Electrical Control of Hole Spin Relaxation in Charge Tunable InAs/GaAs Quantum Dots

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We report on optical orientation of singly charged excitons (trions) in charge-tunable self-assembled InAs/GaAs quantum dots. When the charge varies from 0 to -2, the trion photoluminescence of a single quantum dot shows up and under quasiresonant excitation gets progressively polarized from zero to $\sim$100%. This behavior is interpreted as the electric control of the trion thermalization process, which subsequently acts on the hole-spin relaxation driven in nanosecond time scale by the anisotropic electron-hole exchange. This is supported by the excitation spectroscopy and time-resolved measurements of a quantum dot ensemble.

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Although the usual spin relaxation mechanisms of electrons in semiconductors are theoretically quenched in zero-dimensional systems because of their localization [1], the electron confinement in a quantum dot (QD) gives rise itself to a new efficient mechanism via the hyperfine interaction with the subsystem of nuclear spins in the dot [2–4]. This effect could be a dramatic drawback in the perspective of using the spin of electron in QDs for quantum information processing, particularly in the case of InAs/GaAs self-assembled QDs where a spin relaxation time of a few hundreds ps is calculated [4]. Even if more experimental work is required to confirm this conclusion, it is interesting to consider as an alternate candidate a valence band hole, which because of its p-like symmetry at atomic scale, does not interact with the nuclear spins. Besides, in self-assembled InAs QDs, the hole ground state is a nearly pure heavy hole (with angular momentum $J_z = \pm\frac{3}{2}$) which should significantly limits its spin relaxation usually based on the mixing with the light-hole band ($J_z = \pm\frac{1}{2}$).

In this work we studied self-assembled QDs embedded in a $n-i$-Schottky diode, a system allowing efficient control of the QD charge [5–8]. By charging QDs with a single excess electron in order to form under interband optical excitation *trions* (i.e., a singly charged electron-hole pair itself named *exciton* in the following), we can study the spin state of the photo-hole without being perturbed by the electron-hole exchange interaction. Indeed, in the trion ground state, the latter vanishes because of the singlet configuration of the doubly occupied S_e-electron ground state. As a result, the fine structure of the optically active excitons, which is observed in QDs with no rotational symmetry [9] and produces the quantum beats of exciton spin [10,11] disappears for a trion. The circular polarization of a negative (positive) trion photoluminescence (PL) reflects thus the spin state of the hole (electron) as already reported for CdSe QDs [12]. In this Letter, we report on optical orientation experiments under quasiresonant excitation of individual and ensemble InAs/GaAs QDs. We particularly focus on the regime of the negative trion X^- luminescence and show that its circular polarization strongly depends on the QD excess charge (1 or 2) before the optical excitation. We discuss these observations as the consequence of the hole-spin dynamics driven by the anisotropic exchange interaction which can be switched on and off by electrically controlling the QD charge. Time-resolved optical orientation of a QD ensemble supports our interpretation, and straightforwardly demonstrates the quenching in nanosecond time scale of hole-spin relaxation for ground state trions.

We studied two samples (A and B) grown using molecular beam epitaxy on a [001]-oriented semi-insulating GaAs substrate. Their structure is similar to the one studied in Ref. [5]. It consists of a 200 nm-thick n^+-GaAs layer followed by a 175 nm-thick intrinsic GaAs/AlGaAs/GaAs multilayer, which contains the InAs QDs 25 nm above the n^+-GaAs layer. Sample A has a low QD density ($\leq 10^9$ cm^{-2}) allowing single QD spectroscopy. The QD charge is controlled by a bias V_g between a Schottky contact (a semitransparent NiCr film) and a deep ohmic contact on the sample surface. For spectroscopy of individual QDs, we used 1 μm-diameter optical apertures through an opaque metallic mask evaporated on the Schottky contact.

