Spin Relaxation Quenching in Semiconductor Quantum Dots

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We have studied the spin dynamics in self-organized InAs/GaAs quantum dots by time-resolved photoluminescence performed under strictly resonant excitation. At low temperature, we observe strictly no decay of both the linear and the circular luminescence polarization. This demonstrates that the carrier spins are totally frozen on the exciton lifetime scale.

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The discrete energy levels in artificial atoms such as semiconductor quantum dots and the corresponding lack of energy dispersion lead to a predicted modification of the spin relaxation dynamics compared to bulk or two-dimensional structures. Extensive experimental and theoretical works in the latter systems have identified the main carrier spin-flip mechanisms, including exchange interaction, motional narrowing due to the lack of inversion symmetry, and mixing between the conduction and valence band states through spin-orbit coupling [1,2]. In quantum dots (QD), all these processes have to be reconsidered since the wave vector k is no longer a good quantum number [3,4]. The absolute lack of energy states between QD energy levels is expected to inhibit not only the elastic processes of spin relaxation but also the inelastic ones such as phonon scattering. The expected long spin relaxation time thus makes QD structures promising candidates for the implementation of spintronic and quantum information processing devices [5,6].

Recent optical pumping experiments have indeed given good indications of a slowing down of the carrier spin relaxation processes in QD compared to bulk or quantum well (QW) structures. The spin orientation in Wurtzite-type nanocrystals has been studied both in continuous wave (cw) and in time-resolved experiments [3,7]. These investigations have revealed the role of both the QD size and the carrier trapping at surface sites on the spin polarization decay time. In single GaAs QD formed by QW interface fluctuations, Gammon *et al.* deduced from cw photoluminescence (PL) experiments that the spin-flip scattering rate was lower than the radiative recombination rate of the ground state [8]. Similar experiments performed in self-organized InAs/GaAs QD lead to the same qualitative conclusion [9]. Recently, Gotoh *et al.* reported on an exciton spin relaxation time of about 900 ps in zero-dimensional InGaAs quantum disks, which is almost twice as long as the radiative recombination lifetime [10]. However, up to now, the QD polarization dynamics have been studied mostly in nonresonant excitation conditions (i.e., photogenerating the carriers in the barrier). The observed spin polarization dynamics of the QD ground state is then the result of all the spin relaxation mechanisms which have occurred in the bulk barrier, in the QD excited states, and finally in the QD ground state, including any spin-flip scattering due to the energy relaxation process itself. In order to study the intrinsic spin dynamics, a strictly resonant excitation of the QD ground state is highly desirable [11]. Note that the free exciton radiative and spin relaxation times in semiconductor QW have only been identified in such clear resonant excitation conditions [2,12,13]. In this Letter, we present a detailed time-resolved investigation of the exciton spin dynamics in self-organized InAs/GaAs QD performed under strictly resonant excitation. These experiments evidence a spin relaxation quenching at low temperature ($T \leq 30$ K). Time-resolved experiments under magnetic field applied along the growth axis (Faraday geometry) allow us to probe the exciton fine-structure without resolving it spectrally.

The investigated structure, grown by molecular beam epitaxy on a (001) GaAs substrate, consists of 5 InAs QD planes embedded in a GaAs λ planar cavity, inserted between two GaAs/AlAs Bragg mirrors [14]. The QD layers are localized near the electromagnetic field antinodes in the microcavity which operates in the weak coupling regime. The QD are obtained after a nominal deposition of 2.2 InAs monolayers. The QD density is $\sim 4 \times 10^{10}$ cm^{-2} per array, and the ground state emission is centered around 1.15 eV with a full width at half maximum (FWHM) of $\sim$60 meV. A variation of the cavity thickness along the radial direction of the wafer allows us to tune the cavity resonance by moving the laser beam across the sample surface. The microcavity is designed so that the cavity mode (FWHM $\sim$3 meV) can be tuned in the energy range of the QD ground state emission. Inserting the QD into a microcavity brings two major improvements with respect to bare QD: (i) the filtering operated by the optical mode results in a reduction of the energetic inhomogeneity of the studied QD; (ii) the narrowing of the radiation pattern emitted by the microcavity allows us to collect the QD emission efficiently in spite of the very small acceptance solid angle ($\sim 10^{-3}$ sr) of the two color up-conversion detection setup we have used [15].

