Optically Driven Spin Memory in *n*-Doped InAs-GaAs Quantum Dots

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We show that the spin state of the resident electron in an *n*-doped self-assembled InAs-GaAs quantum dot can be *written* and *read* using nonresonant, circularly polarized optical pumping. A simple theoretical model is presented and accounts for the remarkable dynamics producing counterpolarized photoluminescence.

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Because of their atomiclike energy spectrum, self-assembled quantum dots (QDs) have a strong potential for the realization of an elementary quantum gate in condensed matter [1]. In particular, the electron spin in a QD is a good candidate for a quantum information carrier because a long coherence time is anticipated, as a result of the inhibition of classical spin relaxation mechanisms [2]. For instance, spin coherence times up to 3 ns were reported for electrons in CdSe QDs [3]. In undoped QDs, the electron-hole pair fine structure is strongly affected by the interplay between the exchange interaction and the reduced QD symmetry (usually C_{2v}). As a result, the excitonic ground state is characterized by two linearly polarized eigenstates [4–6], split by an anisotropic exchange energy. Recent experiments performed under strictly resonant excitation [7] demonstrated the stability of the photoluminescence (PL) linear polarization (i.e., optical alignment) during the whole radiative lifetime up to a temperature of 30 K, and rapid damping of the circular polarization. Under nonresonant excitation conditions, the circular orientation of excitons also experiences a fast decay [8,9] ($\tau \approx 300$ ps).

In this Letter, we address the new topic of optical manipulation and dynamics of electronic spin in *n*-doped QDs that contain one resident electron. First, we show that the spin of the electron trapped in such a QD can be modified by photoinjection of a spin-polarized electron-hole pair, which results in *writing* the spin state. Next, we probe the degree of spin polarization of the electron remaining after the photoinjected electron-hole pair has recombined, which corresponds to *reading* the quantum information associated with the electron spin.

We studied simultaneously QD samples with and without *n* doping. They both consist of 20 layers of lens-shaped self-assembled InAs/GaAs QDs separated by 40-nm-thick GaAs layers (for a review on QD growth, see Ref. [10]). The *n*-doped sample contains a Si-delta doping layer 2 nm below each QD layer with a nominal donor concentration of 8×10^{10} cm^{-2}. The average QD density is about 4×10^{10} cm^{-2}, but it seems that most of the QDs contain a single electron: Observation of intraband absorption [11] indicates that the doping is effective. Conversely, we observe ground state luminescence under strictly resonant excitation (with the up-conversion PL technique [8]), which proves that the spin-degenerated electronic *S* states are not fully occupied. The low temperature (10 K) luminescence is centered around 1.15 eV, which corresponds to a typical dot size (as estimated within an effective mass model) of 2 nm in height and 11 nm in radius. Such a dot size implies the existence of at least three shells of confined electronic states. As shown in Fig. 1, under cw excitation in the InAs wetting layer (WL) (1.44 eV), the doped sample luminescence exhibits a remarkable *negative* degree of circular polarization

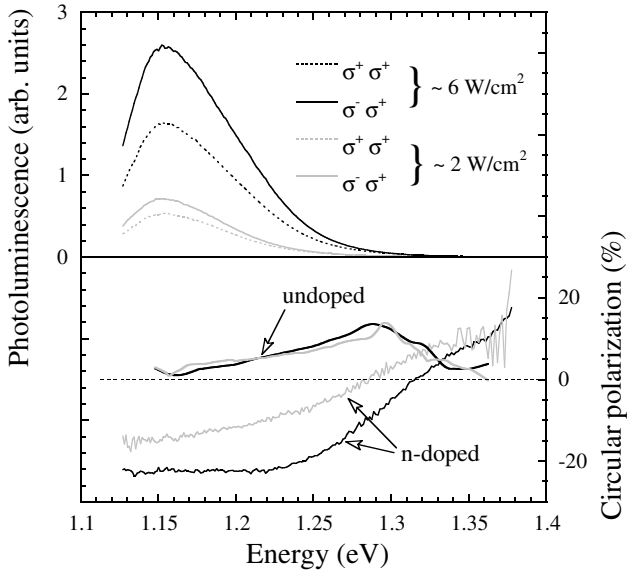

FIG. 1. Upper curves: polarization-resolved photoluminescence spectra at $T = 10$ K of the *n*-doped sample under WL excitation with σ^+-polarized light for two different powers. Lower curves: corresponding circular polarization ratios together with the undoped sample's ratios measured under equivalent excitation conditions.