Although the charging of a single QD is best determined by optical spectroscopy (see further Fig. 2), we first discuss this mechanism with the capacitance-voltage curve shown in Fig. 1 in order to stress out an important feature of such a system. When the gate voltage V_g varies from -0.2 to $+0.35$ V, the capacitance increases by a step reflecting the filling of the QD ground state. Its position at $V_g \approx 0$ V agrees fairly well with the electron confinement energy $\sim$150 meV below the GaAs conduction band, the 0.8 eV Schottky barrier and the structure lever arm $L = 7$ as discussed in [13]. The lower part of Fig. 1 shows a schematic diagram of the electron occupation N for a single QD. The charging $N-1 \rightarrow N$ occurs when the potential μ_E of the n$^+$ electrode reaches the QD chemical potential $\mu_{\mathrm{QD}}(N-1)$. The latter can be seen as the single particle energy of one electron added in the QD charged with $N-1$ electrons [14] (e.g., $\mu_{\mathrm{QD}}(0) = E_S$). Similarly, when the dot

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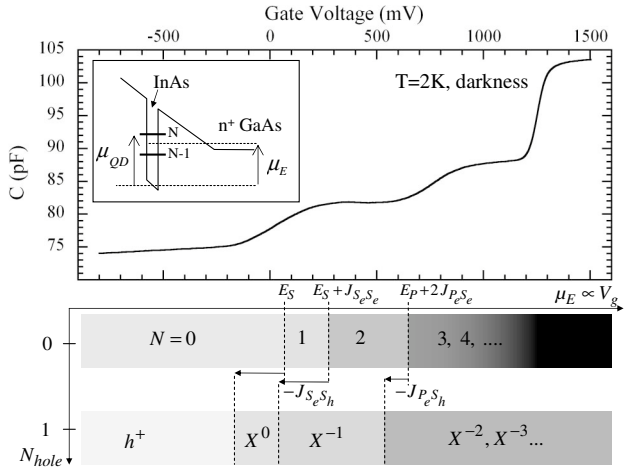

FIG. 1. C-V characteristic of sample B. Inset: conduction band diagram in the vicinity of the QD layer. Bottom: schematic diagram of the charging of a single QD, in darkness ($N_{\text{hole}} = 0$) and in the presence of a photocreated hole ($N_{\text{hole}} = 1$) as a function of the bias.

has captured a photocreated hole under optical excitation, we can deduce the stability diagram of the charged excitons X^{-n}. To the first order, the domains are simply shifted by the direct electron-hole Coulomb interaction (denoted $J_{S_eS_h}$ or $J_{P_eS_h}$) as illustrated in Fig. 1. A remarkable feature shown by this diagram is the wide stability domain of the trion complex X^- as a consequence of the large energy separation $E_P - E_S$ (~60 meV) between the S_e and P_e electron levels. Hence, the trions can form within a voltage range where the QD equilibrium occupation N is 1 or 2. As we show in the following, the trion spin dynamics depends on this initial charge.

Optical orientation experiments have been performed at low temperature (<20 K). The polarization of the excitation is set to σ^+, and the PL polarization σ^+ or σ^- is analyzed by a quarter-wave plate placed in front of a fixed Glan-Thomson linear polarizer. The whole setup provides a rejection ratio better than 5% in $\sigma^+\sigma^-$ configuration, over the spectral region of interest. The luminescence is spectrally filtered by a double monochromator and detected by a cooled charge coupled device array.

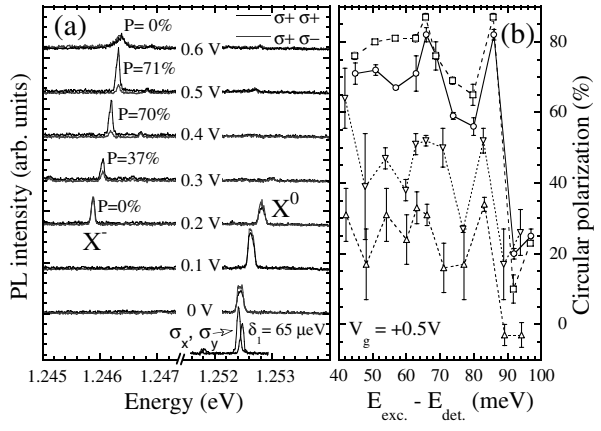

FIG. 2. (a) Polarization-resolved PL spectra of a single QD under σ^+ excitation at 1.32 eV. (b) Polarization of 4 trion lines as a function of the excitation energy. Error bars correspond to the deviation between two successive measurements.

