m_e)^2(\beta_h/\beta_e)^2$. With typical parameters for GaAs $\gamma_c = 24.5$ eV Å^3 [21], $|\beta_h| = 11$ meV Å [24], $m_e = 0.067 m_0$, and $m_{lh\perp} = 0.2 m_0$ [12], the (exciton-bound) light-hole spin relaxation significantly dominates since $\tau^{(h)} \sim 0.01\tau^{(e)}$ at $l_z \approx 8$ nm (reported in Ref. [6]). For both the (exciton-bound) electron and hole, the relaxation rates $\tau^{-1}_{U\to 1}$ and $\tau^{-1}_{1\to L}$ differ by the energy splitting between the involved states. For a small energy transfer ($E_n - E_m) < \hbar s/a$, long-wave acoustic phonons mainly contribute to the relaxation process, the envelope integral Eq. (7) is limited to unity, and, consequently, the relaxation is the more efficient the larger is the density of the phonon states. At a large energy transfer ($E_n - E_m) > \hbar s/a$, however, the contribution of the short-wave phonons becomes increasingly important and the envelope integral results in a decrease of the relaxation rate. Note that in Ref. [6] the experimentally deduced splittings $E_U - E_1 = 200$–600 μeV and $E_1 - E_L = 30$–40 μeV are reported, whereas the characteristic energy $\hbar s/a \sim 100$ μeV is calculated at $a \approx 25$ nm and $s \sim 3 \times 10^5$ cm/s.

In order to evaluate numerically the (light-hole) spin-relaxation rates from Eqs. (4)–(7) we take $E_U - E_1 = 0.4$ meV, $E_1 - E_L = 0.04$ meV, and $a = 25$ nm. For the other parameters we use $|\beta_h| = 11$ meV Å [24], $M = 0.3 m_0$, $m_{lh\perp} = 2.3 m_0$, $\hbar\Omega = 0.1$ meV, $\rho = 5$ g/cm^3, and $eh_{14} = 1.2 \times 10^7$ eV/cm [12]. In Fig. 2 we show the temperature dependence of the spin lifetimes $\tau^{(h)}_{U\to 1}$ (solid line) and $\tau^{(h)}_{1\to L}$ (dashed line) in a wide temperature range. Separately, at low and moderate temperatures these relaxation times are shown in the inset in Fig. 2. It is seen in Fig. 2 that at low temperature both the $|0^U\rangle$ and $|\pm 1\rangle$ bright states decay with the relaxation time on the order of a few tens of nanoseconds. At very low temperature the $1 \to L$ transition, which is characterized by a small energy transfer, is noticeably slower than the $U \to 1$ transition, while both processes are characterized by similar rates already at the temperatures of a few kelvins, as is seen in the inset in Fig. 2. At moderate temperature these processes show relaxation times on the order of a few nanoseconds and therefore compete with the radiative decay, which for typical (hh exciton) QDs occurs usually on the nanosecond scale [25].

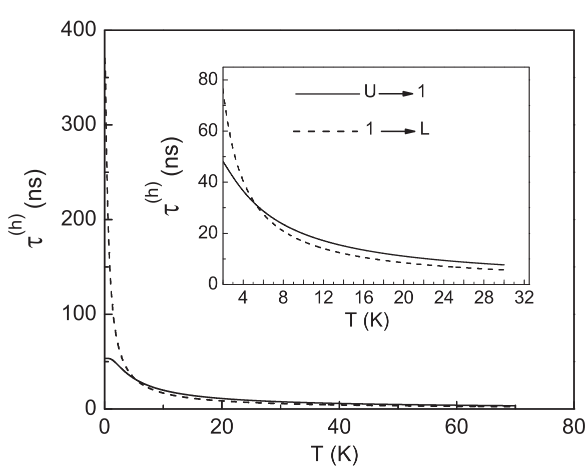

FIG. 2. The one-phonon relaxation times within the fine structure of the light-hole exciton ground state as a function of temperature. For the parameters and details see the text.

