Dynamics of Coherent and Incoherent Spin Polarizations in Ensembles of Quantum Dots

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The temperature dependence of spin coherence in InGaAs quantum dots is obtained from quantum beats observed in polarization-resolved pump-probe experiments. Within the same sample we clearly distinguish between coherent spin dynamics leading to quantum beats and incoherent long-lived spin-memory effects. Analysis of the coherent data using a theoretical model reveals ≈ 10 times greater stability of the spin coherence at high temperature compared to that found previously for exciton states in four-wave-mixing experiments by Borri *et al.* [Phys. Rev. Lett. **87**, 157401 (2001)]. The data on incoherent polarization reveal a new form of spin memory based on charged quantum dots.

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The temporal coherence of zero-dimensional excitons in quantum dots (QD) is of particular interest for fundamental physics as well as for future applications in quantum logic devices. It has been addressed in coherent control [1] as well as four-wave-mixing experiments [2]. Future quantum devices could also benefit from the expected long spin coherence [3,4].

In many QD systems the lowered symmetry of the confining potential leads to the breakdown of a pure-spin picture of exciton states [1,5–10] leading to the formation of new linearly polarized eigenstates split by the electron-hole exchange interaction [see Fig. 1(a)]. It has been thus suggested that long-lived spin polarization is only likely to be achieved for a single charge carrier in doped dots[11]. The QD exciton emission, on the other hand, can exhibit a long lasting stability of linear polarization at low T after linearly polarized resonant excitation [12]. However, the experiments in Refs. [11,12] do not address spin-coherence effects and deal with incoherent population decay. Spin coherence can be probed only in experiments on dots where a coherent superposition of the linearly polarized eigenstates is excited and the evolution of their relative phase [leading to quantum beats (QB)] is then monitored [1,7,13].

In our work we perform polarization-resolved pump-probe experiments on ensembles of InGaAs QDs. From differential transmission data we deduce both the dynamics of coherent spin polarization manifested in quantum beats [13] and also identify long-lived spin effects due to the incoherent population decay of charged excitons. Our main aim in this work is to study the effect of the coupling of QDs to their environment [2,12,14,15] on the dynamics of coherent and incoherent spin polarizations. We show the robustness of the spin coherence compared to other optical coherence phenomena in the QD system: this is deduced from the order of magnitude longer spin coherence decay (or slower spin dephasing) time we find at high T, as compared to the exciton dephasing measured in four-wave-mixing experiments [2]. We also find a long-lived spin memory in the sample, which is identified as arising due to the slow incoherent decay of the spin-polarized population of charged excitons.

The sample investigated consists of 16 InGaAs QD layers (dot density $\approx 5 \times 10^{10}$ cm^{-2}) grown using molecular beam epitaxy. Each layer is grown in the middle of a 20 nm GaAs quantum well clad by 10 nm thick AlGaAs barriers. The sample was thermally annealed at 700 °C for 12 min [16]. Differential pump-probe transmission $\Delta P/P$ (P—laser power transmitted through the sample) was measured using lock-in techniques. Pulses of a femtosecond Ti-sapphire laser were tuned into resonance with the peak of the QD exciton ground state (GS). In a relatively large range of energies ($\pm$ 15 meV) around

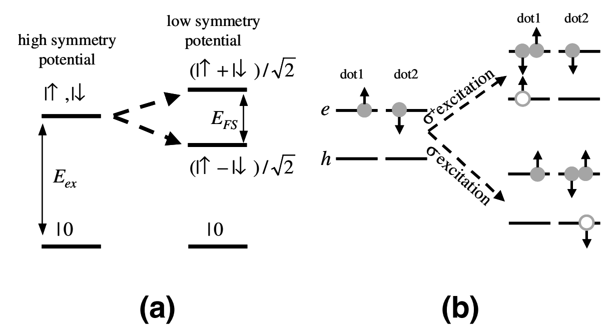

FIG. 1. (a) Schematic diagram of QD exciton states in the case of confining potential of high (left) and low (right) symmetry. $|0\rangle$, $|\uparrow\rangle$, and $|\downarrow\rangle$ denote the initial photon and spin-up and -down exciton states, respectively. In the low symmetry case, mixing of the otherwise degenerate $|\uparrow\rangle$ and $|\downarrow\rangle$ states is observed. Fine structure splitting of the new linearly polarized eigenstates is denoted E_{FS}. E_{ex} denotes the exciton energy. (b) Schematic diagram of the excitation by σ-polarized light of dot 1 (2) charged with spin-up (-down) electron. Dot 1 (2) can absorb $\sigma^+(\sigma^-)$ light only.

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the GS peak the dynamics are similar to those reported here. A notable modification to the dynamics is observed when the laser is tuned >30 meV above the GS peak [17], close to the first excited state. Thus the contribution of the excited state absorption to the data measured at the GS peak is very weak. Either pump or probe pulses excite <0.1 excitons per dot [18], excluding contributions from biexciton effects.

