Optically Driven Spin Memory in \( n \)-Doped InAs-GaAs Quantum Dots


1Laboratoire de Physique de la Matière Condensée de l’Ecole Normale Supérieure, 24 rue Lhomond, 75005 Paris, France
2CNRS-Laboratoire de Photonique et Nanostructures, Route de Nozay, 91460 Marcoussis, France
3Laboratoire de Physique de la Matière Condensée, INSA-CNRS, 335 Avenue de Rangueil, 31077 Toulouse Cedex, France

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

DOI: 10.1103/PhysRevLett.89.207401

PACS numbers: 78.47.+p, 71.70.-d, 72.25.Fe, 73.21.La

Because of their atomic-like 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 \textit{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 \textit{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 \( 8 \times 10^{10} \text{ cm}^{-2} \). The average QD density is about \( 4 \times 10^{10} \text{ cm}^{-2} \), 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 \textit{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 \( \sigma^{+} \)-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 $\sigma^+$ polarization and detected in $\sigma^+(-)$ polarization. This optical pumping rate increases (in absolute value) up to $-22\%$ when increasing excitation power from $\sim 2\ \text{W cm}^{-2}$ to $\sim 6\ \text{W cm}^{-2}$, which corresponds to average excitation densities less than one photojected pair per dot. On the contrary, the undoped sample luminescence under the same excitation conditions shows a weak positive circular polarization ($\pm 6\%$) with weak power dependence. The countercircular 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 photojected 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 exciting and recombinining transitions imply the same bands (here, $\Gamma_6$ 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 photocreaited 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 $K_{ee} = 5$ meV [15]),
while electron-hole exchange ($\Delta_0$) 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 $P_e$ and $S_e$ electrons, and the ground state singlet $S_0$ formed with two $S_e$ electrons. The index denotes the total-spin projection on the growth axis. By combination with the two hole states ($S_h^{0,\pm}$), we obtain ten trion states.

Let us remark that electron states with zero total spin (i.e., ground state ($/0.0028$)), both cases, the spin orientation of the injected electron is completely depolarized luminescence due to the random interaction couples the electronic and dot shape anisotropy. Indeed, if the so-called $S_e$ and $P_e$ 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 $S_0 \otimes S_h^+$ again. We assign this electron spin-flip relaxation to the slow increase of counterpolarization during the recombination lifetime. A similar mechanism might be expected to occur in the case of the dark trion $T_{-1} \otimes S_h^+$ 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 $S_e$ and $P_e$ 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 $S_0 \otimes S_h^+$ 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 photojected electron-hole pair is polarized, with an increase of the spin $\downarrow$ population (for $\sigma^+$ 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,
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.

We thank A. Vasanelli for her $\mathbf{k} \cdot \mathbf{p}$ band structure calculations and K. Kavokin for fruitful discussions. The LPMC-ENS is “Unité Associée au CNRS et aux Universités Paris 6 et Paris 7.” Two of us (G.B. and R.E.) are supported by a NEDO grant and by the E.E.C. IST project SQUID.

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

*Present address: CEA/DRFMC/SP2M/PSC, 17 avenue des Martyrs, 38054 Grenoble, Cedex 9, France.


[21] Note that due to the high 80 MHz repetition rate of the Ti:Sa laser, the probe acts on the system just like a copolarized pump for itself, at a 12.5 ns delay.