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 microphotoluminescence 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 [1–5]. As a result a large number of schemes for QD spin based qbit 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 (CE) 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]. The splitting in energy between $|X\rangle$ and $|Y\rangle$ can be expressed in terms of an effective magnetic field B_{AEI} in the QD plane acting on the exciton pseudo spin [8,9]. The magnitude of B_{AEI} characterizes the strength of the anisotropic CE.

The electron in a QD is also interacting with the nuclear spins of the atoms that form the dot [1,10]. The electron-hole CE interaction cancels out ($B_{AEI} = 0$) in the ground state of a singly charged exciton X^+ (2 holes + 1 electron) [11]. 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 [12]. The electron spin present during the radiative lifetime of the X^+ interacts strongly with the nuclear spins, the hole spin left behind after recombination interacts only weakly [1]. The created dynamic nuclear polarization (DNP) can be expressed as an effective magnetic field B_N that can in turn stabilize the electron spin [1,12–14].

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. This has to the best of our knowledge never been reported before in InAs dots or any other material system. We show novel effects resulting from the combined effect of the effective nuclear magnetic field B_N and the CE interaction (i.e., B_{AEI}) on the electron spin in an InGaAs QD: the control of the nuclear field B_N via nonresonant 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 [1,12–14]. As compared to charged excitons, we show that the robust electron spin injection for X^0 has the advantage that in the presence of an effective magnetic field B_{AEI} perpendicular to the light propagation axis, the QD can act as a *tunable* light polarization converter. Light polarization conversion was previously reported in variable, external magnetic fields [9]. Zero field optical polarization conversion reported in [8] in CdSe/ZnSe dot ensembles reached a fixed, not tunable maximum of 3%. In our novel scheme 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 [15]. We show that the buildup of B_N is possible due to the presence of charged excitons X^+ appearing under nonresonant 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, 5 nm GaAs. The samples are intentionally n doped, but spectroscopy shows that (residual) p-type doping prevails, leading to the observation of X^0 and X^+. The photoluminescence (PL) and PL excitation (PLE) measurements at 4 K on individual QDs were carried out with a confocal microscope build around attocube nanopositioners connected to a spectrometer and a charge coupled device (CCD) camera. With a Fabry-Perot interferometer in front of the spectrometer, we obtain QD PL spectra with a FWHM of 15 μeV. The transition energy is determined through a Lorentzian fit with an error of ± 0.2 μeV. For low signal levels the number of counts on the CCD is increased by a factor of 50 when removing the interferometer. The signal to noise ratio of 10^4 obtained by placing a solid immersion lens on the sample allows us to obtain a precision of ± 1 μeV for the transition energy by fitting the spectra with Lorentzian lineshapes (FWHM 80 μeV). 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 $P_l = (I^X - I^Y)/(I^X + I^Y)$.

Typical PL emission for $E_{\text{laser}} = 1.44$ eV of a continuous wave Ti-Sapphire laser exciting the heavy hole to electron transition in the WL about 90 meV above the QD X^0 emission [16]shows three transitions : X^0 [Fig. 1(a)], the neutral biexciton $2X^0$ (2 electrons + 2 holes) which is blue shifted (not shown), and the positively charged exciton X^+. The X^+ shows as anticipated no fine structure when the laser is linearly polarized [Fig. 1(b) lower panel] and a circular polarization P_c in the order of 50% under strong pumping with circularly polarized light [Fig. 1(b) upper panel] [17].

