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Dynamic nuclear polarization of a single charge-tunable InAs/GaAs quantum dot

B. Eble,[1] O. Krebs,[1,*] A. Lemaître,[1] K. Kowalik,[1,2] A. Kudelski,[1] P. Voisin,[1] B. Urbaszek,[3] X. Marie,[3] and T. Amand[3]
[1]*CNRS Laboratoire de Photonique et Nanostructures, Route de Nozay, 91460 Marcoussis, France*
[2]*Institute of Experimental Physics, Warsaw University, Hoża 69, 00-681 Warsaw, Poland*
[3]*Laboratoire de Nanophysique Magnétisme et Optoélectronique, INSA, 31077 Toulouse Cedex 4, France*

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We report on the dynamic nuclear polarization of a single charge-tunable self-assembled InAs/GaAs quantum dot in a longitudinal magnetic field of ~0.2 T. The hyperfine interaction between the optically oriented electron and nuclear spins leads to the polarization of the quantum dot nuclei measured by the Overhauser shift of the singly charged excitons (X^+ and X^-). When going from X^+ to X^-, we observe a reversal of this shift, which reflects the average electron spin optically written in the quantum dot either in the X^+ state or in the final state of X^- recombination. We discuss a theoretical model which indicates an efficient depolarization mechanism for the nuclei limiting their polarization to ~10%.

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The spin dynamics of an electron confined in a self-assembled semiconductor quantum dot (QD) is currently the subject of intense research.[1–7] It indeed represents a promising direction for implementing quantum computation algorithms in the solid state, because once the electron is confined in a quantum dot, its spin dynamics at low temperature is *almost* no longer subjected to the random perturbations which lead to relaxation and decoherence in bulk or quantum wells. For example, the usual spin relaxation due to spin-orbit interaction turns out to be quite negligible.[1,8] The subsisting sources of relaxation that have been identified in real QDs are (i) the exchange interaction with additional hole(s) or electron(s)[6] and (ii) the hyperfine interaction with the QD nuclear spins.[3,4] In order to address this issue with optical techniques, field-effect structures embedding charge-tunable[9] QDs offer an amazing potential: the exchange-induced spin relaxation can be kept under control, e.g., by extracting the hole from a photoexcited electron-hole pair[1] (called an exciton) or by adding a charge preventing exchange from operating,[6,10] while the effective role of the hyperfine interaction which only affects the conduction band electrons can be investigated by controlling the nature (electron or hole) of the spin-polarized carrier.

In this Rapid Communication we address the issue of hyperfine interaction in a self-assembled charge-tunable InAs/GaAs quantum dot submitted to an external magnetic field of ~0.2 T. Optical excitation with circularly polarized light is used for writing the electron and/or hole spins, whereas an external bias applied to the *n*-Schottky-type sample controls the electronic charge.[6,9,11,12] The same single QD has been studied in three different regimes: when the electron spin $\hat{S}^e$ interacting with the nuclear spins $\hat{I}^j$ (i) forms with two photoexcited holes a positive trion X^+, (ii) forms a neutral exciton X^0, and (iii) results from the radiative recombination of a negative trion X^- made of two electrons and one hole. By measuring the Overhauser shift of the X^+ or X^- Zeeman splitting,[13–15] we show that the small applied magnetic field leads to optically induced polarization of the QD nuclei, with a nonlinear dependence on the average electron spin $\langle \hat{S}_z^e \rangle$ deduced from the photoluminescence (PL) circular polarization. Remarkably, this shift changes sign on crossing from X^+ to X^-. We present a theoretical description of the nuclear polarization dynamics in InAs/GaAs QDs explaining most of our results.