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The sample is excited by 1.5 ps pulses generated by an optical parametric oscillator, synchronously pumped by a mode-locked Ti-doped sapphire laser. The time-resolved PL is then recorded by up-converting the luminescence signal in a $LiIO_3$ nonlinear crystal with the Ti:Sa laser pulses [11,15]. The time resolution is limited by the laser pulse width ($\sim$1.5 ps).

The linear and the circular polarization degrees of the luminescence are defined as $P_{\text{lin}} = (I^X - I^Y)/(I^X + I^Y)$ and $P_C = (I^+ - I^-)/(I^+ + I^-)$, respectively. Here I^X (I^Y) and I^+ (I^-) denote, respectively, the X (Y) linearly polarized and the right (left) circularly polarized luminescence components (X and Y are chosen parallel to the [110] and [1$\bar{1}$0] sample directions). The experiments have been performed at low excitation power ($\sim$7 W cm^{-2}) which corresponds to an average estimated density of photoexcited carriers less than one electron-hole pair per QD. In the following, the laser excitation energy is set to 1.137 eV; it coincides with both the cavity mode and the ground state energy of a family of QD. The detection energy is strictly the same as the excitation one [16].

Let us first recall that in a (InGa)As/GaAs (001)-grown QW, the relevant symmetry is D_{2d}. If the growth direction Oz is chosen as the quantization axis for the angular momentum, the conduction band is s-like, with two spin states $s_{e,z} = \pm 1/2$; the upper valence band is split into a heavy-hole band with the angular momentum projection $j_{h,z} = \pm 3/2$ and a light-hole band with $j_{h,z} = \pm 1/2$ at the center of the Brillouin zone. The heavy-hole exciton states can be described using the basis set $|J_z\rangle \equiv |j_{h,z}, s_{e,z}\rangle$, i.e., $|J_z = 1\rangle \equiv |3/2, -1/2\rangle$, $|J_z = -1\rangle \equiv |-3/2, 1/2\rangle$, $|J_z = 2\rangle \equiv |3/2, 1/2\rangle$, $|J_z = -2\rangle \equiv |-3/2, -1/2\rangle$ (J_z represents the total angular momentum projection of the exciton state on the quantization axis). This basis set is diagonal with respect to the exciton exchange interaction and the twofold degenerate optically active $J_z = |\pm 1\rangle$ states are split from the nonoptically active $J_z = |\pm 2\rangle$ states by the electron-hole exchange interaction energy [17].

In QD structures, the symmetry is lowered and the exchange interaction is thus no longer isotropic [8,18]. The anisotropic exchange interaction splits the $|\pm 1\rangle$ radiative doublet into two eigenstates labeled $|X\rangle = (|1\rangle + |-1\rangle)/\sqrt{2}$ and $|Y\rangle = (|1\rangle - |-1\rangle)/(i\sqrt{2})$, linearly polarized along the [110] and [1$\bar{1}$0] directions, respectively. Continuous wave single dot spectroscopy experiments have clearly evidenced these two linearly polarized lines in self-organized InGaAs QD with an exchange splitting of $\hbar\omega \simeq 150$ μeV [9]. This anisotropic exchange splitting may originate from QD elongation [8,18,19] and/or interface optical anisotropy [18,20].

Figure 1(a) displays the time-resolved PL intensity with polarization parallel (I^X) and perpendicular (I^Y) to the linearly polarized σ^X excitation laser (the initial peak on the I^X luminescence component intensity corresponds to backscattered laser light from the sample surface). The corresponding linear polarization (P_{lin}) kinetics is also plotted. The PL intensity decays with a characteristic time $\tau_{\text{rad}} \sim 800$ ps. After the pulsed excitation, the QD emission exhibits a strong linear polarization ($P_{\text{lin}} \sim 0.75$) which remains strictly constant within our experimental accuracy during the exciton emission (i.e., over $\sim$2.5 ns). From this observation, we can infer that P_{lin} decay time is greater than 20 ns. This behavior differs strongly from the exciton linear polarization dynamics in bulk or type-I QW structures, characterized by a linear polarization decay time of a few tens of picoseconds [1,15]. In the latter structures, it has been demonstrated that any scattering mechanism (phonon, exchange interaction, electron, and/or hole single-particle spin relaxation) results in the destruction of the coherent superposition between the $|1\rangle$ and $|-1\rangle$ components of the linear exciton. The experimental observation of a QD exciton linear polarization which does not decay with time proves that neither the electron, nor the hole spin relax on the exciton lifetime scale.

