Slow Exciton Spin Relaxation in Single Self-Assembled In$_{1-x}$Ga$_x$As/GaAs Quantum Dots

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We calculate the acoustic phonon-assisted exciton spin relaxation in single self-assembled In$_{1-x}$Ga$_x$As/GaAs quantum dots using an atomic empirical pseudopotential method. We show that the transition from bright to dark exciton states is induced by Coulomb correlation effects. The exciton spin relaxation time obtained from sophisticated configuration interaction calculations is approximately 15–55 µs in pure InAs/GaAs QDs and even longer in alloy dots. These results contradict previous theoretical and experimental results, which suggest very short exciton spin times (a few ns), but agree with more recent experiments that suggest that excitons have long spin relaxation times (> 1 µs).

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Self-assembled quantum dots (QDs) have many attractive features as fundamental building blocks for quantum information processing. However, their short spin lifetime is still a major obstacle for such applications. There have been extensive studies of single electron and hole spin relaxation in QDs caused by hyperfine interaction with nuclear spins [1, 2] and the spin-phonon interaction due to spin-orbit coupling (SOC) [3–10]. However, exciton spin relaxation has been less commonly studied.

Excitons and biexcitons in QDs have been used to generate single photons [11] or entangled photon pairs [12]. Bright and dark excitons have also been proposed as possible quantum bits (qubits) [13, 14]. The fast nonradiative relaxation of bright excitons limits the maximal single-photon device emission rate and thus lowers the source efficiency [15]. This property also lowers the quality of the single photons and the fidelity of the entangled photon pairs generated by biexciton cascade decay [16]. Despite its importance, spin relaxation in excitons is still not well understood and full of controversy.

Exciton spin relaxation has been measured by several groups in different types of QDs [17–19]. The measured spin relaxation time ranges from 200 ps [17] to 167 ns [18]. The spin relaxation time calculated from perturbation theory is approximately 2 ns in In(Ga)As/GaAs QDs at 4 K [20], which seems to be in good agreement with experimental values [19]. All of these studies suggest fast spin relaxation for excitons. However, recent direct measurements [21, 22] of dark exciton lifetimes show that dark excitons actually have rather long lifetimes (∼ 1.5 µs), which serve as a lower bound for exciton spin relaxation, in sharp contrast to previous results.

To solve the controversy, we calculate the first-order phonon-assisted exciton spin relaxation in single self-assembled In$_{1-x}$Ga$_x$As/GaAs QDs using an atomistic empirical pseudopotential method (EPM) [23]. Remarkably, we find that in the Hartree-Fork (HF) approximation, the transition from a bright to dark state is forbidden, suggesting that the transition is induced by Coulomb correlation effects. Sophisticated configuration interaction (CI) [24] calculations suggest that the bright-to-dark exciton transition is on the order of tens of µs in InAs/GaAs QDs, much longer than previous calculations [20] and early experimental values [17, 19], but supported by more recent measurements [21].

In In$_{1-x}$Ga$_x$As/GaAs QDs, the electron-hole exchange interaction splits the ground neutral exciton (X) states into two optically active (bright) states with higher energies and two optically inactive (dark) states with lower energies. Single-dot spectroscopy shows that the typical energy space between bright and dark states, ∆BD, is approximately 100–300 µeV [25]. Because of the asymmetry of the exchange interaction [25, 26, 27], the bright (dark) states further split into two sub-levels B1 and B2 (D1 and D2), as schematically shown in Fig. 1. The energy splitting between B1 and B2, known as fine structure splitting (FSS), is usually a few tens of µeV [25, 26, 27].

