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Controlling the polarization eigenstate of a quantum dot exciton with light

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We demonstrate optical control of the polarization eigenstates of a neutral quantum dot exciton without any external fields. By varying the excitation power of a circularly polarized laser in micro-photoluminescence experiments on individual InGaAs quantum dots we control the magnitude and direction of an effective internal magnetic field created via optical pumping of nuclear spins. The adjustable nuclear magnetic field allows us to tune the linear and circular polarization degree of the neutral exciton emission. The quantum dot can thus act as a tunable light polarization converter.

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Semiconductor quantum dots (QDs) are nanometer sized objects that contain typically several thousand atoms resulting in a confinement of electrons in all three spatial directions. The absence of translational motion prolongs the carrier spin lifetimes as compared to bulk (3D) and quantum well (2D) structures [1, 2, 3, 4, 5]. As a result a large number of schemes for QD spin based q-bit manipulations have been proposed [6]. After optical excitation, a conduction electron and a valence hole form a neutral exciton X^0 in the dot. For the model system of self assembled InGaAs QDs in GaAs, the anisotropic electron hole Coulomb exchange interaction for the QD symmetry C_{2v} gives rise to a bright X^0 doublet of eigenstates $|X\rangle$ and $|Y\rangle$ polarized along the $[1\bar{1}0]$ and $[110]$ crystallographic directions, respectively [7, 8]. To characterize the strength of the anisotropic Coulomb exchange an effective magnetic field B_{AEI} in the QD plane acting on the exciton pseudo spin can be introduced [9, 10, 11].

In analogy to the electron and the proton in a hydrogen atom, the electron in a QD is also interacting with the *magnetic* moment of the nuclear spins of the atoms that form the dot [12]. The electron-hole Coulomb exchange interaction cancels out in the ground state of a singly charged exciton as for example the X^+ (2 holes + 1 electron) as the holes form a spin singlet [13]. Under suitable excitation conditions, the electron polarization created through optical pumping of the X^+ exciton can be transferred to the nuclear spins in the dot via the hyperfine interaction even at zero applied magnetic field, giving rise to an effective magnetic field B_N that can in turn stabilize the electron spin [1, 14, 15, 16].

In this Letter we demonstrate optical control of the polarization eigenstate of a neutral quantum dot exciton X^0 in the absence of any external magnetic or electric field. We show novel effects resulting from the combined effect of the effective nuclear magnetic field B_N and the

Coulomb exchange interaction (i.e. B_{AEI}) on the electron spin in an InGaAs QD: the control over B_N via non-resonant optical pumping allows us to orientate the pseudo spin of a neutral exciton and therefore achieve substantial optical orientation, previously only reported for charged excitons. As compared to charged excitons, we show that the robust electron spin injection for X^0 has the advantage that in the presence of B_{AEI} the quantum dot can act as a *tunable* light polarization converter. The degree of circular to linear polarization conversion can be adjusted through a slight variation in excitation laser power, which could provide a new approach to switching the polarization of QD based single photon emitters [17]. We show that the build-up of B_N is possible due to the presence of charged excitons X^+ appearing under non-resonant pumping conditions.

The sample consists of: GaAs substrate, 20 nm of GaAlAs, 98 nm GaAs, delta doping Si 10^9 cm⁻², 2 nm GaAs, InGaAs wetting layer (WL) and QDs, 100 nm GaAs, 20 nm of GaAlAs, 5nm GaAs. Although the samples are intentionally n-doped, detailed spectroscopic analysis shows that (residual) p-type doping prevails, leading to the observation of neutral and singly positively charged excitons. The photoluminescence (PL) and PL excitation (PLE) measurements at 4K on individual QDs were carried out with a confocal microscope build around attocube nano-positioners connected to a spectrometer and a charge coupled device (CCD) camera. The signal to noise ratio of 10^4 obtained by placing a solid immersion lens on the sample allows to obtain a spectral precision of ± 1 μ eV for the transition energy by fitting the spectra with Lorentzian lineshapes. The excitation energy E_{laser} of a continuous wave Ti-Sapphire laser is varied between 1.38 and 1.48 eV, covering the heavy hole and light hole to electron transitions in the WL [18, 19]. The circular polarization degree of the QD PL is defined as $P_c = (I^+ - I^-)/(I^+ + I^-)$, where $I^{+(-)}$ is the $\sigma^{+(-)}$ polarized PL intensity integrated over the spectral domain covering the X^0 doublet (X^+ singlet) emission. The linear polarization degree is defined as