B. Two-phonon processes

Consider first the two-phonon processes contributing to the exciton spin relaxation within the in-plane polarized radiative doublet of the light-hole exciton ground state, so-called longitudinal spin relaxation. For a symmetrical dot, a direct (single-phonon) relaxation between these states is completely absent because of the zero density of the phonon states involved, so that the indirect channel is of paramount importance. Let us begin with the "indirect" process in which the dark state serves as the intermediate state; see Fig. 1. In this process a phonon is scattered from the state $\vec{q}$ to the state $\vec{q}\,'$ while the spin of the bright exciton flips. The corresponding relaxation rate calculated from Fermi's golden rule is given by

$$\frac{1}{\tau^{(e,h)}_{|+1\rangle\,\to\,|-1\rangle}} = \frac{2\pi}{\hbar}\sum_{\vec{q},\vec{q}\,'}\left|M^{e,h}_{\vec{q},-\vec{q}\,'}\right|^2 \delta(E_{+1} - E_{-1} + \hbar\omega_{\vec{q}} - \hbar\omega_{\vec{q}\,'}) \times N_{\vec{q}}(N_{\vec{q}\,'} + 1)\,. \tag{8}$$

The effective matrix element involved in Eq. (8) contains transitions from one of the bright states to the dark state with the emission of a phonon and then back to the other (orthogonally polarized) bright state with the absorption of a phonon; see Fig. 1. The exciton spin is changed in both the first and second transitions. The resonant part of the transition matrix element has the form [26,27]

$$M^{e,h}_{\vec{q},-\vec{q}\,'} = \frac{\langle -1|H^{e,h}_{\vec{q}}|0^L\rangle\langle 0^L|H^{e,h}_{\vec{q}\,'}|+1\rangle}{E_{-1} - E_L - \hbar\omega_{\vec{q}} + i\hbar/2\tau_L}, \tag{9}$$

where the phonon matrix elements are calculated to first order of the SO coupling strength β_e (β_h) and τ_L is the dark state lifetime with respect to all allowed destroying processes. At low temperature the dark (lh exciton ground) state lifetime can be approximated by

$$\frac{1}{\tau_L} = \frac{1}{\tau_{nr}} + \sum_{i=e,h}\frac{1}{\tau^{(i)}_{|0^L\rangle\to|+1\rangle}} + \sum_{i=e,h}\frac{1}{\tau^{(i)}_{|0^L\rangle\to|-1\rangle}}, \tag{10}$$

where τ_{nr} is the dark exciton nonradiative lifetime. (A nonradiative decay of $\tau_{nr} \approx 10$ ns, e.g., has been reported for conventional InGaAs/GaAs self-assembled QDs in Ref. [28].) Using the expression $|1/(\varepsilon + i\Gamma_L/2)|^2 = (2\pi/\Gamma_L)\delta(\varepsilon)$, where $\Gamma_L = \hbar/\tau_L$ is the dark state width, from Eq. (8) one immediately obtains

$$\frac{1}{\tau^{(e,h)}_{|+1\rangle\to|0^L\rangle\to|-1\rangle}} = \frac{1}{\tau^{(e,h)}_{|+1\rangle\to|0^L\rangle}}\,\frac{\tau_L}{\tau^{(e,h)}_{|0^L\rangle\to|-1\rangle}}\,, \tag{11}$$

where the factor $\tau_L/\tau^{(e,h)}_{|0^L\rangle\to|-1\rangle}$ can be viewed as the conditional probability that the lh exciton being in the dark state relaxes just to the $|-1\rangle$ bright state by means of the electron (hole) spin flip. From the above result, which is relevant for a symmetrical dot with no anisotropic exchange splitting, the relation $\tau_{|+1\rangle\to|0^L\rangle\to|-1\rangle} = \tau_{|-1\rangle\to|0^L\rangle\to|+1\rangle}$ follows. Similarly, for the longitudinal relaxation, which proceeds via the $|0^U\rangle$ bright state as the intermediate state (see Fig. 1), one obtains

$$\frac{1}{\tau^{(e,h)}_{|+1\rangle\to|0^U\rangle\to|-1\rangle}} = \frac{1}{\tau^{(e,h)}_{|+1\rangle\to|0^U\rangle}}\,\frac{\tau_0}{\tau^{(e,h)}_{|0^U\rangle\to|-1\rangle}}. \tag{12}$$