FIG. 1: (Color online) Schematic depiction of exciton spin relaxation. τ$^{BD}$ (τ$^{DD}$) is the transition time from bright (dark) to dark (bright) states. τ$^{B0}$ (τ$^{D0}$) is the radiative decay time of the bright (dark) excitons. ∆BD is the exchange splitting between bright and dark excitons.
In QDs, spin–phonon interaction due to SOC can cause spin flip. In excitons, the spin flip results in a transition from a bright to dark exciton and the emission of a phonon or vice versa (see Fig. 1). The exciton spin relaxation rate from a bright (B) to a dark (D) state is given by the first-order Fermi’s Golden Rule:

\[ \frac{1}{\tau_{BD}} = \frac{2\pi}{\hbar} \sum_{\mathbf{q}} |M_{BD}^{\nu}(\mathbf{q})|^2 (N_{\nu,\mathbf{q}} + 1)\delta(\Delta_{BD} - \hbar\omega_{\nu,\mathbf{q}}). \]

where \( N_{\nu,\mathbf{q}} = (e^{\hbar\omega_{\nu,\mathbf{q}}/k_BT} - 1)^{-1} \) is the Bose-Einstein distribution function for phonons. \( \hbar\omega_{\nu,\mathbf{q}} \) is the phonon energy, with \( c_\nu = |\mathbf{q}| \), where \( c_\nu \) is the speed of sound for the \( \nu \) = LA (longitudinal acoustic phonon) and TA (transverse acoustic phonon) modes. Because \( \Delta_{BD} \) is very small, only acoustic phonons are involved in the process. The exciton-phonon-coupling matrix element is given by:

\[ M_{BD}^{\nu}(\mathbf{q}) = \alpha_\nu^e(\mathbf{q}) \langle \Psi_X^D | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_e} | \Psi_X^B \rangle - \alpha_\nu^h(\mathbf{q}) \langle \Psi_X^B | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_h} | \Psi_X^D \rangle, \]

where \( \Psi_X^D \) (\( \Psi_X^B \)) are the bright (dark) state wave functions and \( \alpha_\nu^e(\mathbf{q}) \) (\( \alpha_\nu^h(\mathbf{q}) \)) is the electron(hole)-phonon-coupling strength. We have considered three mechanisms in QDs, including electron(hole)-acoustic-phonon interaction due to (i) the deformation potential \( \nu = \text{LAPD} \), (ii) the piezoelectric field for the longitudinal modes \( \nu = \text{LAZ} \), and (iii) the piezoelectric field for the transverse modes \( \nu = \text{TAPZ} \). Details of \( \alpha_\nu^e(\mathbf{q}) \) and \( \alpha_\nu^h(\mathbf{q}) \) and related parameters can be found in Ref. 4. The overall spin relaxation time from bright to dark states, \( T_1 \), is

\[ 1/T_1 = \sum_{\nu} \sum_{B} \sum_{D} 1/\tau_{BD}^{\nu}. \]

It is essential to have high-quality exciton wave functions to obtain accurate exciton spin relaxation times. In this work, we use EPM to calculate single-particle energy and wave functions. This method has been successfully applied to study the electronic and optical properties of self-assembled In_{1-x}Ga_xAs/GaAs QDs. We simulate lens-shaped In_{1-x}Ga_xAs/GaAs QDs embedded in a cubic GaAs matrix. We obtain the electron and hole energy levels and wave functions by solving the Schrödinger equation via a linear combination of band basis (LCBB) method, in which the SOC is included in the non-local part of the pseudopotentials. When single particle wave functions have been obtained, Slater determinants are built as a basis for excitonic states. The exciton wave functions are obtained via the CI method by expanding them as a linear combination of Slater determinants. The \( \alpha \)-th \( (\alpha = D_1, D_2, B_1, B_2) \) exciton wave function is written as,

\[ \Psi_X^{\alpha}(\mathbf{r}_e, \mathbf{r}_h) = \sum_{v} \sum_{c} C_{v,c}^\alpha \Phi_{v,c}(\mathbf{r}_e, \mathbf{r}_h), \]

where \( N_e \) and \( N_c \) are the numbers of valence and conduction states included in the expansion. The coefficients \( \{C_{v,c}^\alpha\} \) as well as the exciton energies are obtained by diagonalizing the many-particle Hamiltonian in terms of the Slater determinants basis set \( \{\Phi_{v,c}\} \). The exciton energies and wave functions are well converged using \( N_e=20 \) and \( N_c=12 \) (including spin) in our calculations.