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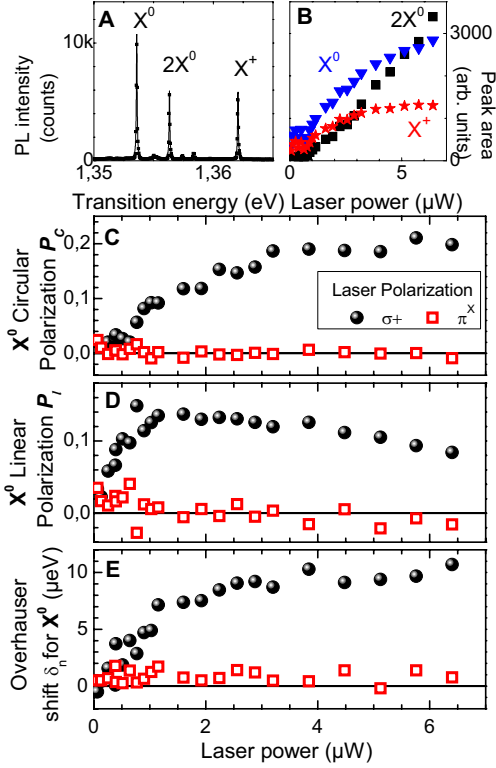


FIG. 1: (Color online) QD I. $E_{\text{laser}} = 1.44$ eV. (A) PL spectrum at 4K for $P=2.5\mu\text{W}$ and linear π^X excitation and detection. (B) Integrated PL intensity as a function of laser power for the X^0 (triangles), X^+ (stars) and $2X^0$ (squares). The circular polarization degree P_c (C), the linear polarization degree P_l (D) and the Overhauser shift δ_n (E) are shown as a function of laser power for X^0 . Solid circles (hollow squares) represent σ^+ (π^X) laser polarization.

$$P_l = (I^X - I^Y)/(I^X + I^Y).$$

Figure 1.A shows a typical PL spectrum for $E_{\text{laser}}=1.44$ eV exciting the heavy hole to electron transition in the WL about 90meV above the QD neutral exciton X^0 emission. Three transitions are observed: X^0 , the neutral biexciton $2X^0$ (2 electrons + 2 holes) which is blue shifted [18], and the positively charged exciton X^+ . The X^+ shows as anticipated no fine structure and a P_c in the order of 50% under strong pumping with circularly polarized light (not shown) [20]. The attribution of the three transitions is in agreement with the power dependence of the PL intensity shown in figure 1B.

Under linearly polarized excitation, the two bright X^0 states $|X\rangle = \frac{|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle}{\sqrt{2}}$ and $|Y\rangle = \frac{|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle}{i\sqrt{2}}$ are separated in energy by $\delta_1 \equiv E_X - E_Y \simeq -9\mu\text{eV}$ for dot I due to B_{AEI} [7]. Here \uparrow (\downarrow) stands for the heavy hole pseudo spin up (down) and \uparrow (\downarrow) for the electron spin up (down) projections onto the z -axis, which is also the light propagation axis and the sample growth axis [21]. The same splitting $|\delta_1|$ is found for the $2X^0$, but as expected with the order of the peaks reversed. It is important to note

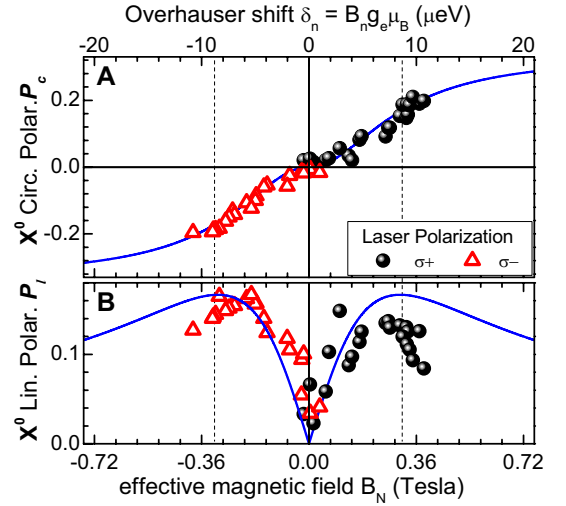


FIG. 2: (Color online) QD I. $E_{\text{laser}} = 1.44$ eV. $\delta_1 = -9\mu\text{eV}$. Circular polarization degree P_c (B) and Linear polarization degree P_l (C) for the X^0 states as a function of the effective nuclear field B_N for σ^+ (σ^-) laser polarization shown as solid circles (hollow triangles) assuming $|g_e| = 0.48$. Solid lines are calculated with equations 2 and 3. Dashed vertical line: measured value of $|\delta_1|$.

that equal intensities $I^X = I^Y$ of the linearly polarized transitions $|X\rangle$ and $|Y\rangle$ result in a net PL polarization $P_l = 0$ when integrating over both transitions.