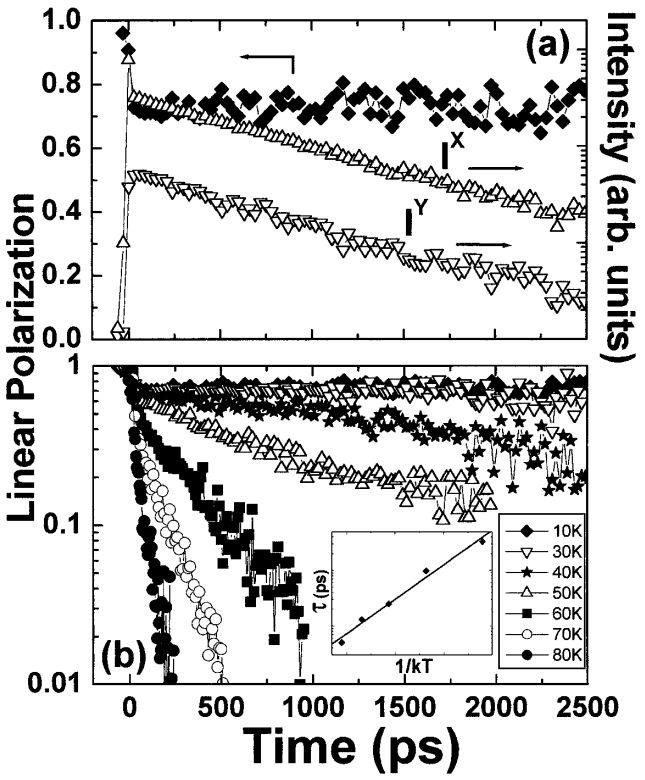

FIG. 1. (a) Time dependence of the PL components copolarized I^X ($\triangle$) and cross polarized I^Y (∇) to the σ^X polarized excitation laser ($T = 10$ K) and the corresponding linear polarization P_{lin} ($\blacklozenge$). (b) Temperature dependence of the linear polarization dynamics. Inset: P_{lin} decay time τ as a function of $1/(k_B T)$.

Figure 1(b) presents the dependence of the exciton PL linear polarization dynamics upon the lattice temperature. A clear temporal decay of P_{lin} is observed above 30 K: the linear polarization decay time drops from $\sim$3500 ps

at 40 K down to 50 ps at 80 K with an activation energy $E_a = 30 \pm 1$ meV [see the inset of Fig. 1(b)]. This very strong P_{lin} temperature dependence can be due either to any electron, hole, exciton spin-flip scattering, or any spin-conserving scattering processes which break the coherent superposition of the linearly photogenerated excitons $|X\rangle = (|1\rangle + |-1\rangle)/\sqrt{2}$. To the best of our knowledge there is no available theory on the exciton spin flip and its temperature dependence in QD. However, one can speculate that the depolarization mechanism is due to hole scattering to higher QD excited states since the measured activation energy is close both to the energy splitting between the ground and first excited heavy-hole state and to the InAs LO phonon energy [21]. In order to clarify the origin of this temperature induced depolarization process, the study of the spin dynamics in *n*- or *p*-doped QD samples is desirable.

A circularly polarized (σ^+) excitation should lead to the observation of circular polarization quantum beats at the pulsation corresponding to the anisotropic exchange splitting. After about 15 ps, the time required for the QD PL signal to overcome the backscattered laser light, we do not observe any polarization at $T = 10$ K and as a consequence any beats in this excitation configuration. This absence of polarization is interpreted as a consequence of the exchange splitting energy statistical fluctuations among the QD whose energy coincides with the cavity mode.