Here τ_0 is the lifetime of the (lh exciton ground) z-polarized bright state (with respect to all allowed destroying processes), which can be approximated by

$$\frac{1}{\tau_0} = \frac{1}{\tau_{r\parallel}} + \frac{1}{\tau_{nr}} + \sum_{i=e,h} \frac{1}{\tau^{(i)}_{|0^U\rangle\to|+1\rangle}} + \sum_{i=e,h} \frac{1}{\tau^{(i)}_{|0^U\rangle|\to-1\rangle}}, \quad (13)$$

where $\tau_{r\parallel}$ is the corresponding radiative lifetime. The factor $\tau_0/\tau^{(e,h)}_{|0^U\rangle\to|-1\rangle}$ in Eq. (12) can be viewed as the conditional probability that the lh exciton in the $|0^U\rangle$ intermediate state relaxes just to the $|-1\rangle$ bright state. For QDs of high quality, the radiative recombination can dominate in a wide temperature range, that is, $\tau_{r\parallel} \ll \tau_{nr}$. Supposing also that the hole spin relaxation dominates, the total relaxation rate is given by (we recall that for symmetrical QDs the equality $\tau_{|+1\rangle\leftrightarrow|0^{L(U)}\rangle} = \tau_{|-1\rangle\leftrightarrow|0^{L(U)}\rangle} = \tau_{1\leftrightarrow L(U)}$ holds)

$$\frac{1}{\tau^{(h)}_{|+1\rangle\to|-1\rangle}} = \frac{1}{\tau^{(h)}_{1\to L}}\,\frac{\tau_{nr}}{\tau^{(h)}_{L\to 1} + 2\tau_{nr}} + \frac{1}{\tau^{(h)}_{1\to U}}\,\frac{\tau_{r\parallel}}{\tau^{(h)}_{U\to 1} + 2\tau_{r\parallel}}. \quad (14)$$

As expected, the relaxation rate Eq. (14) tends to zero at zero temperature since one of the sequential transitions is assisted by the absorption of a phonon.

Consider now the spin-flip transition from the z-polarized bright to the dark state. Although both of these states are characterized by the same total momentum $F_z = 0$, direct (thermally activated) transition between the symmetric $|0^U\rangle$ and antisymmetric $|0^L\rangle$ superposition of the basis states is, evidently, absent. The desirable transitions, however, can happen with a participation of the intermediate states, as is shown in Fig. 1. For the $U \to L$ process, which is accompanied by the emission of two phonons, the relaxation rate is given by (for simplicity, we concentrate as before on the hole spin relaxation)

$$\frac{1}{\tau^{(h)}_{U\to L}} = \frac{2\pi}{\hbar}\sum_{\vec q,\vec q'} \left|M^h_{\vec q,\vec q'}\right|^2 \delta(E_U - E_L - \hbar\omega_{\vec q} - \hbar\omega_{\vec q'}) \times (N_{\vec q}+1)(N_{\vec q'}+1). \quad (15)$$

