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

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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 ~0.2T. The hyperfine interaction between the optically oriented electron and nuclei 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 down 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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Spin dynamics of an electron confined in a self-assembled semiconductor quantum dot (QD) is currently the subject of an intense research. It indeed represents a promising direction for implementing quantum computation algorithms in solid state, because once the electron is confined in a quantum dot, its spin dynamics at low temperature is almost no longer subject 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. The subsisting sources of relaxation which have been identified in real QDs are (i) the exchange interaction with additional hole(s) or electron(s) and (ii) the hyperfine interaction with the QD nuclei spins. In order to address this issue with optical techniques, field-effect structures embedding charge-tunable QDs offer an amazing potential: the exchange-induced spin relaxation can be kept under control e.g. by extracting the hole from a photo-excited electron-hole pair (named further exciton) or by adding a charge preventing the exchange to operate, 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 Letter 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 down the electron and/or hole spins, whereas an external bias applied to the n-Schottky-type sample controls the electronic charge. The same single QD has been studied in three different regimes: when the electron spin $S^e$ interacting with the nuclear spins $I^e$ (i) forms with two photo-excited 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, we show that the small applied magnetic field leads to the optically-induced polarization of the QD nuclei, with a non-linear dependence on the average electron spin $\langle S^e_z \rangle$ deduced from the photoluminescence (PL) circular polarization. Remarkably, this shift changes sign with the 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. 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 200nm-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 1μm-diameter optical apertures to spatially select single QDs.

![Image](https://example.com/image.png)

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 intra-dot excitation at 1.31 eV.
Figure 1 shows the T=5K PL intensity contour plot 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 V and ~0.15 V we only observe the PL emission from the ground state exciton $X^0$ clearly identified by its fine structure [Fig. 2(b)] and by the biexciton (hardly perceptible in Fig. 1) appearing under stronger excitation at lower energy. (ii) Above 0.15 V the $X^-$ trion red-shifted by ~6 meV shows up indicating the charging of the QD with an electron $e$. Both lines ($X^0$ and $X^-$) still coexist because under non strictly-resonant excitation (here 1.31 eV) a single photo-hole can be 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 the occupation with 2 electrons. (iv) For negative bias a symmetrical charging effect occurs for holes. The neutral exciton line which disappears as a result of the electron tunnelling out of the QD, is replaced by a 3 meV blue-shifted line assigned to the trion $X^+_2$. Although the applied bias only controls the conduction band chemical potential and thus cannot itself generate the QC charging with holes, this effect is achieved under strong intra-dot excitation. It directly creates a hole within the QD, which does not escape as the electron thanks to its larger effective mass.

To study the influence of hyperfine interaction on spin dynamics, optical orientation experiments have been performed in presence of a small longitudinal magnetic field parallel to the QD growth axis $z$. We used a standard micro-PL setup based on a x50 microscope objective, a double spectrometer of 0.6m-focal length and a Nitrogen-cooled CCD array detector, providing a spectral resolution of 30 µeV and a precision on line position of about 1 µeV after de-convolution with a Lorentzian fit. The optical excitation and detection were both performed along the $z$ axis. Thus the degree of PL circular polarization defined by $\rho_c = (I_{g^+} - I_{g^-})/(I_{g^+} + I_{g^-})$, where $I_{g^+(-)}$ denotes the PL intensity measured in $\sigma^{(+)}$-(−) polarization, traces the average spin $\langle S^z \rangle = -\rho_c/2$ of the electron participating in the PL signal. This results from the usually accepted assumption of pure heavy-hole ground state with angular momentum projection $m_z = \pm \frac{1}{2}$ in InAs QDs leading to optically active electron-hole pairs $|\pm\rangle = |\pm\frac{1}{2}, \pm\frac{1}{2}\rangle$. 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 in-plane anisotropy of real QDs the latter is further split by an energy $\delta_1 \approx 50$ meV into $|X\rangle$ and $|Y\rangle$ eigenstates linearly polarized along the crystallographic axes $(110)$ and $(110)$. 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. 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 $X^2$. In magnetic field, the Zeeman interaction separates the $\sigma^\pm$-polarized components of the trion lines by $\delta_Z = g_X |\mu_B B^\text{ext}|$ where $g_X$ is the exciton g-factor (supposed to be constant for the three lines considered here) and $\mu_B = 58\mu$eV/T is the Bohr magneton. It also increases the bright $X^0$ splitting to $\delta_0 = \sqrt{\delta_1^2 + \delta_Z^2}$. Figure 2(c) shows the PL spectra resolved in circular polarization. Under linearly polarized excitation we find for the three lines a Zeeman splitting $\delta_2 \approx 28$ µeV in agreement with an exciton g-factor of $\sim 3$. Under circularly polarized excitation a significant deviation from the sole Zeeman interaction is now observed: the $X^+$ splitting gets larger in $\sigma^+$ excitation by +10 µeV and smaller in $\sigma^-$ by -15 µeV. This so-called Overhauser shift (OHS) denoted $\delta_o$ 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 $\delta_n=+15$ $\mu$eV in $\sigma^+$ and $\delta_n=-25$ $\mu$eV in $\sigma^+$, 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 doesn’t operate during $X^-$ 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 $X^+$ (situation of Fig. 2(c) because of excitation at higher energy), in which case it only acts as a probe of OHS produced by $X^+$. An other feature of these results is the pronounced OHS asymmetry when changing the excitation from $\sigma^+$ to $\sigma^-$. This clearly appears in Fig. 3 which reports the bias dependence of circular polarization at higher energy), in which case it only acts as a probe of OHS produced by $X^+$. 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 $X^+$ (situation of Fig. 2(c) because of excitation at higher energy), in which case it only acts as a probe of OHS produced by $X^+$. An other feature of these results is the pronounced OHS asymmetry when changing the excitation from $\sigma^+$ to $\sigma^-$. This clearly appears in Fig. 3 which reports the bias dependence of circular polarization and of the trion spin splitting. This asymmetry means that polarizing the nuclear spins in the direction which 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-nuclei flip-flop process (the Zeeman 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 [12 20]:

$$\hat{H}_{hf} = \frac{\nu_0}{2} \sum_j A_j |\psi(r_j)|^2 \left( \hat{I}_j \hat{\sigma}_z + \frac{\hat{I}_j \hat{\sigma}_- + \hat{I}_j \hat{\sigma}_+}{2} \right) \tag{1}$$

where $\nu_0$ is the two-atom unit cell volume, $r_j$ is the position of the nuclei $j$ with spin $\hat{I}_j$, $A_j$ is the constant of hyperfine interaction with the electron and $\psi(r)$ is the electron envelope function. The sum goes over the nuclei interacting significantly with the electron (i.e. essentially in the effective QD volume defined by $V = (\int |\psi(r)|^4 \, dr)^{-1} = \nu_0 N/2$). This interaction has two important effects on the electron-nuclei spin system. (i) It acts as an effective magnetic field $B_n \approx \sum_j A_j \mathbf{r}_j/(N g_e \mu_B)$ on the electron spin of g-factor $g_e$. In absence of nuclear polarization, this random nuclear field averages to zero but shows fluctuations $\propto A/\sqrt{N}$ of the order of 30 mT [2, 3]. In a classical description the electron spin precess around the total magnetic field $\mathbf{B} = B_{ext} + B_n$, which determines the spin dynamics of a single electron [2] as well as of $X^+$ [2]. (ii) Since this precession stops randomly within a correlation time $\tau_c$ (due to optical excitation/recombination or QD charging), the conservation of angular momentum leads to the transfer of spin polarization towards the nuclei. Quantum mechanically this flip-flop 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 diffusion driven by 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 as a first order a uniform electron wavefunction $\psi(r) = \sqrt{2/N} \nu_0$ over the involved nuclei. This also amounts to consider a uniform nuclear polarization $\rho_n = \langle I^n_z \rangle / I^n$ for each isotopic specie $\alpha$ of the QD. The theory of time-dependent perturbation up to the second order applied to the flip-flop interaction characterized by a correlation time $\tau_c$ yields the following equation rate [24]:

$$\frac{d\langle I^n_z \rangle}{dt} = -\frac{1}{T^n} \langle (I^n_z) - Q^n (S^n_z) \rangle \tag{2}$$

where $Q^n = I^n(I^n+1)/\overline{S^+S^-}$ and $T^n$ is given by:

$$\frac{1}{T^n} = \frac{2f_e \tau_c}{1 + \left[ (\frac{g_s}{g} \delta_Z + \delta_n) T_e / \hbar \right]^2} \left( \frac{A^n}{N \hbar} \right)^2 \tag{3}$$