A surprising power dependence of P_c and P_l for X^0 following circularly polarized excitation in the WL is shown in figure 1C and D. We observe an increase from $P_c \simeq 0$ up to 22% when exciting with P_{exc} above $4\mu\text{W}$ (figure 1C), so substantial optical orientation has been achieved. Even more intriguing, we observe under *circularly* polarized excitation that the *linear* polarization increases abruptly with Laser power from $P_l \simeq 0$ to 17% before gradually decreasing for $P_{\text{exc}} > 1\mu\text{W}$, (figure 1D). These effects do not purely depend on laser power but also polarization, as can be seen in figures 1C and D: exciting with linearly polarized light results in $P_c \simeq P_l \simeq 0$ for X^0 with no dependence on P_{exc} [22].

With only the exchange interaction B_{AEI} present the X^0 eigenstates $|X\rangle$ and $|Y\rangle$ are linearly polarized. Excitation with circularly polarized light should result in beats in the time domain between $|\uparrow, \downarrow\rangle$ and $|\downarrow, \uparrow\rangle$ as those are not the X^0 polarization eigenstates [23, 24]. So assuming (i) an exponential radiative decay for the X^0 with a characteristic time $\tau_r = 700\text{ps}$ [3] and (ii) an exciton spin lifetime $\tau_s \gg \tau_r$, the measured P_c in cw PL would be $P_c = P_c^0(1 + \omega^2\tau_r^2)^{-1}$ with $\hbar\omega = \delta_1$ and P_c^0 is the P_c created in the dot at time $t=0$ for X^0 [10]. For the measured $\delta_1 \simeq -9\mu\text{eV}$ one would only expect $P_c^{\text{MAX}} \simeq 1\%$, and not 22% as found in the experiment. Concerning P_l , circularly polarized excitation should result in PL with $I^X = I^Y$ and hence $P_l = 0$ which is in contradiction to the 17% measured.

The observed optical orientation and polarization con-

version can not be explained without invoking new X^0 eigenstates. We will argue in the following that non-resonant optical pumping has created a dynamic nuclear spin polarization (DNP) that acts on the electron spin like an effective internal magnetic field of several hundred mT along the z -axis (see figure 2A and B). The coupling of the nuclear spins to the electron spin via the Fermi-contact interaction (neglected here for hole spins [12]) can be expressed as

$$H_{HF} = \sum_k^N A_k \left(I_z^k S_z + \frac{I_-^k S_+ + I_+^k S_-}{2} \right) \quad (1)$$

and $\langle H_{HF} \rangle = A \langle \vec{I} \rangle \vec{S} \equiv g_e \mu_B \vec{B}_N \vec{S}$, where \vec{I}^k and \vec{S} are the spin operator for nucleus k (out of $N \simeq 10^4 - 10^5$) and for the electron spin, respectively. g_e is the longitudinal electron g factor and I_z^{MAX} for In, Ga and As is $9/2$, $3/2$ and $3/2$, respectively. The combined effect of an *external* longitudinal magnetic field and B_{AEI} on the bright exciton doublet are detailed in [3, 7, 9, 11, 25]. Here we simply replace the Zeeman Hamiltonian by $\langle H_{HF} \rangle$ resulting in a Zeeman splitting (called Overhauser shift) purely due to $\vec{B}_N = (0, 0, B_N)$ of $\delta_n = g_e \mu_B B_N$.

The presence of a magnetic field component along the z -axis will result in

(i) a splitting $\sqrt{\delta_1^2 + \delta_n^2}$ of the bright X^0 doublet and
(ii) new eigenstates $|+\rangle = \alpha|X\rangle + i\beta|Y\rangle$ and $|-\rangle = \beta|X\rangle - i\alpha|Y\rangle$ where $\alpha^2 = 1$ ($\alpha^2 \rightarrow 1/2$) for $B_N = 0$ ($B_N \rightarrow \infty$) and $\alpha = \alpha(\delta_1, \delta_n)$ and $\beta = \beta(\delta_1, \delta_n)$. Assuming that $\tau_r \gg \Omega^{-1}$ where $\hbar\Omega = \sqrt{\delta_1^2 + \delta_n^2}$ we find:

$$P_c(\delta_n) = 4\alpha^2 \beta^2 P_c^0 = \delta_n^2 P_c^0 / (\delta_n^2 + \delta_1^2) \quad (2)$$

$$P_l(\delta_n) = 2\alpha\beta(\alpha^2 - \beta^2) P_c^0 = -\delta_n \delta_1 P_c^0 / (\delta_n^2 + \delta_1^2) \quad (3)$$