In the following a magnetic field is applied along the growth direction ($B \parallel [001]$). If the Zeeman splitting $\hbar\Omega_z = g\mu_B B$ is much larger than the exchange energy $\hbar\omega$, the QD exciton eigenstates are no longer the $|X\rangle$ and $|Y\rangle$ linearly polarized states but the $|+1\rangle$ and $|-1\rangle$ circular ones (g is the exciton longitudinal Landé factor). We thus expect to observe a circularly polarized PL under σ^+-polarized excitation, as already measured in cw spectroscopy [9,18]. This circular polarization is indeed observed under σ^+-polarized excitation for any magnetic field value up to 5 T (see Fig. 2 where $B = 2.5$ T) [22–25]. Again, the striking feature is the absence of any polarization decay on the exciton emission scale which confirms that the QD exciton spin is totally frozen.

The circular or linear exciton polarization dynamics can be described in the framework of an effective pseudospin with $S = 1/2$ [18]. In this formalism, the exciton states $|+1\rangle$ and $|-1\rangle$ are equivalent to a pseudospin polarized parallel or antiparallel to the z-axis, respectively ($S_z = 1/2$ or $-1/2$), and the linear exciton states $|X\rangle$ and $|Y\rangle$ are described by a pseudospin $S_x = 1/2$ and $-1/2$, respectively. The circular and linear exciton PL polarization writes simply $P_C = 2S_z$ and $P_{\text{lin}} = 2S_x$. The pseudospin Hamiltonian which takes into account the exchange and the Zeeman terms is simply equal to $\mathcal{H} = \hbar/2(\omega\sigma_x + \Omega_z\sigma_z)$, where $\sigma_{x,z}$ are the Pauli matrices [18].

The schematic representation shown in the inset of Fig. 2 shows that the pseudospin rotates around the vector $\vec{\Omega}$ $(\omega, 0, \Omega_z)$ after a circularly polarized excitation characterized by an initial pseudospin $\vec{S}(0)$ parallel to the Oz axis. The projection of $\vec{S}(t)$ on the Oz axis yields the circular PL polarization which should oscillate as a function of time. We do not observe in Fig. 2 any beating structure in the PL circular polarization. As already discussed above, this absence comes from the exchange energy statistical fluctuations among the detected QD. We emphasize that these fluctuations, which lead to the destruction of the PL beats, do not prevent the observation of circular polarization since the mean QD pseudospin S_z is nonzero and tends to $1/2$ when B increases.

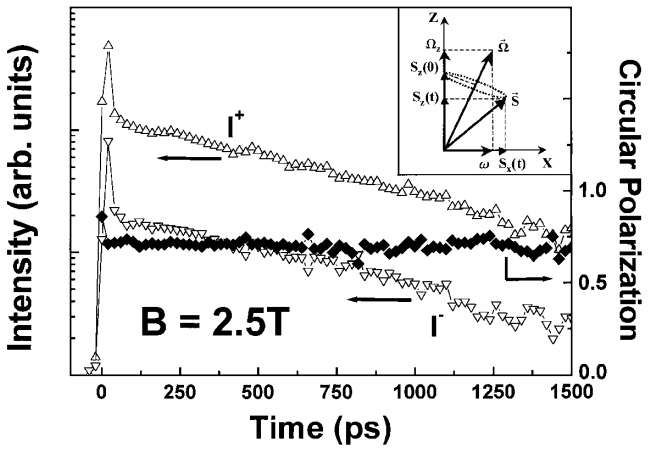

FIG. 2. Time dependence of the PL components copolarized I^+ (△) and counterpolarized I^- (▽) to the σ^+ polarized excitation laser ($T = 1.7$ K) and the corresponding circular polarization P_C (◆). Inset: schematic representation of the exciton pseudospin $\vec{S}$ rotating around the vector $\vec{\Omega}$ after a σ^+ polarized excitation (see text).

Under linearly polarized excitation, the initial pseudospin $\vec{S}(0)$ is parallel to the Ox axis. Increasing the magnetic field yields a reduction of the average value of the PL exciton linear polarization $P_{\text{lin}} = 2S_x$. The observed magnetic field dependence of the PL circular and linear polarization displayed in Figs. 3(a) and 3(b), respectively, confirms this interpretation.

Moreover we see in the inset of Fig. 2 that, under a linearly (circularly) polarized excitation, one expects to observe a nonzero circular P_C (linear P_{lin}) photoluminescence polarization. This conversion from optical orientation to alignment and vice versa, reported in Figs. 3(c) and 3(d), was previously observed for excitons in type-II QW and in InAlAs/AlGaAs QD; it is a direct consequence of the anisotropic exciton exchange interaction [18,20].