For this process, the $F_z = \pm 1$ states of the lh exciton serve as the intermediate states (which we label below by $|k\rangle$) and the effective matrix element has the form [26]

$$M^h_{\vec q,\vec q'} = \sum_{k=|+1\rangle,|-1\rangle}\left\{\frac{\langle 0^L|H^h_{\vec q'}|k\rangle\langle k|H^h_{\vec q}|0^U\rangle}{E_U - E_k - \hbar\omega_{\vec q} + i\hbar/2\tau_k} + \frac{\langle 0^L|H^h_{\vec q}|k\rangle\langle k|H^h_{\vec q'}|0^U\rangle}{E_U - E_k - \hbar\omega_{\vec q'} + i\hbar/2\tau_k}\right\}, \quad (16)$$

where τ_k is the lifetime of the intermediate $|k\rangle$ state with respect to all allowed destroying processes. Below for both intermediate states we take the same lifetime $\tau_{|+1\rangle} = \tau_{|-1\rangle} = \tau_1$. One can check that for the matrix elements involved in Eq. (16) the equality $\langle 0^L|H^h_{\vec q'}|+1\rangle\langle +1|H^h_{\vec q}|0^U\rangle = -\langle 0^L|H^h_{\vec q'}|-1\rangle\langle -1|H^h_{\vec q}|0^U\rangle$ holds. Consequently, the considered process is not relevant for ideally symmetrical QDs with no anisotropic exchange splitting. Introducing a slight hybridization of the $F_z = \pm 1$ states, we suppose that the respective anisotropic splitting Δ_{xy} is small as compared to the energy transfer in both sequential transitions. Considering the resonant contributions and using the expression $|1/(\varepsilon + i\Gamma_1/2)|^2 = (2\pi/\Gamma_1)\delta(\varepsilon)$, where $\Gamma_1 = \hbar/\tau_1$ is the intermediate state width, one obtains

$$\frac{1}{\tau^{(h)}_{U\to L}} = \frac{\Delta^2_{xy}}{\Delta^2_{xy} + \Gamma^2_1}\,\frac{4}{\tau^{(h)}_{|0^U\rangle\to|+1\rangle}}\,\frac{\tau_1}{\tau^{(h)}_{|+1\rangle\to|0^L\rangle}}, \quad (17)$$

where the factor $\tau_1/\tau^{(h)}_{|+1\rangle\to|0^L\rangle}$ is the conditional probability that the lh exciton in the intermediate state relaxes just to the dark state by means of a hole spin flip. Note that from the above expression the obvious relation $\tau^{(h)}_{L\to U} = \tau^{(h)}_{U\to L} e^{(E_L - E_U)/k_BT}$ follows. Hence for the relaxation Eq. (15) the correlation between the intermediate level separation Δ_{xy} and the width Γ_1 is important [29]. In the low-temperature regime, the lifetime of the in-plane polarized bright (lh exciton ground) state can be approximated by

$$\frac{1}{\tau_1} = \frac{1}{\tau_{r\perp}} + \frac{1}{\tau_{nr}} + \sum_{i=e,h}\frac{1}{\tau^{(i)}_{|+1\rangle\to|0^L\rangle}} + \sum_{i=e,h}\frac{1}{\tau^{(i)}_{|+1\rangle\to|0^U\rangle}}, \quad (18)$$

where $\tau_{r\perp}$ is the corresponding radiative lifetime.

Believing as before that the hole spin relaxation dominates, that is, $\tau^{(e)} \gg \tau^{(h)}$, the relaxation rate Eq. (17) reduces to (at $\tau_{r\perp} \ll \tau_{nr}$)

$$\frac{1}{\tau^{(h)}_{U\to L}} = \frac{\Delta^2_{xy}}{\Delta^2_{xy} + \Gamma^2_1}\,\frac{4\,\tau_{r\perp}\, e^{(E_U - E_1)/k_BT}}{\left[\tau^{(h)}_{1\to L}\tau^{(h)}_{1\to U} + \tau_{r\perp}\left(\tau^{(h)}_{1\to L} + \tau^{(h)}_{1\to U}\right)\right]}. \quad (19)$$

As follows from Eqs. (14) and (19), the relaxation rates of the considered two-phonon processes are determined by the competition between different decay channels of the intermediate states. If the intermediate states contribute mainly to radiative and nonradiative decay, the indirect relaxation is strongly suppressed in comparison with the involved single-phonon transitions. When the spin-flip processes strongly dominate, which can happen at elevated temperatures, the indirect relaxation is governed by single-phonon transitions.