With our fitting procedure we can extract δ_n for X^0 as a function of Laser power for σ^+ excitation (see figure 1E) [26]. An identical splitting with opposite sign is found for σ^- excitation (not shown) and for linearly polarized excitation we measure $\delta_n \simeq 0$, as $B_N = 0$. As the dependence of δ_n on P_{exc} is non-linear, it is more instructive to plot P_c (figure 2.A) and P_l (figure 2.B) achieved for X^0 as a function of the created field $B_N \propto \delta_n$ (assuming an electron g -factor of $|g_e| = 0.48$ [13]). The experimental curves in figure 2 are very well reproduced using $|P_c^0| = 33\%$ as the only fitting parameter in equations 2 and 3. The measured P_c of $\pm 22\%$ for $\sigma\pm$ excitation represents 65% of the maximum achievable $P_c^0 = \pm 33\%$ for $\delta_n \rightarrow \infty$, although only a low nuclear polarization of roughly 5% is achieved as $\delta_n \simeq 10 \mu\text{eV}$. We demonstrate a wide range of tunability for the circular to linear conversion as we go from $P_l \simeq 0$ to the theoretical limit of maximum conversion $P_l = P_c^0/2$ for $|\delta_n| = |\delta_1|$. For $|\delta_n| > |\delta_1|$ P_l decreases in both theory and experiment. For P_l not all experimental points are on the theoretical curve and we notice a slight asymmetry between σ^+ and σ^- excitation. Our simple model does not take into account strain induced heavy hole - light hole coupling which results in X^0 eigenstates which are already at $B_N = 0$ different

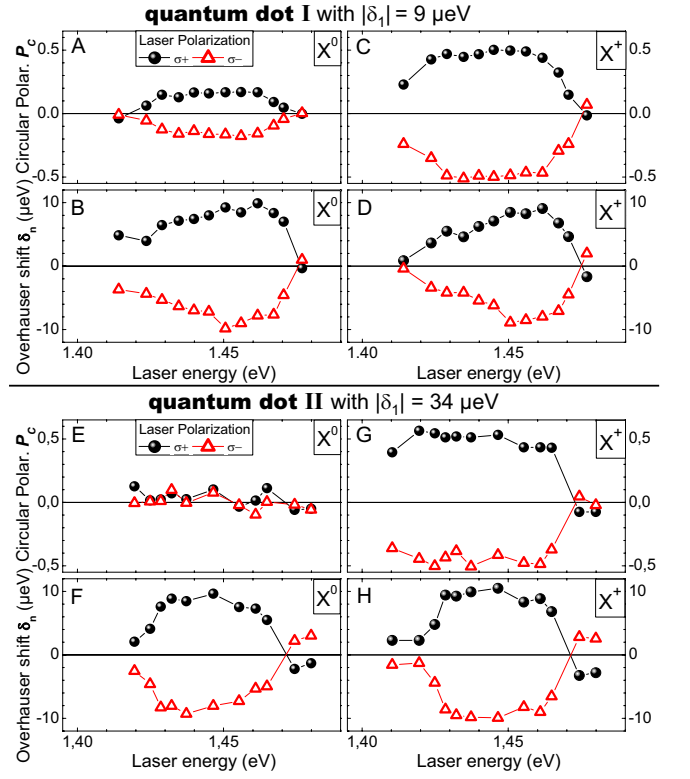


FIG. 3: (Color online) PL detection energy dot I (II) $\simeq 1.358$ eV ($\simeq 1.342$ eV). Circular polarization degree P_c and Overhauser shift δ_n as a function of laser energy for σ^+ (σ^-) laser polarization shown as solid circles (hollow triangles) for (A,B) X^0 of dot I, (C,D) X^+ of dot I, (E,F) X^0 of dot II and (G,H) X^+ of dot II.

from $|X\rangle$ and $|Y\rangle$ [11, 27]. This could be at the origin of the observed discrepancy.