defined as $(I_{\sigma^+\sigma^+} - I_{\sigma^+\sigma^-})/(I_{\sigma^+\sigma^+} + I_{\sigma^+\sigma^-})$, where $I_{\sigma^+\sigma^{+(-)}}$ denotes the PL intensity excited with σ^+ polarization and detected in $\sigma^{+(-)}$ polarization. This optical pumping rate increases (in absolute value) up to -22% when increasing excitation power from $\sim 2\ \mathrm{W\,cm^{-2}}$ to $\sim 6\ \mathrm{W\,cm^{-2}}$, which corresponds to average excitation densities less than one photoinjected pair per dot. On the contrary, the undoped sample luminescence under the same excitation conditions shows a weak positive circular polarization (+ 6%) with weak power dependence. The counterpolarized luminescence in *n*-doped QDs is also observed when exciting in the GaAs barrier at 1.55 eV. Hence it is not due to details of the WL band-edge structure that would (unlikely) favor a light-hole optical transition. The short rise time of the luminescence signal (30 ps) proves the short capture time in these QDs and supports the assumption that a photoinjected electron keeps its original spin during the capture process and first stages of thermalization. On the other hand, hole spin relaxation is known to be much more efficient, in bulk or 2D heterostructures [12,13]. For this reason, we consider that heavy holes are completely depolarized before their capture in the QDs. Besides, observation of a 50% circular polarization of the WL luminescence, when exciting in the GaAs barrier, is consistent with this assumption. These simple observations suggest that *n*-doped QDs behave like *spin memories:* the spin polarization of the electron that remains in a dot influences the polarization of the luminescence of an injected electron-hole pair, giving rise to the observed power dependence.

Much more is learned by examining the dynamics of this mechanism on the picosecond time scale. The time dependence of the luminescence and circular polarization of the ground state emission under excitation in the WL is shown in Fig. 2. We used a 80 MHz pulsed Ti:Sa laser in ps mode and a single color, luminescence up-conversion technique [8]. The collected luminescence was detected for each delay both in co- and counterpolarized geometry with respect to the exciting beam. The dynamics presents two regimes. During the first 10 ps (inset of Fig. 2), while the luminescence intensity is rising, we observe a positive polarization, up to +20%, that decreases and reverses sign with a typical decay time of 10 ps. This process is followed by a slow evolution of the negative circular polarization during the whole radiative lifetime, down to -45%. The luminescence signal is measured up to 2 ns after the excitation pulse and shows an exponential decay with a typical lifetime of 800 ps. The polarization is observed up to 30 K with no change in the dynamics and then slowly vanishes when increasing the temperature above 100 K. When performing the same experiment under excitation in the GaAs barrier, we observe the same polarization dynamics, with typical polarization reduced by half due to absorption by both heavy- and light-hole transitions.

From the theoretical point of view, it is rather surprising to observe a *negative* circular orientation when both

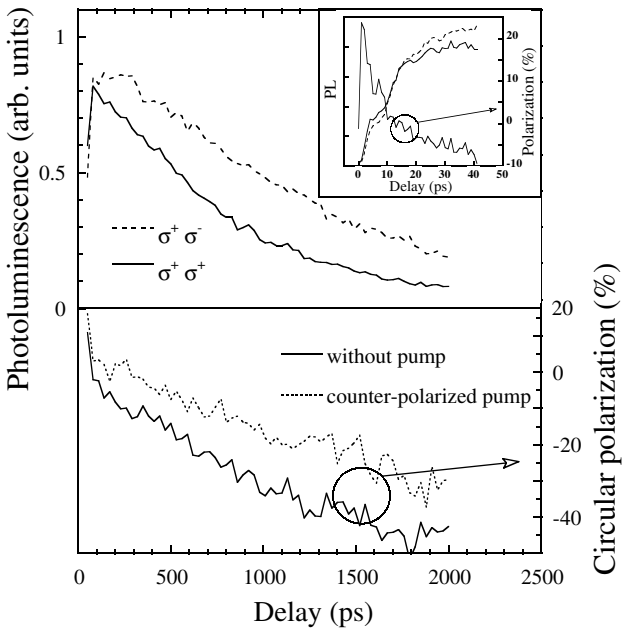

FIG. 2. Top: time-resolved photoluminescence detected at 1.15 eV under circularly polarized excitation at 1.44 eV of the *n*-doped QD sample. The inset shows a blowup of the fast dynamics at short times. Bottom: the corresponding circular polarization is shown (dark line), as well as the polarization in the presence of a counterpolarized pump pulse 10 ns earlier (grey line).

exciting and recombining transitions imply the same bands (here, Γ_6 electrons and heavy holes). To interpret consistently the whole set of experimental results, we propose a scenario providing a simplified description of the three-particle complex (or *trion*) dynamics under the driving forces acting on spins in this system: exchange interaction between carriers and spin-orbit interaction. The reduced C_{2v} symmetry of a quantum dot is a major ingredient, since the total angular momentum is no longer a good quantum number. Yet, the conduction (valence) single particle states, which are not eigenstates of the angular momentum L_z, can still be described to zeroth order, as the S_c (S_v) and P_c (P_v) 2D atomiclike orbitals. In the following, we use this convenient labeling for the *actual* orbitals.