Figure 2(a) shows $\Delta P/P$ pump-probe traces measured at $T = 7$ K with co- and cross-circularly polarized pulses. In the $\sigma^+\sigma^+$ configuration the $\Delta P/P$ signal is maximum at zero pump-probe delay. The signal reaches a local minimum at a delay time $\tau_d \approx 100$ ps, a maximum at $\tau_d \approx 170$ ps, and subsequently decays with a time constant of ≈ 550 ps. The behavior for the $\sigma^+\sigma^-$ configuration is in antiphase to that of the $\sigma^+\sigma^+$ trace for $\tau_d < 170$ ps, with a negligible signal at short delays $\tau_d \approx 0$ and decay with a time constant ≈ 420 ps at long delays.

At long delays the $\sigma^+\sigma^-$ signal is markedly weaker than that in the $\sigma^+\sigma^+$ configuration. This difference results in a "residual" circular polarization or exciton spin memory in the sample. This is seen clearly in Fig. 2(b) where we plot the degree of circular polarization $\Pi = (I_{\sigma^+\sigma^+} - I_{\sigma^+\sigma^-})/(I_{\sigma^+\sigma^+} + I_{\sigma^+\sigma^-})$. Here $I_{\sigma^+\sigma^+}$ ($I_{\sigma^+\sigma^-}$) is the signal intensity in the $\sigma^+\sigma^+$ ($\sigma^+\sigma^-$) configuration. $\Pi \approx 1$ at zero delay and then reaches a minimum of $\Pi = -0.147$ at $\tau_d \approx 90$ ps. At long delays ($\tau_d > 200$ ps) $\Pi > 0.2$ and increases gradually with time.

The oscillatory behavior observed at short delays originates from the fine structure splitting (E_{FS}) of the two linearly polarized QD exciton modes [1,7,13] and can be described within the framework of the quantum beat theory of Ref. [19]. We consider the populations of the two linearly polarized exciton modes split by E_{FS}, which are excited simultaneously with an initial relative phase $\Delta\Theta_0$ by the circularly polarized pump pulse. Following the excitation, $\Delta\Theta$ then oscillates in time with period $2\pi/\omega = h/E_{FS}$. This leads to a periodic variation of the coherent polarization in the sample. In a large ensemble of dots a slight dot-to-dot variation of E_{FS} is present (described in our model by a Gaussian of width δ) leading to a gradual loss of the amplitude in the collective beat pattern. In addition, the phase relation between different dots as well as the periodic evolution of $\Delta\Theta$ within a dot can be perturbed by scattering, such as interactions with phonons, etc. These effects are introduced in the equation below via the polarization coherence time T_{coh}. The carrier population decay is given by τ_X, describing the incoherent contribution to the observed signal. We thus obtain

$$I_{\sigma^+\sigma^+/\sigma^+\sigma^-} = e^{-t/\tau_X} \pm e^{-t/T_{coh}} e^{-\delta^2 t^2} \cos(\omega t). \quad (1)$$

Equation (1) gives $P_{\sigma^+\sigma^+} \approx P_{\sigma^+\sigma^-} \approx e^{-t/\tau_X}$ at long delays. However, the residual polarization observed in Fig. 2 indicates that in addition to the dots exhibiting the oscillating behavior there is a subset of the dot ensemble with no oscillatory component, i.e., for which $\omega \approx 0$. We attribute the nonoscillating contribution to dots containing a charge carrier prior to the pump excitation, arising from the carriers ionized from impurities in the AlGaAs barriers. This attribution is based in part on cw single-dot experiments, which showed the absence of the fine structure for charged excitons [8,9], for which the electron-hole exchange interaction is blocked by the presence of an additional carrier. This situation is illustrated in Fig. 1(b) [20]. As seen from the figure for dot 1 (2) the $\sigma^+(\sigma^-)$-polarized excitation creates a charged exciton, where due to the Pauli principle the electrons form a spin singlet state and hence the exchange interaction in such a *e*-*h* complex is suppressed [8,9].

It is notable in Fig. 1(b) that only dot 1 (2) can be excited with a $\sigma^+(\sigma^-)$-polarized pulse. At the same time the creation of a charged exciton in dot 1 by a σ^+-polarized pump pulse will not change the absorption of a σ^--polarized probe pulse, since the absorption of the latter pulse in dot 1 was blocked even before the pump excitation. Thus, the signal $I_{\sigma^+\sigma^+}$ measured from the whole ensemble of dots has contributions from both neutral and charged dots, while $I_{\sigma^+\sigma^-}$ originates from neutral dots only. The contribution from the charged dots to $I_{\sigma^+\sigma^+}$ is taken into account by adding a term $A_{chX}\exp(-t/\tau_{chX})$ to Eq. (1). Here A_{chX} is half of the weight of charged dots in the ensemble [20] and τ_{chX} is the charged exciton lifetime. It also follows that τ_X in Eq. (1) can be measured directly from the decay of $I_{\sigma^+\sigma^-}$ at long delays and corresponds to the neutral exciton lifetime.