Under nonresonant excitation at $h\nu \sim 1.45$ eV, a few tens meV above the band edge of the wetting layer (WL) 2D-continuum, the heavy-hole band with $J_z = \pm\frac{3}{2}$ is selectively excited because of the strong biaxial strain of the InAs WL which splits the heavy- and light-hole bands. In principle, this gives rise to a fully spin-polarized population of photoelectrons with, as a result, a significant PL circular polarization (defined as $(I_{\sigma^+} - I_{\sigma^-})/(I_{\sigma^+} + I_{\sigma^-})$ where $I_{\sigma^{+(-)}}$ denotes the PL intensity measured in $\sigma^{+(-)}$ polarization). We actually measure a weak polarization that slightly increases up to ~10% when applying a positive bias in the 0.5–1 V range. This outcome is due to the thermodynamic equilibrium through a tunnel barrier (see Fig. 1 inset) between InAs (QDs and WL) and the adjacent $n^+ -$ GaAs layer: spin-polarized electrons photocreated in the WL relax by fast tunneling into GaAs, within a time $\tau_{\text{tun.}} < 1$ ps for $V_g < 0.5$ V (estimated with a simple 1D-model as in Ref. [15]). Photocreated holes which relax their spin orientation before capture recombine thus with unpolarized electrons mainly brought by the $n^+ -$ GaAs electrode. Note that this strikingly contrasts with the *negative* circular polarization we previously reported for chemically n-doped QDs [16] where a hot $P_e - S_e$ electron pair could be formed within the QDs during the photoelectron relaxation. In order to reduce this effect, we consider in the following the regime of quasiresonant (or *intradot*) excitation corresponding to a laser energy fixed at least 50 meV below the WL interband transition.

Figure 2(a) shows the polarization-resolved PL spectra of an individual QD for different gate voltages. The X^0 neutral exciton is basically not polarized like the trion feature X^- which appears at $V_g = 0.2$ V when this QD starts being filled with one electron. The identification of theses lines is confirmed by the characteristic trion redshift of ~5 meV [5,7,17] and by the electron-hole exchange induced splitting δ_1 which vanishes for a trion. When increasing the bias, the trion gets highly polarized up to 70% and then above 0.5 V starts broadening while its polarization simultaneously collapses. All the trion lines we measured through a 1 μm-size aperture exhibit this remarkable dependence, with yet a scatter of the maximum polarization in the 30–85% range, that we attribute to the random perturbation produced by a small aperture on the PL emission of a given QD [18]. The precise *intradot* excitation energy did not turn out to be critical in order to observe this effect. This is illustrated in Fig. 2(b) which shows the circular polarization of 4 different single trion lines at 0.5 V as a function of the excitation detuning. The polarization remains large on a broad excitation spectral range (40 meV) which rules out any field induced optical resonance. In conclusion the measurements on single QDs

reveal that the trion optical orientation strongly depends on the bias, which controls the QD equilibrium occupation N before the optical excitation. The maximum is reached when the quantum dots are filled with two electrons, as expected $L/e \times J_{S_e S_e} \sim 175$ mV above the bias of X^- appearance.

In the following, we explain how the applied bias acts on the hole-spin relaxation by controlling the trion thermalization, a term used to describe all processes (including tunneling) that the carriers undergo to establish the QD ground state. We first note that the quasiresonant excitation with a detuning in the 50–70 meV range involves mixed transitions [19] (i.e., transitions from discrete states to a continuum) that we assign here to the transitions from a confined hole state to the conduction quasi-2D continuum. The conjugate process implying the hole 2D-continuum and the electron ground state can be reasonably discarded because holes photocreated in the WL escape into GaAs due to the internal electric field, and thus cannot contribute to the PL [20]. Therefore, the photo-hole is assumed to be created on a QD discrete level and to relax (if necessary) on the QD ground state without losing its optical spin orientation, whereas the dynamics of the photoelectron depends on the electric bias, as illustrated in Fig. 3. The bias determines the initial charge state ($N = 1$ or $N = 2$) as well as the final state ($X, X^-, \ldots$) of the thermalization before the optical recombination (see Fig. 1). During this thermalization, the hole spin can relax because of the anisotropic exchange interaction (AEI) with electrons. The latter is responsible in anisotropic QDs for the splitting of the excitons with angular quantum number $j = \pm 1$ into linearly polarized eigenstates $|X\rangle$, $|Y\rangle$. The energy splitting δ_1 amounts to a few tens μeV for the InAs/GaAs system, and could be measured in sample A for $\sim 1/3$ of the investigated single QDs with values in the 30–80μ eV range [see, e.g., Fig. 2(a)]. This gives rise to spin quantum beats of excitons photocreated as a coherent superposition of $|X\rangle$ and $|Y\rangle$ ([10,11]), with a period $\tau_{\mathrm{AEI}} = h/\delta_1$ falling in the 5–100 ps range. For a trion in its ground state, this interaction vanishes because of the 2-electron singlet configuration. Therefore, for $N = 1$ [Fig. 3(a)], the hole-spin dynamics is driven by AEI with the nonthermalized electron pair (or with the single S_e electron) before being quenched as soon as the trion has reached its ground state. This produces the complete hole-spin relaxation observed at the threshold of trion appearance [Fig. 2(a) at 0.2 V], when its thermalization time amounts about to the exciton radiative lifetime ($\tau_r \sim 1$ ns). For $N = 2$ [Fig. 3(b)] and as long as the field is above $\sim$25 kV/cm, the photogenerated electron rapidly tunnels out from the InAs WL layer before the hole spin flips under AEI, leaving the dot with a spin-polarized trion. This simple model explains the bias-dependent polarization of the trion emission, reaching a maximum when the dot is initially occupied by two electrons.