The sample which has already been used in Ref. 6 was grown by molecular beam epitaxy on a [001]-oriented semi-insulating GaAs substrate. The InAs QDs are grown in the Stranski-Krastanov mode 25 nm above a 200-nm-thick n^+-GaAs layer and capped by an intrinsic GaAs (25 nm)/$Al_{0.3}Ga_{0.7}As$ (120 nm)/GaAs (5 nm) multilayer. The QD charge is controlled by an electrical bias applied between a top Schottky contact and a back Ohmic contact. We used a metallic mask evaporated on the Schottky gate with 1-μm-diameter optical apertures to spatially select single QDs. Figure 1 shows the $T=5$ K PL contour plot of intensity against bias and detection energy of the single QD that has been extensively studied in this work. The identification of the different spectral lines and of the associated QD charge relies on several robust observations. (i) Between 0 and ~0.15 V we observe PL emission from only the ground-state exciton X^0, clearly identified by its fine structure[16] [Fig. 2(b)] and the biexciton (hardly perceptible in Fig. 1) appearing under stronger excitation at lower energy. (ii) Above 0.15 V the X^- trion redshifted by ~6 meV shows up, indicating the charging of the QD with an electron.[6,17,18] Both lines (X^0 and X^-) still coexist because under non-strictly-resonant excitation (here 1.31 eV) a single photohole can be

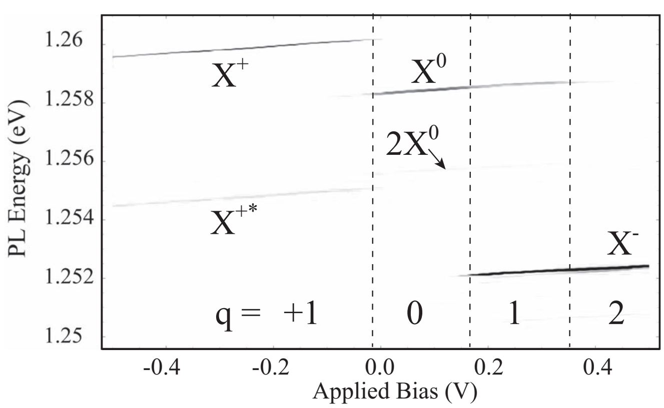

FIG. 1. Gray-scale contour plot of the PL intensity from a single InAs QD at $T=5$ K versus the detection energy and applied bias under intradot excitation at 1.31 eV.

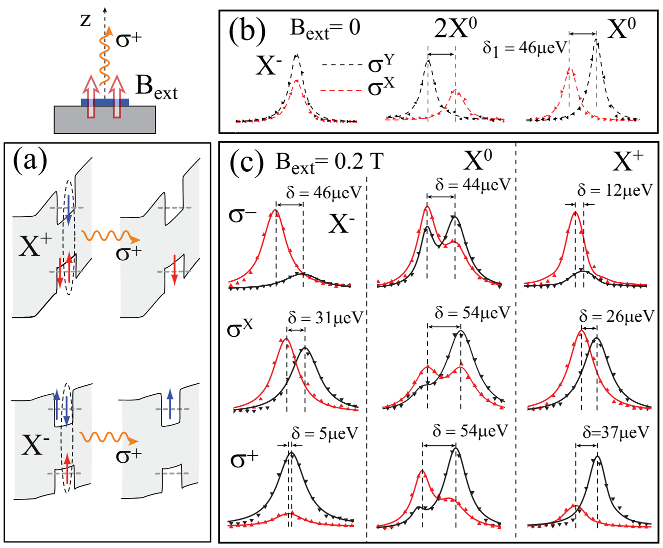

FIG. 2. (Color online) (a) Sketch of the spin configuration for X^+ and X^- for σ^+ emission. (b), (c) Zoom over a 200 μeV range of PL spectra excited at 1.34 eV in zero and 0.2 T magnetic field, respectively. (b) Detection in linear polarization. (c) Detection in σ^- [red (gray)] and σ^+ (black) for three different excitation polarizations as indicated on the left.

created in the QD, giving rise to optical recombination with the resident electron. (iii) Above 0.35 V, the neutral exciton line definitely disappears, indicating occupation with two electrons. (iv) For negative bias a symmetrical charging effect occurs for holes. The neutral exciton line, which disappears as a result of the electron tunneling out of the QD, is replaced by a 3 meV blueshifted line assigned to the trion X^+.[12,19] Although the applied bias controls only the conduction band chemical potential and thus cannot itself generate the QD charging with holes, this effect is achieved under strong intradot excitation. It directly creates a hole within the QD, which does not escape like the electron because of its larger effective mass.