In order to estimate the above relaxation rates, we use the same parameters as in Fig. 2 and we take $\Delta_{xy} = 3\ \mu$eV, $\tau_{nr} = 10$ ns, and $\tau_r = 0.5$ ns (we recall that the involved radiative lifetimes $\tau_{r\perp} = 3\tau_r$ and $\tau_{r\parallel} = 3\tau_r/2$). In Fig. 3 we show the dependence of the (hole) spin lifetimes $\tau^{(h)}_{|+1\rangle\to|-1\rangle}$ and $\tau^{(h)}_{U\to L}$ on the temperature at low (the main panel) and moderate (the inset) temperatures. For comparison, the one-phonon relaxation times $\tau^{(h)}_{U\to 1}$ and $\tau^{(h)}_{1\to L}$ are also presented in the inset. In Fig. 3 it is seen that both considered indirect processes, being very slow at low temperature, are significantly accelerated with growth of the temperature. The respective decay times are about 1 μs at 2 K and decrease to a couple of nanoseconds at 70 K. In the inset in Fig. 3 it is also seen that at a high enough temperature the two- and one-phonon transitions are characterized by similar decay times, as expected.

Hence, according to the numerical estimates obtained (with the splitting parameters reported in Ref. [6] for GaAs QDs of the light-hole type), for the radiative triplet of the lh exciton ground state the main decay channel at low temperature is radiative decay, whereas at moderate temperature the spin-flip transitions also become important. In this case radiative decay of the z-polarized exciton is limited by the spin-flip transition

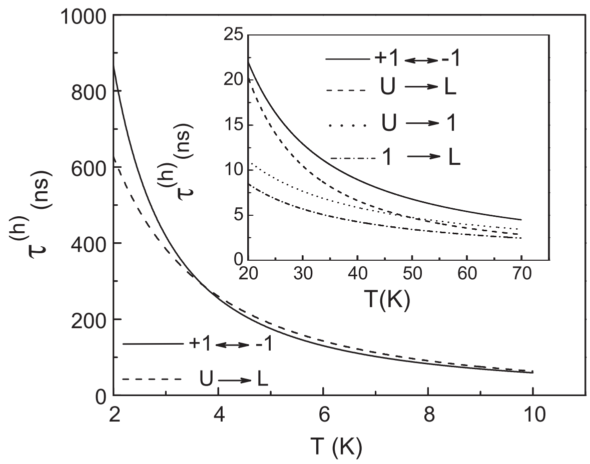

FIG. 3. The two–phonon relaxation times within the fine structure of the light-hole exciton ground state as a function of temperature. Inset: The one- and two-phonon relaxation times. For the parameters see the text.

to the bright doublet located below, while the thermalization process to the dark state is still suppressed. Similarly, radiative decay of the in-plane polarized bright state at moderate temperature is limited by decay to the dark state and the (z-polarized) bright state as well, while the longitudinal relaxation is much less important. Here we would like to note that the presented numerical results for the lh exciton spin lifetimes are calculated with dot parameters, such as the electron and light-hole masses, the SO strengths, etc., which are typical for unstrained GaAs QDs. To obtain the exact quantitative results for strained QDs, similar to those studied in Ref. [6], a separate detailed analysis probably must be carried out.

C. Magnetic field effects

Now we consider the impact of the longitudinal (parallel to the z direction) magnetic field on the lh exciton spin relaxation. We will discuss this just briefly since a complete solution of this problem requires separate consideration. The Zeeman effect leads to a further transformation of the spin states of the lh exciton. For the states with $F_z = \pm 1$, the resulting wave functions (in an asymmetrical QD with the anisotropic splitting Δ_{xy}) are