Once we have obtained exciton wave functions, the exciton wave function overlap in Eq. (2) can be calculated as

\[ \langle \Psi_X^D | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_e} | \Psi_X^B \rangle = \sum_{v} \sum_{c} (C_{v,c}^D)^* C_{v,c}^B \langle \psi_{c,v}^D | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_e} | \psi_{c,v}^B \rangle, \]

and

\[ \langle \Psi_X^D | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_h} | \Psi_X^B \rangle = \sum_{v} \sum_{c} (C_{v,c}^D)^* C_{v,c}^B \langle \psi_{c,v}^h | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_h} | \psi_{c,v}^h \rangle, \]

where \( \psi_{c,v}^e \) (\( \psi_{c,v}^h \)) is the \( e \)-th (\( h \)-th) electron (hole) wave function.

The single particle matrix elements, \( \langle \psi_{c,v}^e | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_e} | \psi_{c,v}^e \rangle \), are calculated in the Bloch basis of bulk InAs at the \( \Gamma \) point. The bright (dark) exciton wave functions are dominated by configurations in which electron and hole have the same (opposite) pseudo-spin. The mixture of the configurations, in which electron and hole have the same (opposite) pseudo-spin because of heavy hole-light hole mixing, is rather small. Therefore, the matrix elements in Eq. (3) are expected to be very small because, if electrons \( \psi_{c,v}^e \) and \( \psi_{c,v}^e \) have the same pseudo spins, ensuring that \( \langle \psi_{c,v}^e | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_e} | \psi_{c,v}^e \rangle \) is large \((\sim 1)\), \( \langle \psi_{c,v}^e | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_e} | \psi_{c,v}^e \rangle \) is small \((< 0.01)\). However, if electrons \( \psi_{c,v}^e \) and \( \psi_{c,v}^h \) have opposite pseudo spins, \( \langle \psi_{c,v}^e \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_e} | \psi_{c,v}^h \rangle \) is large \((\sim 0.5)\), but the single-particle wave function overlaps, \( \langle \psi_{c,v}^e | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_e} | \psi_{c,v}^e \rangle \), must be very small. The same arguments also apply to the holes.

We start with the simplest case, using only the lowest electron and hole \((N_e=N_c=2)\) states to construct the exciton wave functions, which is equivalent to the HF approximation. Surprisingly, we find that the exciton spin relaxation rate is zero in this approximation. To understand this result, we examine Eq. (4) and Eq. (5) under the HF approximation in greater detail. We first examine the electron part of the exciton wave function overlap, Eq. (5). Under the HF approximation, Eq. (5) can be written as

\[ \langle \Psi_X^D | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_e} | \Psi_X^B \rangle_{HF} = \xi_{11} \sum_{v=1}^{2} (C_{v,1}^D)^* C_{v,1} + \xi_{12} \sum_{v=1}^{2} (C_{v,2}^D)^* C_{v,2}, \]

where \( \xi_{11} = \langle \psi_{c,v}^e | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_e} | \psi_{c,v}^e \rangle = \langle \psi_{c,v}^h | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_e} | \psi_{c,v}^h \rangle \), and \( \xi_{12} = \langle \psi_{c,v}^e | \text{e}^{\text{i} \mathbf{q} \cdot \mathbf{r}_e} | \psi_{c,v}^h \rangle \). \( \psi_{1} \) \((\psi_{2})\) are the electron spin up (down)
wave functions of the lowest energy level. Because $\psi_1^+$ and $\psi_2^+$ are Kramers degenerate states that are related by time reversal symmetry, it is easy to prove that $\xi_{12}=0$. Furthermore, the bright ($\Psi^+_X$) and dark ($\Psi^-_X$) exciton states are orthogonal:

$$\langle \Psi^+_X | \Psi^-_X \rangle_{HF} = \sum_{v=1,2} [(C^D_{v,1})^* C^B_{v,1} + (C^D_{v,2})^* C^B_{v,2}] = 0 \quad (8)$$