In spite of the inhomogeneous distribution of InAs QDs in size and composition, which produces in the bias domain a broadening of the QD charge state, the trion orientation can still be evidenced on a QD ensemble. Figure 4(a) shows the polarization-resolved PL excitation spectra of sample B at different gate voltages. We mainly focus here on the phonon-assisted excitation resonance observed 35 meV above the detection energy between the GaAs LO- and TO-phonon features. Remarkably its polarization goes from $\sim$5% to basically 100% when the bias varies from 0.2 to 0.5 V, i.e., again when the QDs are progressively filled with electrons. Although the precise nature of this 1LO phonon resonance is still a matter of investigations (see, e.g., [21]), this strong polarization indicates very likely the *instantaneous* quenching of the hole-spin relaxation in a trion *resonantly* excited in its ground state. For higher energy excitation, the PL excitation spectra consists of several steplike transitions which can be ascribed to free-to-bound transitions [19]. Their polarization shows the same bias dependance as the single

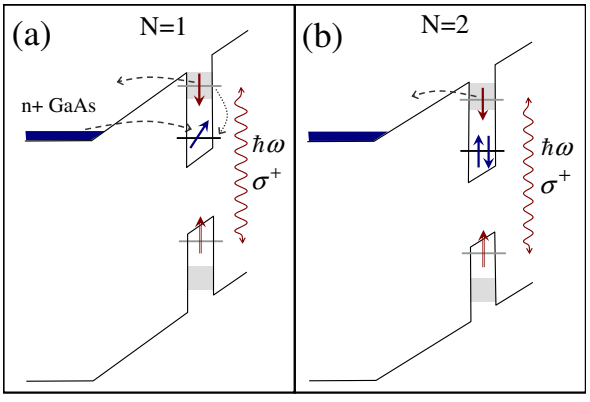

FIG. 3 (color online). Sketch of the charged exciton thermalization dynamics in a QD for $N = 1$ (a) or $N = 2$ (b) electrons before the quasiresonant σ^+ excitation. The gray-shaded areas represent the WL 2D-continuum of the QDs. In panel (a), the dotted and dashed arrows illustrate two possible paths for the photoelectron.

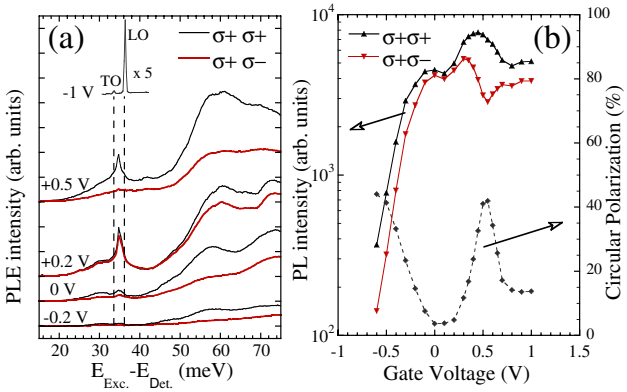

FIG. 4 (color online). (a) Excitation spectra of sample B PL detected at 1.3 eV. The GaAs LO- and TO-phonon features (dashed lines) were identified at -1 V by Raman diffusion in $\sigma^+\sigma^-$ configuration. (b) PL and polarization at 1.265 eV under excitation at 1.34 eV against gate voltage.