$$\Psi_1^{U,L} = \frac{1}{\sqrt{2}}\left[\sqrt{1 \pm \frac{\Delta_B^{(1)}}{\sqrt{\left(\Delta_B^{(1)}\right)^2 + \Delta_{xy}^2}}}\,|+1\rangle \pm \sqrt{1 \mp \frac{\Delta_B^{(1)}}{\sqrt{\left(\Delta_B^{(1)}\right)^2 + \Delta_{xy}^2}}}\,|-1\rangle\right], \qquad (20)$$

where the Zeeman splitting $\Delta_B^{(1)} = \mu_B(g_e - g_h)B$ with g_e (g_h) the electron (hole) g factor [14]. Similarly, for states with $F_z = 0$, the resulting wave functions are

$$\Psi_0^{U,L} = \frac{1}{\sqrt{2}}\left[\sqrt{1 \pm \frac{\Delta_B^{(0)}}{\sqrt{\left(\Delta_B^{(0)}\right)^2 + (\Delta_{UL})^2}}}\,|0^+\rangle \pm \sqrt{1 \mp \frac{\Delta_B^{(0)}}{\sqrt{\left(\Delta_B^{(0)}\right)^2 + (\Delta_{UL})^2}}}\,|0^-\rangle\right], \qquad (21)$$

where $\Delta_B^{(0)} = \mu_B(g_e + g_h)B_z$ and $\Delta_{UL} = E_U - E_L$. The basis states now contribute to the wave functions Eqs. (20) and (21) with different weights, which depend on the magnitude of the magnetic field. Consequently, the initially linearly (in-plane) polarized bright states become elliptically polarized, while the initially dark state becomes (at least partially) optically active with the oscillator strength increasing with an increase in the magnetic field. Moreover, at strong enough magnetic fields, when the Zeeman splitting prevails over the exchange-induced splitting, the $|s_z, j_z\rangle$ basis states can be restored and then all of the four lh exciton states will be bright. For in-plane polarized bright states with anisotropic splitting of a few micro electron volts, recovery of the $|\pm 1\rangle$ unperturbed states happens already at small fields, as was observed in Ref. [6]. To reach a similar result for the $F_z = 0$ states with exchange-induced splitting of a few hundreds of micro electron volts, large magnetic fields are required.

The magnetic field affects the one- and two-phonon transitions within the fine structure of the lh exciton in different ways. In principle, the direct (single-phonon) transitions are still allowed. However, the spin-flip transitions from (to) the two in-plane polarized states, which are split by the Zeeman interaction, are characterized now by a different energy transfer. Additionally, the field-induced change of the wave functions of the states involved leads to various transition probabilities. As a result, since the basis states are coupled by the SO interaction only in pairs [30], not all of the one-phonon relaxation channels survive at large fields, which leads to a quenching of the two-phonon processes. To illustrate, note that for symmetrical QDs the momentum and spin operators included in the SO interaction [see Eqs. (1) and (2)] act separately. The probabilities of the one-phonon transitions therefore are effectively governed by the (square) matrix element of a spin Hamiltonian of the form $H_s^{(e,h)} = \sigma_y \mp \sigma_x$ (in the electron and hole bases, respectively) constructed on the spin eigenfunctions. For single-phonon processes, the respective (normalized) probabilities are shown in (the main panel of) Fig. 4 in dependence on the magnetic field. The calculations are performed with the set of parameters $\Delta_{xy} = 10\ \mu$eV, $\Delta_{UL} = 480\ \mu$eV, $g_e - g_h = 1.4$, and $g_e + g_h = -2.4$, which is relevant for the GaAs QDs from Ref. [6]. It is seen in Fig. 4 that among the four possible transitions (for each carrier, the hole and electron) only two of them survive at large fields. For example, when the exciton-bound hole flips its spin, the transition probability between the $|\Psi_0^U\rangle$ and $|\Psi_1^U\rangle$ states (and between the $|\Psi_0^L\rangle$ and $|\Psi_1^L\rangle$ states as well) tends to zero with growth of the field. For the exciton-bound electron, the two other spin-flip processes, between the $|\Psi_0^U\rangle$ and $|\Psi_1^L\rangle$ states (and between the $|\Psi_0^L\rangle$ and $|\Psi_1^U\rangle$ states as well), are not effective at large fields. Schematically, this is shown in the inset