By substituting $\xi_{12}$ and Eq. (8) into Eq. (7), we have the electron part of the exciton wave function overlap, $\langle \Psi^+_X | e^{i\mathbf{q} \cdot \mathbf{r}_e} | \Psi^-_X \rangle_{HF}=0$. For the same reason, the hole part of the exciton wave function overlap is $\langle \Psi^+_X | e^{i\mathbf{q} \cdot \mathbf{r}_h} | \Psi^-_X \rangle_{HF}=0$. Therefore, the exciton-phonon interaction matrix element, $M^{BD}(\mathbf{q})=0$, meaning the exciton spin relaxation rate equals zero under the HF approximation. Because $\xi_{12} \sim 1$ is very large, a small un-orthogonality of the exciton wave functions may cause huge errors in the calculated spin relaxation time. As it is prohibited in the HF approximation, exciton spin relaxation is induced by Coulomb correlation effects; therefore, electron/hole correlation effects (via CI calculations) must be included to obtain the correct relaxation time.

The CI-calculated exciton relaxation times of pure InAs/GaAs QDs are approximately 15–55 $\mu$s. The spin relaxation times of alloy QDs are even longer. The exciton spin relaxation times are determined by three factors: (i) $\Delta_{BD}$, which determines phonon momentum $\mathbf{q}$ (smaller $\Delta_{BD}$ leads to a longer spin relaxation time because of lower phonon density), (ii) the electron and hole single-particle wave functions, which determine single-particle relaxation time, and (iii) the exciton CI coefficients $\{C_{e,v}\}$. All three factors are strongly affected by the geometry and chemical composition of the QDs.

We calculate the exciton relaxation time of lens-shaped pure InAs/GaAs QDs as a function of base diameter (height) while keeping height (base diameter) constant. The exciton spin relaxation times as well as $\Delta_{BD}$ are given in Fig. 2 as black lines. The calculated $\Delta_{BD}$ distributes mostly between 100–300 $\mu$eV, which is in good agreement with experimental values [23]. For pure dots, when dot height increases from 2.0 to 5.5 nm, $\Delta_{BD}$ decreases from 330 to 90 $\mu$eV [Fig. 2(a)]. We find that the exciton spin relaxation time is dominated by the hole spin flip. The decrease of $\Delta_{BD}$ tends to slow the spin relaxation time. At the same time, the hole spin flip time drops quickly with increasing dot height [2]. These two factors compete with each other, and the overall effect is that the spin flip time decreases first when dot height changes from 2.0 to 3.0 nm, reaching a relatively constant value as dot height further increases [Fig. 2(c)]. On the contrary, as the base diameter increases from 20 to 30 nm, $\Delta_{BD}$ decreases from 310 to 220 $\mu$eV [Fig. 2(b)], whereas increasing dot diameter also slows hole spin relaxation [2], and the exciton spin relaxation time increases [Fig. 2(b)].

We also calculate the exciton spin relaxation time of lens-shaped alloy In$_{0.7}$Ga$_{0.3}$As/GaAs QDs. The results are shown in Fig. 2 as red lines. These results are similar to those of pure QDs. Generally, the exciton spin relaxation time of alloy dots due to first-order spin-phonon coupling is much longer than that of the pure dots because alloy dots usually have smaller $\Delta_{BD}$. In fact, exciton spin relaxation time is very sensitive to $\Delta_{BD}$, as demonstrated in Fig. 3. We compare the spin relaxation times of two QDs. One QD is a lens-shaped InAs/GaAs dot with diameter=20 nm and height=3.5 nm. The other QD is an alloy dot with the same geometry but with Ga composition of $x=0.3$. As we artificially change $\Delta_{BD}$, the spin relaxation times increase dramatically with decreasing $\Delta_{BD}$, as $T_1 \sim \Delta_{BD}^{-1}$, where $\gamma = 2.9$ for the pure dot and $\gamma = 2.3$ for the alloy dot from the numerical fit.