For simplicity, we consider that the dots contain ideally a single resident electron and that at most a single electron-hole pair is optically injected in a dot during the radiative lifetime. We assume that the heavy holes photogenerated in the wetting layer are completely depolarized before they are captured by the QDs [12–14]. When a photocreated pair is captured, the injected electron (say with spin $\downarrow$) relaxes in a few ps down to the $P_c^{\downarrow}$ states, while the heavy hole relaxes down to its ground state. Direct Coulomb terms merely change the discrete single particle energies by some meVs, while exchange terms introduce a spin-dependent interaction in the effective Hamiltonian. Clearly, the strongest exchange term is the electron-electron one (typically $K_{\mathrm{e\text{-}e}} = 5$ meV [15]),

while electron-hole exchange (Δ_0) is a small correction to it. Hence the correct physical picture of a trion state is a spin-correlated electron pair perturbed by exchange interaction with a heavy hole.

Figure 3 shows the corresponding fine structure, where the relevant two-electron states are the triplet states T_1, T_0, T_{-1} and the excited singlet S_0^*, formed with a pair of P_c and S_c electrons, and the ground state singlet S_0 formed with two S_c electrons. The index denotes the total-spin projection on the growth axis. By combination with the two hole states ($S_v^{\uparrow \text{or} \downarrow}$), we obtain ten trion states. Let us remark that electron states with zero total spin (i.e., T_0 and S_0^*) are expected to thermalize very rapidly to the ground state ($\tau_{\text{s.c.}}$ in the ps range) due to an efficient phonon-assisted spin-conserving mechanism [16].

We can distinguish two equiprobable situations: (i) When the resident and photoinjected electrons have antiparallel spins, they form either the T_0 or S_0^* states. In both cases, the spin orientation of the injected electron is definitely lost and the radiative recombination yields completely depolarized luminescence due to the random hole spin orientation. (ii) When both electrons have parallel spin (say $\downarrow\downarrow$ for σ^+ excitation), it forms the T_{-1} state which cannot relax to the ground singlet S_0 without spin flip. In the next two paragraphs, we discuss specific spin-flip mechanisms that lead to the relaxation of this trion and to the observed luminescence polarization.

Depending on the hole spin, the hot trion formed with T_{-1} electrons is either *bright* or *dark* (see Fig. 3). The bright trion can recombine radiatively with σ^+ polarization and leaves a $P_c^{\downarrow}$ electron (the latter will further thermalize on the $S_c^{\downarrow}$ state). This explains our observation of copolarized luminescence at the very beginning of the recombination. But, another dynamical process can occur for this bright hot trion, driven by the electron-hole *anisotropic* exchange interaction (AEI). In neutral QDs, the latter splits the bright excitonic states ($J_z = \pm 1$) into linearly polarized states, by up to 150 μeV [5,17,18]. Here, it implies a coupling of the bright trion $T_{-1} \otimes S_v^{\uparrow}$ to the $S_0^* \otimes S_v^{\downarrow}$ trion, opening an efficient thermalization path towards $S_0 \otimes S_v^{\downarrow}$. In practice, everything behaves just as if the hole and one electron undergo a simultaneous spin reversal or *flip-flop* process, followed by an irreversible relaxation down to $S_0 \otimes S_v^{\downarrow}$. The radiative recombination of the resulting frozen trion produces then counterpolarized light, due to the hole spin state. This path being much faster than the direct recombination as soon as the coupling is of the order of a few μeVs, we assign this mechanism to the reversal of luminescence polarization after a few tens of picoseconds.

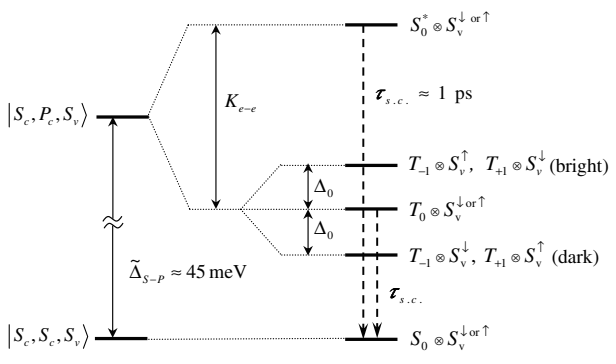

FIG. 3. Hot trion fine structure in a symmetrical QD. $\tilde{\Delta}_{\text{S-P}}$ is the energy separation between excited and ground trion states, corrected by the direct Coulomb terms. The electron-hole exchange interaction (Δ_0) slightly splits the triplet-based states.