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
spins by \( \delta_n = -2 \sum \alpha x_\alpha \langle I_\alpha^z \rangle A^\alpha \) where \( x_\alpha \) is the fraction of specie \( \alpha \). Note Eq. (2) is only valid under the condition of weak nuclear polarization \([21, 24]\). This is experimentally verified here with a maximum polarization \( \rho_e \approx 0.1 \) deduced from the measured OHS (\( \approx 25 \) \( \mu \)eV) divided by its maximum theoretical value (\( \approx 250 \)\( \mu \)eV for a realistic InGaAs QD). The stationary solution of Eq. (2) driven by the electron polarization \( \langle S^z_e \rangle / S \) 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, that in spite of the screening by the applied magnetic field opens a way for nuclear spin diffusion due to the time-dependent hyperfine interaction with electron spin \([18]\). However, since we could not investigate further this effect by varying the field, we simply describe it by adding to Eq. (2) the term \(- f_e \langle I_\alpha^z \rangle / T_d \) where \( T_d \) is a depolarization time constant independent on \( \alpha \). The OHS reached at equilibrium is then given by the implicit equation:

\[
\delta_n = \frac{-\Delta^* \langle S^z_e \rangle}{1 + \kappa \left( \frac{\hbar}{\tau_e} \right)^2 + \left( \frac{g_e}{g_x} \delta_Z + \delta_n \right)^2} \tag{4}
\]

where \( \Delta^* = 2A \sum \alpha x_\alpha Q^\alpha \), \( \kappa = \tau_e / T_d (N / \bar{A})^2 \) and we have used \( A \) (\( \approx 50 \) \( \mu \)eV) instead of \( A^\alpha \) which indeed weakly depends on \( \alpha \). Here we treat \( \Delta^*, \tau_e \) and \( \kappa \) as fitting parameters, while \( \delta_n, \delta_Z, \langle S^z_e \rangle \) can be determined from the experiments. Note yet that \( \Delta^* \) 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 \( \left( \frac{g_e}{g_x} \delta_Z + \delta_n \right) \). In particular it predicts the observed OHS asymmetry when changing the excitation polarization from \( \sigma^+ \) to \( \sigma^- \) since \( \delta_n \) changes sign with respect to \( \delta_Z \). Note the amplitude of this feedback depends directly on the finite nuclear spin depolarization time included in \( \kappa \). Let us now examine in more details the agreement of this model with our experimental results. For \( X^+ \), the correlation time \( \tau_e \) of the hyperfine interaction is given by its lifetime (\( \approx 1 \) ns if radiatively limited) and the average electron spin is determined from the PL circular polarization itself. The latter remains essentially constant around 70% \([\text{Fig. 3}] \) whereas the OHS increases continuously with the electric field amplitude. This trend agrees thus 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 \( \delta_0 \) which averages to almost 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. (2) 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 \( \tau_e \). 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 \( \tau_e \) leading first to the reduction of \( \kappa \) and then to the increase of \( (\hbar / \tau_e)^2 \) in Eq. (4). However, since \( \tau_e, \rho_e \) 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 \( \rho_e \) and \( \delta_n \) are reported in Fig. 3(c) together with a theoretical fit. The good agreement, in particular regarding the asymmetrical and non-linear dependence on \( \rho_e \) gives a strong support to our theoretical description. More intriguing, we couldn’t observe the direct influence of the hyperfine field fluctuations on the electron spin relaxation (i.e. on the circular polarization of \( X^+ \)) \([3]\) for the mere reason that the circular polarization of \( X^+ \) was already quite strong in the 70–80% range in zero magnetic field. This contrasts to Ref. \([2]\) where an ensemble of p-doped QDs was studied. Further investigations are thus required to elucidate this zero field result.

In conclusion, we have shown that a significative 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 \( X^- \). It demonstrates unambiguously that the nuclear polarization results from the hyperfine interaction with an unpaired electron within the quantum dot. Our results are discussed with 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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[1] M. Krounva et al., Nature 432, 81 (2004).
[2] P.-F. Braun et al., Phys. Rev. Lett. 94, 116601 (2005).
[3] I. Merkulov et al., Phys. Rev. B 65, 205309 (2002).
[4] A. V. Khaetskii et al., Phys. Rev. Lett. 88, 186802 (2002).
[5] J. M. Smith et al., Phys. Rev. Lett. 94, 197402 (2005).
[6] S. Laurent et al., Phys. Rev. Lett. 94, 147401 (2005).
[7] M. V. G. Dutt et al., Phys. Rev. Lett. 94, 227403 (2005).
[8] A. V. Khaetskii and Y. V. Nazarov, Phys. Rev. B 61, 12639 (2000).
[9] R. J. Warburton et al., Phys. Rev. Lett. 79, 5282 (1997).
[10] T. Flissikowski et al., Phys. Rev. B 68, 161309(R) (2003).
The weaker line denoted $X^+\star$ is assigned to a hot trion because of its broad Lorentzian lineshape and its 5 meV redshift resulting of triplet configuration.