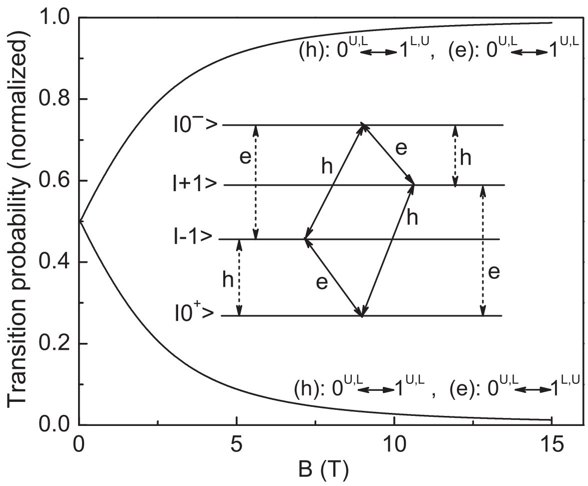

FIG. 4. The one-phonon transition probabilities within the fine structure of the light-hole exciton ground state as a function of magnetic field. Inset: Allowed (solid arrows) and quenched (dashed arrows) transitions at large magnetic fields. For the parameters see the text.

in Fig. 4, where the solid (dashed) arrows refer to the allowed (quenched) transitions. In the inset in Fig. 4 it is seen that at large fields the two-phonon processes become practically forbidden and only an independent flip of the hole and then the electron spins (or vice versa) results in the indirect spin flip transitions between the $|\pm 1\rangle$ bright states and between the $|0^{\pm}\rangle$ bright states as well.

IV. ADDITIONAL REMARKS

Below we would like to provide some remarks and additions. (i) For the light-hole-related relaxation rates, the presented results Eqs. (4)–(7) are obtained with the $\vec{k}$-linear SO interaction Eq. (2), which is of relativistic origin. Actually, both the light-hole subband and the electron subband transform according to the same spinor representation Γ_6 [22] and, consequently, for the light hole, the SO interaction of the form Eq. (1) is also relevant,

$$H_{SO}^{cub} = \alpha_h(\sigma_x k_x - \sigma_y k_y), \tag{22}$$

where the strength $\alpha_h = \gamma_h \langle k_z^2 \rangle$ has a nonrelativistic nature and depends on the height of the QD. The relative magnitude of the relaxation times τ_{lin} and τ_{cub} due to the relativistic and the Dresselhaus SO coupling, respectively, is given by

$$\tau_{lin} = \tau_{cub}\left(\frac{\alpha_h}{\beta_h}\right)^2. \tag{23}$$

Using the parameters for GaAs, $|\beta_h| = 11$ meV Å [24] and $|\gamma_h| = 9$ eV Å^3 [31], from Eq. (23) we obtain that $\tau_{cub} \lesssim \tau_{lin}$ at $l_z \lesssim 3$ nm, that is, the Dresselhaus term Eq. (22) is important in flat enough QDs, as expected. For the case considered here, the relativistic term Eq. (2) strongly dominates since $\tau_{lin} \approx 0.02\ \tau_{cub}$ at $l_z \approx 8$ nm. (ii) For QDs with an asymmetric confining potential in the growth direction, an additional source for the spin relaxation can arise from the Rashba spin-orbit coupling [32]. Similarly to the Dresselhaus term Eq. (22), the Rashba term is relevant for both the exciton-bound electron and the light hole, whereas the Rashba coupling contributes to the hh splitting only when the hh-lh mixing is taken into account [33]. (iii) In principle, for the light holes, as for electrons, the spin-flip transitions in QDs can be mediated by the hyperfine interaction with nuclei. For electrons in GaAs QDs, the nucleus-mediated spin-flip transition rate is found to be lower than the spin-orbit rates [34]. (iv) The presented results for single-phonon processes are calculated for the piezoelectric type of carrier-phonon interaction which is known to be most effective in polar crystals for small energy transfer [26]. The relative magnitudes of the relaxation times τ_{def} and τ_{piezo} due to the deformation potential and the piezoelectric type of carrier-phonon interaction, respectively, are given by [9]