To study the influence of hyperfine interaction on spin dynamics, optical orientation experiments have been performed in the presence of a small longitudinal magnetic field parallel to the QD growth axis z.[2,13,20] The field was provided by a permanent magnet simply put below the sample within the cryostat cold finger. Its amplitude B_{ext} at the sample position was estimated to ≈ 0.2 T. We used a standard micro-PL setup based on a $\times 50$ microscope objective, a double spectrometer of 0.6 m focal length, and a nitrogen-cooled charge-coupled device array detector, providing a spectral resolution of 30 μeV and a precision in line position of about 1 μeV after deconvolution by a Lorentzian fit. The optical excitation and detection were both performed along the z axis. Thus the degree of PL circular polarization defined by $\rho_c=(I_{\sigma^+}-I_{\sigma^-})/(I_{\sigma^+}+I_{\sigma^-})$, where $I_{\sigma^{+(-)}}$ denotes the PL intensity measured in $\sigma^{+(-)}$ polarization, traces the average spin $\langle S_z^e\rangle=-\rho_c/2$ of the electron participating in the PL signal.[20] This results from the usually accepted assumption of a pure heavy-hole ground state with angular momentum projection $m_z=\pm\frac{3}{2}$ in InAs QDs leading to optically active electron-hole pairs $|\pm 1\rangle=|\mp\frac{1}{2},\pm\frac{3}{2}\rangle$. As a result, depending on the QD charge state, we are able to read out either the spin of the electron for both X^+ and X^0 states, or the spin of the hole in X^- from which we deduce the electron spin $\langle S_z^e\rangle=+\rho_c/2$ left in the QD after the optical recombination, as sketched in Fig. 2(a).

Figure 2 shows the influence of the applied magnetic field on the (charged) exciton fine structure. In InAs QDs, the electron-hole exchange interaction leads to the splitting of X^0 between the dark states (angular quantum number $J=2$) uncoupled to light and the bright states ($J=1$) which form the X^0 line observed in PL experiments. Due to the in-plane anisotropy of real QDs the latter is further split by energy $\delta_1\sim 50$ meV into $|X\rangle$ and $|Y\rangle$ eigenstates linearly polarized along the crystallographic axes $\langle 110\rangle$.[16,21] This is illustrated in Fig. 2(b) which presents linear-polarization-resolved spectra in zero magnetic field. The X^0 line is split by $\delta_1=46$ μeV and the biexciton $2X^0$ shows the same splitting with reversed sequence of polarization.[22] In contrast, the trion lines X^+ and X^- are not split, in agreement with the vanishing of the exchange interaction between one electron (hole) and two holes (electrons) forming a spin singlet in the ground state.[23] In the magnetic field, the Zeeman interaction separates the $\sigma^{\pm}$-polarized components of the trion lines by $\delta_Z=|g_X|\mu_B B_{ext}$, where g_X is the exciton g factor (supposed to be constant for the three lines considered here) and $\mu_B=58$ μeV/T is the Bohr magneton. It also increases the bright X^0 splitting to $\sqrt{\delta_1^2+\delta_Z^2}$. Figure 2(c) shows the PL spectra resolved in circular polarization. Under linearly polarized excitation we find for the three lines a Zeeman splitting $\delta_Z\approx 28$ μeV in agreement with an exciton g factor of ~ 3.[16] Under circularly polarized excitation a significant deviation from the sole Zeeman interaction is now observed: the X^+ splitting gets larger in σ^+ excitation by $+10$ μeV and smaller in σ^- by -15 μeV. This so-called Overhauser shift (OHS) denoted δ_n indicates the polarization of the QD nuclear spins which progressively builds up through the hyperfine interaction with the optically oriented electrons in the QD. Remarkably a symmetrical but reversed effect occurs for X^- with a shift $\delta_n=+15$ μeV in σ^- and $\delta_n=-25$ μeV in σ^+, whereas the PL from X^- and X^+ shows the same helicity. This OHS reversal demonstrates that in the case of a spin-polarized X^- for which the total electron spin is zero, the mechanism leading to nuclear polarization does not operate during the X^- lifetime but takes place due to the interaction with the single electron left in the QD after the optical recombination. This contrasts with the results reported for GaAs QDs.[15] For X^0, which still shows a weak polarization at 0.2 T, we observed no significative OHS except when there is an overlap with X^+ [the situation of Fig. 2(c) because of excitation at higher energy], in which case it only acts as a probe of the OHS produced by X^+. Another feature of these results is the pronounced OHS asymmetry when changing the excitation from σ^+ to σ^-. This clearly appears in Fig. 3 which reports the bias dependence of the circular polarization and of the trion spin splitting. This asymmetry means that polarizing the nuclear spins in the direction that produces a larger effective field for the electron is more difficult than in the opposite direction. The total electron spin splitting represents indeed the main energy cost of the electron-nucleus flipflop process (the Zee-