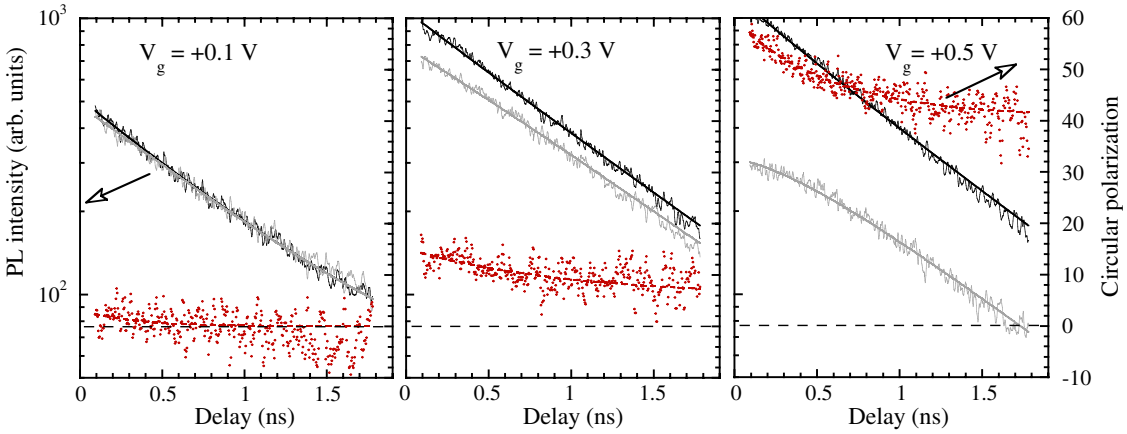

FIG. 5 (color online). Polarization- and time-resolved PL intensity at 1.27 eV (solid lines) in $\sigma^+\sigma^+$ (black) and $\sigma^+\sigma^-$ (gray) configurations, and circular polarization (dots) with a multiexponential fit (dashed line) of sample B under excitation at 1.34 eV.

QDs (weak at 0.2 V and strong at 0.5 V), with moreover a significant polarization for biases below 0 V. A similar dependance is shown in Fig. 4(b) ,which reports the PL intensity and polarization as a function of the bias for a QD ensemble [22]. The resonance associated to the QD filling with two electrons is still observed at 0.5 V. In addition, the polarization of excitons (weak at 0 V) increases for a negative bias. The latter effect actually occurs in the regime where the PL intensity decreases due to the field and could be associated from micro-PL spectroscopy (not shown here) to the emission of positive trions X^+.

To check the consistency of our interpretation and, in particular, the quenching of the spin orientation of thermalized trions, we performed time-resolved spectroscopy of sample B. A standard macro-PL setup equipped with a streak camera synchronized to a mode-locked Ti:Sapphire laser in the ps mode is used together with a simple monochromator for the detection providing a time resolution of 50 ps. Figure 5 gives the main results. At 0.1 V ($N = 0$) the PL of excitons is almost unpolarized during all the recombination (even for the smallest accessible delays $\sim$100 ps) as expected for an ensemble of QDs, due to AEI. At 0.3 V, when the stable complex starts to be predominantly a trion, a significant polarization emerges with a long-lasting component which reaches a maximum at 0.5 V ($N = 2$). In this regime, a multiexponential fit of the polarization gives a hole-spin relaxation time larger than 20 ns.

In conclusion, we have performed optical orientation of negative trions in single and ensemble charge-controlled InAs/GaAs QDs. The circular polarization of trion PL is controlled by the electric bias, with a maximum close to 100% when the dots are just filled with two electrons. This is interpreted as the control of the trion thermalization time during which the electron-hole exchange interaction drives the hole-spin relaxation. This electrical control of optical orientation as well as the measurement of a hole-spin lifetime above 20 ns indicates the absence of other efficient mechanisms, such as phonon-mediated spin flip due to heavy-hole light-hole mixing. This suggests to consider holes as alternative candidates to electrons for spin-based memories and for quantum information processing using spin in QDs.

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