In the following we discuss the origin of the DNP that builds up through a simultaneous spin flip of an electron spin with a nuclear spin through the fluctuating term $(I_-^k S_+ + I_+^k S_-)$ in equation 1 [1, 13]. This flip-flop process is repeated in time and a steady state nuclear polarization is reached. This process is very costly in energy for an electron in a neutral exciton X^0 [5], as the bright and dark states (for example $|\uparrow, \downarrow\rangle$ and $|\uparrow, \uparrow\rangle$) are separated in energy by the isotropic exchange energy of up to $\delta_0 \simeq 500 \mu\text{eV}$ at zero magnetic field in InGaAs dots [28]. As a result the probability for electron-nuclear spin flip-flops is very low, so we argue that (i) the electron of the X^0 does not transfer its spin to the nuclei (see below) (ii) X^0 is robust against decoherence by fluctuating nuclear spins which acts through the same flip-flop term. In the literature nuclear spin effects on carriers forming the X^0 have been studied in applied magnetic fields much larger than B_{AEI} where the Zeeman effect dominates [5, 25]. As shown below, the X^0 is not at the origin of the DNP, but merely experiences the existing field B_N in the dot.

The PL in figure 1A shows that the dot is occupied by X^0 some of the time, by X^+ for the rest of the time. The

different origins of the charged excitons are discussed in the literature [29]. In a simple picture, assuming that the dot contains a doping hole, the capture process for electrons (which are less likely to be trapped by potential fluctuations of the WL) could be faster than for holes and an X_0 is formed. If no hole arrives within $t \leq \tau_r$, the X^0 will recombine, if a hole is trapped for $t \leq \tau_r$, the X^+ exciton is formed. Alternatively, a hole could tunnel into or out of the dot during τ_r to a nearby acceptor. During the radiative lifetime of the X^+ electron-nuclear spin flip-flop processes are far more likely as compared to the X^0 case, because in the absence of Coulomb exchange the energy difference between the X^+ states $|\uparrow\downarrow, \downarrow\rangle$ and $|\uparrow\downarrow, \uparrow\rangle$ is only $\simeq \delta_n$. As B_N is essentially constant over at least ms [30], the electron spin of the X^0 experiences the same B_N as the electron in the X^+ .

To demonstrate this point, we compare PLE measurements on the dot investigated already in figures 1 and 2 (dot I, see figure 3 upper part) with an additional dot II with a considerably larger splitting $\delta_1 = 34\mu\text{eV}$ (see figure 3 lower part). High values of P_c in the order of 50% are created for the X^+ in both dots when exciting with $E_{\text{laser}}=1.425$ to 1.465 eV. When approaching the low energy tail of the density of states of the WL at $E_{\text{laser}} \simeq 1.41$ eV, the carrier absorption rate is too low to create nuclear polarization [13]. At $E_{\text{laser}} \geq 1.48$ eV the P_c drops in absolute value and even changes sign as the light hole transitions in the WL are excited [19, 31]. δ_n changes sign accordingly, which demonstrates that nuclear spin orientation in a QD can also be controlled via the optical selection rules in the WL. Comparing figures

3G and 3H for dot II shows clearly that the P_c created for the X^+ is transferred to the nuclear spins. In stark contrast, the P_c for the X^0 is on average zero in figure 3E. The neutral exciton X^0 in both dots is subject to a nuclear field of several hundred mT (figure 3B and 3F), created by the charged exciton state X^+ , but for dot II $B_{AEI} \gg B_N$, so the projection of the total effective magnetic field onto the z -axis is too small to induce optical orientation. Although the values of δ_1 differ by a factor of 3.5 due to different QD size, shape, composition and strain, the values of δ_0 are less sensitive to the exact dot symmetry as they originate mainly from the short range Coulomb exchange [7, 32]. Similar values of $\delta_0 \gg \delta_1, \delta_n$ of typically a few hundred μeV can be assumed for both dots I and II. This means that the P_c shown in figure 3A for dot I is due to the B_N present in the dot, and not vice versa [33].

In summary, optical orientation of neutral excitons X^0 in single QDs in the absence of any applied fields is achieved as an effective nuclear magnetic field B_N is constructed through non-resonant optical pumping. Varying B_N in the presence of a constant B_{AEI} due to Coulomb exchange allows efficient and tunable conversion of circularly to linearly polarized light mediated by a single QD. Considering the slow evolution of B_N [30] and the robustness of the electron spin during energy relaxation, our all optical approach could evolve in future experiments to orientate both the nuclear and the electron spins electrically in QD based Spin- Light Emitting Diodes [34, 35, 36].

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