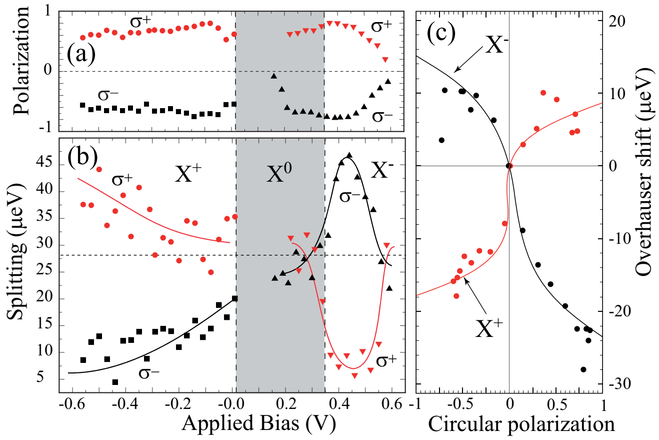

FIG. 3. (Color online) (a) Circular polarization and (b) spin splitting of X^+ and X^- PL lines against applied bias at B_{ext}=0.2 T, under σ^+ [red (gray)] and σ^- (black) polarized excitation at 1.31 eV. The gray-shaded area represents the region of X^0 stability and the solid lines are a guide for the eye. (c) Overhauser shift versus circular polarization for X^- (+0.4 V) and X^+ (−0.2 V) measured for excitation polarizations varying from σ^+ to σ^-. Solid lines are theoretical fits according to Eq. (4) obtained with $(g_e/g_X)\delta_Z$=7 μeV, $\Delta^\star$=1.3(1.3) meV, τ_c=0.6(0.06) ns, and $\kappa^{-1/2}$ =1.8(3.8) μeV for X^+ (X^-), respectively.

man splitting of nuclei being much smaller), which thus produces a negative feedback on the nuclear polarization as we show in the following.

The Hamiltonian describing the hyperfine interaction of a single electron spin $\hat{S}^e=\frac{1}{2}\hat{\sigma}^e$ confined in a QD with N nuclear spins is given by[13,20]

$$\hat{H}_{hf} = \frac{\nu_0}{2}\sum_j A^j|\psi(\mathbf{r}_j)|^2\left(\hat{I}^j_z\hat{\sigma}^e_z + \frac{\hat{I}^j_+\hat{\sigma}^e_- + \hat{I}^j_-\hat{\sigma}^e_+}{2}\right) \tag{1}$$