As seen from Eq. (1) the damping of the quantum beats is strongly dependent on the spin polarization coherence

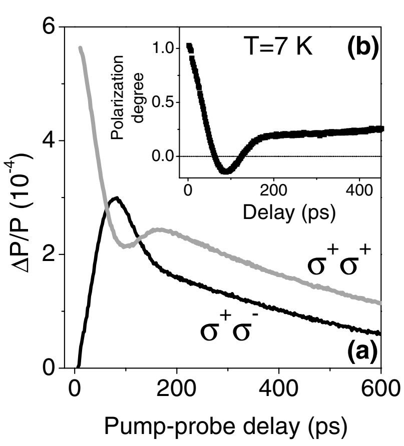

FIG. 2. (a) Dependence of the differential transmission on the pump-probe delay measured at $T = 7$ K for σ^+-polarized pump and σ^+- or σ^--polarized probe (gray and black lines, respectively). Both ≈ 100 fs pulses are tuned to the QD ground state. (b) Temporal dependence of the degree of σ^+ circular polarization after excitation of the sample with a σ^+-polarized pulse at $T = 7$ *K*.

time $T_{\rm coh}$, which implies that the measurement of QBs can serve as a method to study the spin coherence and, in particular, for our purposes, its temperature dependence. An enhanced damping of the beat pattern with increasing T is observed in Fig. 3. A small change in the QB pattern is seen in the whole range of $7 < T < 55$ K, with the decay of the beats governed by the dot-to-dot variation of $E_{\rm FS}$. However, at $T > 60$–65 K a progressively weaker oscillation pattern is seen. At $T \geq 90$ K the beats are washed out and only a strong initial peak (minimum) is observed in $I_{\sigma^+\sigma^+}$ ($I_{\sigma^+\sigma^-}$).

The black curves in Figs. 3(a) and 3(d) show the results of fitting using a modified Eq. (1) with the contribution of charged excitons taken into account. From the low T data we determine δ and ω, which are then kept unchanged for the higher T fits. The change of the other parameters with T is deduced from the experimental results [21].

Although the Gaussian distribution of $E_{\rm FS}$ used in Eq. (1) is a reliable starting point, the actual physical picture can be more complex, as has been discussed for quantum wells in Ref. [22]. From Eq. (1) it is notable that the form of the distribution of $E_{\rm FS}$ could affect mainly $T_{\rm coh}$, the latter influencing the decay of the beat pattern. However, we find that even when using a Lorentz distribution, magnitudes of $T_{\rm coh}$ within 20% of those obtained with a Gaussian distribution are found. The weak sensitivity to the form of the distribution of $E_{\rm FS}$ arises from the relatively small width of the distribution in our case as can be concluded from the fact that unlike in Nickolaus *et al.* [22] quantum beats are observed in our experiments.

Using a Gaussian distribution of $E_{\rm FS}$ we find that the oscillation period corresponds to $E_{\rm FS}$ ($\hbar\omega \pm \delta/2$) of 18 ± 3 μeV [23]. At low temperature the damping of quantum beats is determined by the distribution of $E_{\rm FS}$ with the width of ≈ 6 μeV (see Fig. 4). Above 40 K temperature-induced dephasing starts to contribute to the damping of the beat pattern. Eventually, at 130 K $T_{\rm coh}$ falls to 28 ps. At $T \geq 65$ K the temperature dependence of $T_{\rm coh}$ can be fitted with an ionization-type exponent with activation energy $E_A \approx 22.5$ meV (see inset in Fig. 4), similar to the 30 meV found in measurements of linear polarization decay due to population redistribution in QDs [12]. The magnitude of $T_{\rm coh}$ observed at $T = 130$ K is about 1 order of magnitude larger than that reported at high T for neutral exciton dephasing in four-wave-mixing experiments [2]. This observation indicates the robustness of spin coherence in the QD system.

Equation (1) describes also the long-lived spin memory related to incoherent population decay. The variation of $\tau_{{\rm ch}X}$ and τ_X with temperature is shown in Fig. 4. At $T \leq 35$ K $\tau_{{\rm ch}X} \approx 1300$ ps and is ≈ 3 times longer than $\tau_X = 420$ ps. This difference in incoherent population decay rates is responsible for the long-lived circular polarization (or exciton spin orientation) in the sample excited by the pump beam [see Fig. 2(b)]. As the temperature reaches 50 K a sharp drop in $\tau_{{\rm ch}X}$ to 500 ps is observed. In the whole range $T \geq 50$ K, $\tau_{{\rm ch}X}$ is smaller than τ_X, with the latter growing with T at $T > 35$ K. This in turn leads to decay of the residual circular polarization with time. This is seen, for example, in the inset of Fig. 3(c) at $T = 80$ K, in marked contrast to the growing polarization observed at $T = 7$ K in Fig. 2(b).