$$\tau_{def}^{(e,h)} \sim \tau_{piezo}^{(e,h)}\left(\frac{eh_{14}}{D^{(e,h)}}\right)^2\left(\frac{\hbar s}{\Delta}\right)^2, \tag{24}$$

where D^e (D^h) is the deformation potential constant in the conduction (valence) band and Δ is the energy transfer. For typical values $D \simeq 5$ eV and $s = 2.4 \times 10^5$ cm/s, we obtain $\tau_{def} \lesssim \tau_{piezo}$ at $\Delta \gtrsim 0.4$ meV, so that the deformation potential interaction becomes important for the $U \leftrightarrow 1$ transition, which is calculated here for the energy transfer $E_U - E_1 = 0.4$ meV. The obtained relaxation times are therefore overestimated approximately twice, but the corrected values are still on the order of a few tens of nanoseconds at low temperature, as follows from Fig. 2. (v) For strained QDs, the impact of the valence band mixing (VBM) on the exciton eigenstates due to anisotropic strain effects can be important (a strictly isotropic biaxial strain is probably difficult to reach in experiments). (Another obvious reason of the light-hole–heavy-hole mixing is the in-plane shape anisotropy of the dot.) Definitely, the VBM in itself cannot be a source of the exciton spin relaxation (the electron spin is not affected) and all possible changes are caused by the interplay between the SO coupling and the Bir-Pikus interaction [22]. The hole mixing due to the k-linear interaction of the relativistic origin now also becomes effective [9,22]. [In the light-hole basis this interaction is given by Eq. (2).] Additionally, the interplay between the VBM and the short-range exchange interaction contributes to splitting of the bright doublet, similarly to the heavy-hole exciton [35]. Consequently, even for (strained) QDs with a symmetrical (in-plane) shape, the bright doublet can be split into two linearly polarized states. Further, for the in-plane strain anisotropy, the light-hole states with $F_z = 0$ (± 1) are coupled to the heavy-hole states with $F_z = \pm 2$ (∓ 1), respectively [36]. As a result, transitions between the z-polarized and dark states and within the in-plane polarized doublet as well can still proceed only with participation of two phonons. For the one-phonon transitions, the above effects result in additional terms in the corresponding transition matrix elements, which are proportional to the heavy-hole character (the probability for the hole to be heavy). For the strained dots of interest (with a pronounced light-hole character of the ground state) these additional terms will evidently not be very important.

V. CONCLUSION

In summary, for (strained) quantum dots with the light-hole exciton ground state, scenario of the phonon-induced exciton spin relaxation is more various than for similar (unperturbed) quantum dots with the heavy-hole exciton ground state. Generally, for the light-hole subband, which transforms according to the same spinor representation as the electron subband, additional sources of spin flips appear in comparison with the heavy-hole subband. Besides, for the light-hole exciton, there are additional channels of spin relaxation. Within the fine structure of the heavy-hole exciton the main thermally activated spin-flip process is a decay of the bright state to dark states, which can limit the generation of single photons from a dot. Similar transitions between the in-plane-polarized bright states and the dark state are present for the light-hole exciton as well. Here, however, an additional spin-flip transition—a decay of the z-polarized bright state to the in-plane polarized bright state—is relevant. Moreover, for the z-polarized bright state, the above process is the main decay channel at low temperature. Decay of this state to the dark state becomes important only at high enough temperatures. The numerical estimates obtained allow one to believe that the exciton spin relaxation in the light-hole quantum dots, as in the case of similar conventional quantum dots, is most likely quenched over (at least) tens of nanoseconds at low temperatures.

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