where ν_0 is the two-atom unit cell volume, $\mathbf{r}_j$ is the position of the nucleus j with spin $\hat{I}^j$, A^j is the constant of hyperfine interaction with the electron, and $\psi(\mathbf{r})$ is the electron envelope function. The sum goes over the nuclei interacting significantly with the electron (i.e., essentially over the effective QD volume defined by $V=[\int|\psi(\mathbf{r})|^4 d\mathbf{r}]^{-1}=\nu_0 N/2$). This interaction has two important effects on the electron-nuclei spin system. (i) It acts as an effective magnetic field $\mathbf{B}_n \approx \Sigma_j A^j \mathbf{I}^j/(N g_e \mu_B)$ on the electron spin of g factor g_e. In the absence of nuclear polarization, this random nuclear field averages to zero but shows fluctuations proportional to $A/\sqrt{N}$ of the order of 30 mT.[2,3] In a classical description the electron spins precess around the total magnetic field $\mathbf{B}=\mathbf{B}_{ext.}+\mathbf{B}_n$, which determines the spin dynamics of a single electron[3] as well as of X^+.[2] (ii) Since this precession stops randomly within a correlation time τ_c (due to optical excitation and recombination or QD charging), the conservation of angular momentum leads to the transfer of spin polarization toward the nuclei. Quantum mechanically this flipflop mechanism corresponds to the second term of Eq. (1). The nuclear polarization can then accumulate in the QD giving rise to the OHS through the first term of Eq. (1) under the condition that the spin relaxation due to the dipole-dipole interaction between nuclei is quenched, which is in principle largely achieved for fields above 1 mT.[20]

To derive from Eq. (1) a convenient expression for the nuclear polarization dynamics we assume to first order a uniform electron wave function $\psi(\mathbf{r})=\sqrt{2/N\nu_0}$ over the involved nuclei. This also amounts to considering a uniform nuclear polarization $\rho_\alpha=\langle I^\alpha_z\rangle/I^\alpha$ for each isotopic species α of the QD. The theory of time-dependent perturbation up to the second order applied to the flipflop interaction characterized by a correlation time τ_c yields the following equation rate:[24]

$$\frac{d\langle I^\alpha_z\rangle}{dt} = -\frac{1}{T^\alpha_{1e}}(\langle I^\alpha_z\rangle - Q^\alpha\langle S^e_z\rangle) \tag{2}$$

where $Q^\alpha=I^\alpha(I^\alpha+1)/S(S+1)$ and T^α_{1e} is given by

$$\frac{1}{T^\alpha_{1e}} = \frac{2f_e\tau_c}{1+\left[\left(\frac{g_e}{g_X}\delta_Z+\delta_n\right)\tau_c/\hbar\right]^2}\left(\frac{A^\alpha}{N\hbar}\right)^2 \tag{3}$$

where f_e is the fraction of time that the QD contains an electron, which obviously depends on the excitation power. The OHS is then related to the average nuclear spin by $\delta_n=-2\Sigma_\alpha x_\alpha\langle I^\alpha_z\rangle A^\alpha$ where x_α is the fraction of species α. Note that Eq. (2) is valid only under the condition of weak nuclear polarization.[20,24] This is experimentally verified here with a maximum polarization $\rho_\alpha\approx 0.1$ deduced from the measured OHS ($\approx$25 μeV) divided by its maximum theoretical value ($\approx$250 μeV for a realistic InGaAs QD). The stationary solution of Eq. (2) driven by electron polarization $\langle S^e_z\rangle/S$ close to unity is therefore far from being reached, which means that nuclear depolarization must be taken into account. The physical origin of this mechanism likely relies on the dipolar (or quadrupolar) coupling between nuclei, which in spite of the screening by the applied magnetic field opens a way for nuclear spin relaxation due to the time-dependent hyperfine interaction with the electron spin.[13] However, since we could not investigate this effect further by varying the field, we simply describe it by adding to Eq. (2) the term $-\langle I^\alpha_z\rangle/T_d$ where T_d is a depolarization time constant independent of α. The OHS reached at equilibrium is then given by the implicit equation

$$\delta_n = \frac{-\Delta^*\langle S^e_z\rangle}{1+\kappa\left[(\hbar/\tau_c)^2+\left(\frac{g_e}{g_X}\delta_Z+\delta_n\right)^2\right]} \tag{4}$$