The observed temperature dependence arises from the onset at $35 < T < 50$ K of the efficient thermal excitation of carriers from the QD ground state, which is dominated by the hole activation [14,15]. Other mechanisms such as hole spin flip have also been reported for n-type charged II-VI dots [24], which are, however, likely to be less

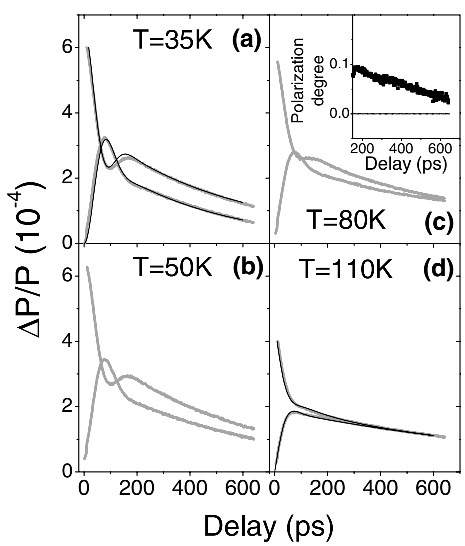

FIG. 3. Dependence of the differential transmission on the pump-probe delay measured at (a) $T = 35$ K, (b) $T = 50$ K, (c) $T = 80$ K, and (d) $T = 110$ K for σ^+-polarized pump and $\sigma^\pm$-polarized probe. The inset in (c) shows the temporal dependence of the degree of circular polarization in the sample at $T = 80$ K.

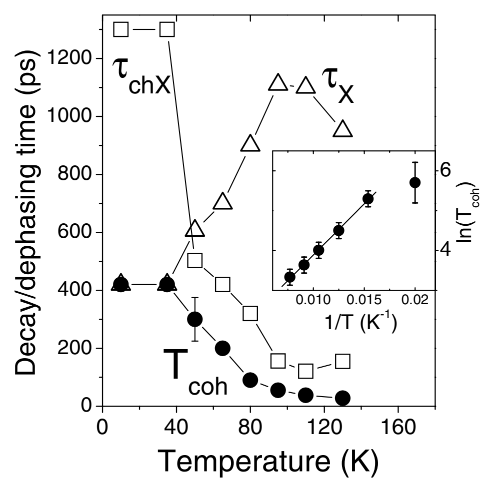

FIG. 4. Temperature dependence of $T_{\rm coh}$ (circles), $\tau_{{\rm ch}X}$ (squares), and τ_X (triangles). Inset shows logarithm of $T_{\rm coh}$ versus $1/T$. The fit is a linear function with slope of ≈ 260 K (22.5 meV).

significant in our case. In our samples the charged excitons are most likely p type, due to the well-known residual p doping in molecular-beam epitaxy grown GaAs structures. In addition, the thermal activation time at $T \approx 70$ K reported from measurements of photoluminescence [14] and photocurrent [15] on individual dots is >10 times faster than spin flip reported in Ref. [24].

The decrease of τ_{chX} indicates the shortening of the time during which the ground state is occupied simultaneously by three carriers, which in turn leads to a faster decrease of the degree of circular polarization in the sample. The increase of τ_X with T is explained by the increased probability of excitation of carriers to excited states and the consequent relaxation to lower energy, thus leading to an increase in the exciton lifetime relative to the radiative recombination time at low T ($\tau_X = 420$ ps at $T \leq 35$ K). The slight deviation from the trend at $T > 110$ K may arise from irreversible excitation of carriers from the dot (leading to the fall in τ_X) and/or contribution from increasingly faster electron dynamics. Because of the complex population dynamics neither the τ_X nor τ_{chX} T dependence can be fitted with a single activation energy. The magnitude of $\tau_{chX} \approx 1300$ ps, observed below the onset of the strong thermal activation, represents the radiative lifetime of charged excitons.

In conclusion, dynamics of spin coherence were studied in a wide range of temperatures in an ensemble of QDs by measuring quantum beats in pump-probe experiments. We find gradual shortening with T of T_{coh} (to 28 ps at $T = 130$ K), which can be fitted by a single energy activation-type process. The same set of experimental data reveals a long-lived spin memory in the sample arising from the slow incoherent decay of the exciton population in charged dots, where the spin oscillation is suppressed. We note that the spin polarization memory can be maximized by employing doped dots and in addition can be controlled by using gated structures.

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