The exciton dark-bright splitting $\Delta_{BD}$ also determines which mechanism is dominant for the spin flip. Figure 4 depicts the contributions of the three exciton-phonon interaction mechanisms to the total exciton spin relaxation rate as a function of temperature. We take a lens-shaped...
InAs/GaAs QDs with base diameter $b=20$ nm and height $h=3.5$ nm, the spin relaxation rates from all mechanisms increase linearly with temperature, which is the signature of the first-order phonon processes. If we use $\Delta_{BD}=310 \mu$eV, which is given by the EPM calculation, then the LADP mechanism contributes the most to the total relaxation rate, as shown in Fig. 4(a). However, if we (artificially) use a smaller $\Delta_{BD}=200 \mu$eV, then the TAPZ mechanism contributes the most, as shown in Fig. 4(b), because the exciton-phonon coupling strength, $\alpha_{LADP} \propto |q|$, whereas $\alpha_{TAPZ} \propto 1/|q|$. Therefore a smaller $\Delta_{BD}$ makes the TAPZ mechanism dominant.

Two exciton spin relaxation mechanisms have been discussed in the literature, namely exchange interaction [30] and SOC [20]. In QDs smaller than the exciton Bohr radius, it has been suggested that the exchange interaction is most significant in exciton spin relaxation, whereas the SOC mechanism dominates in the larger QDs studied here. The calculated spin-flip time is approximately 2 ns in the In(Ga)As/GaAs QDs at 4 K [20], which is a few orders of magnitude faster than that obtained in the present work. However, in the previous calculations, the SOC were treated perturbatively. Cheng et al. have shown that, in the single-particle case, perturbation theory greatly overestimates the spin relaxation rate [3].

The fast relaxation from perturbation theory is due to the failure of perturbation theory to ensure the orthogonality of both single-particle and many-particle wave functions. The (un-orthogonal wave functions) approximation may not be a serious problem in other calculations; however, it is crucial in the present calculation as well as in the single particle case. Because $\xi_{11} \gg \xi_{12}$, a small error in the orthogonality of the wave functions causes large errors as discussed for the HF approximation above.

The exciton spin relaxation time has been measured by several groups for different QDs. Kurtze et al. found that the spin-flip time is approximately 20 ns at 5 K and 1 ns at 110 K in In(Ga)As/GaAs QDs [12]. Snoke et al. found that the dark-to-bright exciton transition time is 200 ps at $T \sim 10$ K in InP QDs [17]. Johansen et al. found that the relaxation time is approximately 77–167 ns in In(Ga)As/GaAs QDs [18]. These experimental values seem to be in good agreement with previous theoretical results [20], all suggesting that the spin relaxation in excitons is very fast. However, in these experiments, spin relaxation times were extracted from the bright exciton decay time, in which the exciton radiative decay is much faster than the spin relaxation. Therefore, there might be very large errors in estimating the spin relaxation time using bright exciton dynamics. A more accurate method for estimation of the exciton spin relaxation time is to measure the dark exciton lifetime, in which the radiative lifetime is extremely long. Indeed, direct manipulation of dark exciton has recently become possible [21, 22]. The measured dark exciton lifetime exceeds 1.5 $\mu$s at 5 K [21], which is the lower bound for the dark-to-bright exciton transition (Note that at this temperature, $\tau_{BD}/\tau_{DB} \approx 1/2$), ruling out the fast spin relaxation in the exciton. This result is supported by the present calculations.

We would like to note that given the very long exciton relaxation time calculated here, the spin relaxation time through a first-order spin-phonon interaction may not be the dominant mechanism for exciton spin relaxation. The roles of other mechanisms need to be further clarified, including hyperfine and second-order spin-phonon interactions. Nevertheless, the exciton spin relaxation time should be much longer than previously reported, which favors quantum information processing.

To conclude, we present an atomistic pseudopotential calculation of the acoustic phonon-assisted exciton spin relaxation from bright to dark exciton in single self-assembled In$_{1-x}$Ga$_x$As/GaAs QDs. We show that the exciton spin relaxation rate is induced by Coulomb correlation effects. The spin relaxation time calculated is 15–55 $\mu$s in pure InAs/GaAs QDs and even longer in alloy dots. The slow spin relaxation in excitons contradicts previous theoretical and experimental results, which claim a very short exciton spin lifetime, but agrees with more recent experiments.

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