A similar mechanism might be expected to occur in the case of the *dark* trion $T_{-1} \otimes S_v^{\downarrow}$ and would lead to copolarized emission. However, the AEI amplitude for a dark exciton is much weaker: experimentally, the corresponding splitting cannot be systematically measured [6,19], while theoretically, the long range exchange interaction which mainly contributes to the AEI of bright excitons vanishes for dark excitons as reported for localized 2D excitons [20]. From these considerations, we can reasonably assume this term to be on average 2 orders of magnitude smaller and hence discard the related mechanism. In fact, the most likely process that allows relaxation of this dark trion is the single particle spin flip of an electron under the effect of spin-orbit (SO) interaction and dot shape anisotropy. Indeed, if the so-called S_c and P_c orbitals are elongated along $\langle 110 \rangle$ directions, the SO interaction couples the electronic T_{-1} triplet and the S_0 singlet, allowing a phonon-assisted relaxation [2] to the bright trion $S_0 \otimes S_v^{\downarrow}$ again. We assign this electron spin-flip relaxation to the slow increase of counterpolarization during the recombination lifetime.

Averaging over the different relaxation paths, we finally conclude that the spin of the electron remaining in the dot after recombination of the photoinjected electron-hole pair is polarized, with an increase of the spin $\downarrow$ population (for σ^+ excitation). Assuming a long spin relaxation time for a resident single electron, this model agrees with the observed increase in absolute value of optical orientation when increasing the excitation power (see Fig. 1), as the generation of antiparallel spins becomes less probable after a few cycles.

In order to demonstrate the electronic spin memory effect we measured the spin lifetime of the resident electrons using a pump-probe experiment: the circularly polarized pump pulse spin-polarizes the QD electrons nonresonantly and the linearly polarized probe pulse triggers luminescence, reflecting the spin polarization of the ground state. To extend the measurement to long delays, we used laser diodes delivering 4-ns pulses with a repetition rate of 80 kHz, exciting carriers in the WL band edge at 1.41 eV. Their relative delay was electronically controlled with a resolution of 2 ns. The probe beam was chopped mechanically, and the time-integrated, polarization-resolved luminescence was detected with a standard lock-in method at 1.15 eV. The probe power is approximately half the pump power and lies within the low excitation regime of Fig. 1. For the *n*-doped sample,

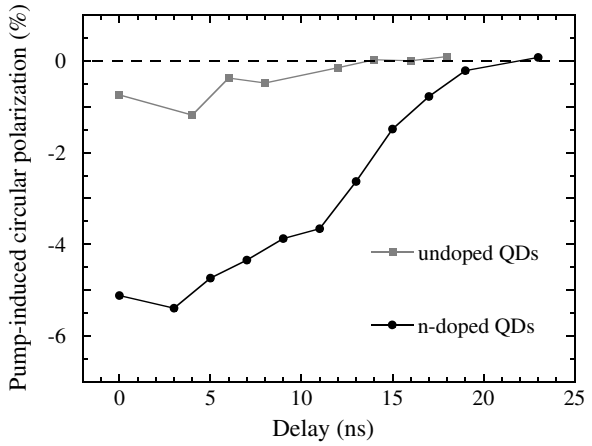

FIG. 4. Circular polarization of the probe luminescence induced by a 4-ns σ^+ pump pulse. For the *n*-doped sample, the spin orientation decay time of the resident electrons is about 15 ns.

we clearly observe a pump-induced circular polarization, as shown on Fig. 4, proving the *writing* of electron spins. This effect, which amounts to -5% at zero delay, is still about -4% at 10 ns when the two pulses no longer overlap, and the spin can still be *read* at a delay of 15 ns, a long time after the optical recombination of the pairs excited by the pump. We attribute this decay time (15 ns) to the typical spin coherence time of an ensemble of resident electrons. In contrast, for the undoped sample the probe luminescence shows almost no pump-induced polarization.

Although the result of the previous experiment clearly shows the memory effect, the measurement itself is hampered by the time integration over a complex dynamics. To overcome this difficulty, we used the time-resolved luminescence setup again in a pump-probe configuration. The time-resolved probe luminescence was measured with or without a counterpolarized pump at the fixed delay of 10 ns [21]. The lower part of Fig. 2 shows that the main effect caused by the counterpolarized pump is to modify the polarization maximum from -45% to -30% at the end of the recombination lifetime. Averaging over the radiative lifetime, we obtain a reduction of the time-integrated polarization of about 10%, which corresponds to -5% of pump-induced optical orientation, in good agreement with the measurements performed with pulsed diodes at 10-ns delay.

In conclusion, we have demonstrated that it is possible to manipulate the spin of the resident electron in an *n*-doped QD using nonresonant optical excitation. A long lasting spin memory effect was observed. A simplified model accounting for the remarkable dynamics producing negative optical orientation was discussed. Development of this work will aim at identifying the mechanism limiting the single resident electron spin lifetime and at manipulating entangled spin states in QD based systems.

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