where $\Delta^*=2\tilde{A}\Sigma_\alpha x_\alpha Q^\alpha$, $\kappa=(N/\tilde{A})^2\tau_c/2f_eT_d$, and we have used $\tilde{A}$ ($\approx$50 μeV) instead of A^α, which indeed weakly depends on α. Here we treat Δ^*, τ_c, and κ as fitting parameters, while δ_n, δ_Z, and $\langle S^e_z\rangle$ can be determined from the experiments. Note yet that Δ^* amounts to $\approx$1.3 meV for a realistic In(Ga)As QD (with x_{In}=0.3, x_{Ga}=0.2, x_{As}=0.5).

Equation (4) clearly shows the negative feedback of the OHS on its equilibrium value through the electron spin splitting $[(g_e/g_X)\delta_Z+\delta_n]$. In particular it predicts the observed OHS asymmetry when changing the excitation polarization from σ^+ to σ^- since δ_n changes sign with respect to δ_Z. Note that the amplitude of this feedback depends directly on the finite nuclear spin depolarization time included in κ. Let us now examine in more detail the agreement of this model with our experimental results. For X^+, the correlation time τ_c of the hyperfine interaction is given by its lifetime ($\sim$1 ns if radiatively limited) and the average electron spin is determined from the PL circular polarization itself. The latter remains essentially constant around 70% (Fig. 3) whereas the OHS increases continuously with the electric field amplitude. This trend thus agrees well with Eq. (4) when assuming a reduction of trion lifetime due to the competition with the field-induced electron escape. For X^0, in addition to the anisotropic exchange term δ_1 which averages almost to zero the electron circular polarization $\langle S_z^e \rangle$ of bright states [see Fig. 2(c)], we should actually add to the spin splitting in Eq. (4) the direct exchange interaction ($\delta_0 \approx 0.5$ meV), since the hyperfine Hamiltonian only couples bright to dark excitons. We thus expect the nuclei polarization by X^0 to be much more difficult than for trions. For X^-, the nuclear polarization dynamics is now driven by the single electron left in the QD after optical recombination, whose lifetime determines τ_c. Above 0.35 V it is mainly limited by the capture time of a second electron tunneling from the n-GaAs reservoir. The dependence observed in Fig. 3(b) showing a maximum of OHS at 0.45 V bias agrees qualitatively well with a progressive reduction of τ_c leading first to the reduction of κ and then to the increase of $(\hbar/\tau_c)^2$ in Eq. (4). However, since τ_c, ρ_c, and likely T_d vary with the electric field it is not possible to fit the results of Fig. 3(b) without any additional assumptions. Therefore, to check the validity of this model we have varied at fixed bias the average spin $\langle S_z^e \rangle$ by rotating the quarter-wave plate which defines the excitation polarization. The measurements of ρ_c and δ_n are reported in Fig. 3(c) together with a theoretical fit. The good agreement, in particular regarding the asymmetrical and nonlinear dependence on ρ_c, gives strong support to our theoretical description. More intriguing, we could not observe the direct influence of the hyperfine field fluctuations on the electron spin relaxation[3] (i.e., on the circular polarization of X^+) for the reason that the circular polarization of X^+ was already quite strong in the 70–80 % range in zero magnetic field. Some finite nuclear polarization achieved in zero field as recently reported in Ref. 25 could explain this result.

In conclusion, we have shown that a significant nuclear polarization can be induced by optically polarized trions in a single InAs/GaAs quantum dot, with a very different signature according to the trion state X^+ or X^-. This demonstrates unambiguously that the nuclear polarization results from the hyperfine interaction with an unpaired electron within the quantum dot. Our results are discussed using a theoretical model which provides a fine understanding of the nuclear polarization mechanism and emphasizes the importance of electron spin splitting on its spin-flip rate.

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*Corresponding author. Email address: Olivier.Krebs@lpn.cnrs.fr

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