The magnetic field dependence of the linear and circular polarization dynamics allows the determination of the exciton fine-structure. Following Dzhioev *et al.,* a quantitative description of these dependences can be given by [18,20]

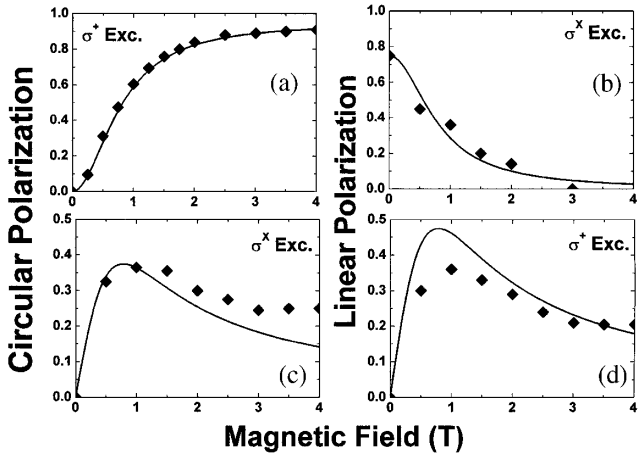

FIG. 3. Magnetic field dependence of the circular (linear) polarization under circularly polarized σ^+ [(a), (d)] and linearly polarized σ^X [(b), (c)] excitations ($T = 1.7$ K). The displayed polarization values are the ones measured at any time delay after the pulsed excitation since we never observe any polarization decay. The solid lines are the best fits obtained with Eqs. (1a)–(1d): $|g|/\tilde{\omega} = 14.5$ ps (see text).

$$P_C \cong P_C^0 \frac{\tilde{\Omega}_z^2}{\tilde{\omega}^2 + \tilde{\Omega}_z^2}, \tag{1a}$$

$$P_{\text{lin}} \cong P_{\text{lin}}^0 \frac{\tilde{\omega}^2}{\tilde{\omega}^2 + \tilde{\Omega}_z^2}, \tag{1b}$$

$$P_C \cong P_{\text{lin}}^0 \frac{\tilde{\omega}\tilde{\Omega}_z}{\tilde{\omega}^2 + \tilde{\Omega}_z^2}, \tag{1c}$$

$$P_{\text{lin}} \cong P_C^0 \frac{\tilde{\omega}\tilde{\Omega}_z}{\tilde{\omega}^2 + \tilde{\Omega}_z^2}, \tag{1d}$$

which correspond, respectively, to the experiments presented in Figs. 3(a)–3(d). Here, $(\tilde{A})^2$ is defined as the statistical average of A^2 over the QD distribution ($A = \omega$, Ω_z); P_C^0 and P_{lin}^0 are fitting parameters which denote, respectively, the circular and linear initial ($t = 0$) polarization.

The four curves (solid lines) displayed in Fig. 3 are the best fits of the experimental data obtained using Eqs. (1a)–(1d) with the following parameters: $P_C^0 = 0.95$, $P_{\text{lin}}^0 = 0.75$, and $|g|/\tilde{\omega} = 14.5$ ps which lead to $\hbar\tilde{\omega} \sim 135$ μeV if we assume that the exciton g factor value measured in Ref. [9] ($|g| \sim 3$) is also relevant for our QD.

We attribute the discrepancy between the fit parameter $P_{\text{lin}}^0 = 0.75$ and the measured initial polarization $P_{\text{lin}}^{\text{las}} = 0.95$ to our very simple approach which ignores any orientation fluctuations from the [110] and [1$\bar{1}$0] eigenstates. These slight fluctuations may originate from statistical fluctuations of the QD shape and orientation [18].

In conclusion, we have studied the linear and circular PL polarization dynamics in self-organized QD under resonant excitation. We never observe at low temperature any measurable temporal decay of the linear or circular luminescence polarization regardless of the excitation polarization and the magnetic field value. These results contrast with the clear spin relaxation previously observed in nonresonant excitation conditions [10]. The main difference between these two types of experiments relies on the higher energy carrier states occupation, including barrier, wetting layer, and QD excited states, which may induce the spin flip of the QD ground state by Coulomb exchange.

This observation of a quenched spin relaxation brings clear experimental support to proposals using electron spins in QD for quantum information encoding and processing in a solid-state system.

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