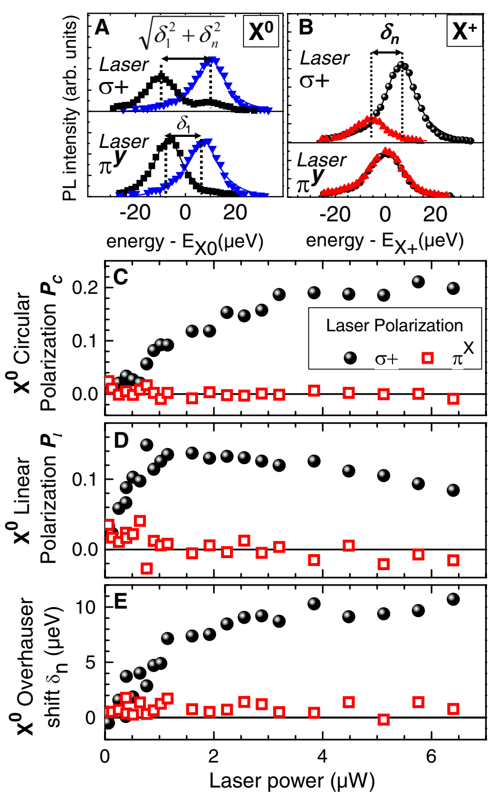

FIG. 1 (color online). QD III. $E_{\text{laser}} = 1.44$ eV. (a) X^0 PL with $E_{X0} = 1.340$ eV recorded with interferometer for $\pi^Y(\sigma^+)$ laser polarization in lower (upper) panel. $\pi^Y(\pi^X)$ detection shown as solid squares (triangles). (b) X^+ PL with $E_{X+} = 1.346$ eV recorded with interferometer for $\pi^Y(\sigma^+)$ laser polarization in lower (upper) panel. $\sigma^+(\sigma^-)$ detection shown as solid circles (triangles). QD I: The circular polarization P_c (c), the linear polarization P_l (d). Overhauser shift δ_n (e) shown as a function of laser power for X^0. Solid circles (hollow squares) represent σ^+ (π^X) laser polarization.

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 $\delta_1 \equiv E_X - E_Y$ 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 [18]. A typical high resolution spectrum is shown for dot III [Fig. 1(a), lower panel] where $\delta_1 = 13.6$ μeV. Note that equal intensities $I^X = I^Y$ of linearly polarized transitions result in a net PL polarization $P_l = 0$ when integrating over both transitions.

As an example for the intriguing characteristics of dots with smalls values of δ_1 we show below a detailed investigation of dot I with $\delta_1 = -9$ μeV. A surprising power dependence of P_c and P_l for X^0 following circularly polarized excitation in the WL is shown in Figs. 1(c) and 1(d). With laser power P_c increases from $\simeq 0$ up to 22% [Fig. 1(c)], so substantial optical orientation has been achieved without any applied magnetic field. 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 $P_{\text{exc}} > 1$ μW, [Fig. 1(d)]. These effects do not purely depend on laser power but also polarization [Figs. 1(c) and 1(d)]: exciting with linearly polarized light results in $P_c \simeq P_l \simeq 0$ for X^0 with no dependence on P_{exc} [19] [compare also lower and upper panel of Fig. 1(a)].

With only the CE interaction present the X^0 eigenstates $|X\rangle$ and $|Y\rangle$ are linearly polarized. Circularly polarized excitation 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 [20]. So assuming (i) an exponential radiative decay for the X^0 with a characteristic time $\tau_r = 700$ ps and (ii) an exciton spin lifetime $\tau_s \gg \tau_r$ (as confirmed in [3]) 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 [8]. For the $\delta_1 \simeq -9$ μeV one would only expect $P_c^{\text{MAX}} \simeq 1\%$, and not the 22% measured. 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 data cannot be explained without invoking new X^0 eigenstates. We argue that nonresonant optical pumping has created DNP that acts on the electron spin like an effective internal magnetic field of several hundred mT along the z axis [see Figs. 2(a) and 2(b)]. The coupling of the nuclear spins to the electron spin via the Fermi contact interaction can be expressed as

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

and $\langle H_{HF}\rangle = (\sum_k^N A_k\langle\vec{I}^k\rangle)\vec{S} \equiv g_e\mu_B\vec{B}_N\vec{S}$, 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 $I_z^{\rm MAX}$ 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 [3,4,7,9]. 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 $|-\rangle = \beta|X\rangle - i\alpha|Y\rangle$, as detailed in [21]. Both (i) and (ii) are clearly visible for dot III in Fig. 1(a). Assuming that $\tau_r \gg \Omega^{-1}$ where $\hbar\Omega = \sqrt{\delta_1^2 + \delta_n^2}$ we find:

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

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

To apply the above formulas to experiments in external magnetic fields, as in [9], the exciton Zeeman splitting has to be added to δ_n. We can extract δ_n for X^0 as a function of laser power for σ^+ excitation [see Fig. 1(e)] [21]. When the signal level is too low for using the Fabry-Perot interferometer, a double Lorentzian for the transitions $|+\rangle$ and $|-\rangle$ is detected, for which the energy splitting $\hbar\Omega = E_+ - E_-$ is much smaller than the detected linewidth. This double peak can be approximated by a single Lorentzian centered at $\frac{E_+ - E_-}{2}$. As shown in [21] changes in the oscillator strength of $|+\rangle$ and $|-\rangle$ (i.e., changes in α and β) shift this single Lorentzian by an energy $\frac{\delta_n}{2}(-\frac{\delta_n}{2})$ away from $\frac{E_+ - E_-}{2}$ when detecting $\sigma^+(\sigma^-)$ polarized PL. As the dependence of δ_n on $P_{\rm exc}$ is nonlinear [10], it is more instructive to plot the polarizations P_c [Fig. 2(a)] and P_l [Fig. 2(b)] achieved for X^0 as a function of the created field $B_N \propto \delta_n$ (assuming an electron g factor of $|g_e| = 0.48$ [11]). A particularity of this plot is that the sign of B_N changes when switching from σ^+ (electron $\downarrow$ injection) to σ^- (electron $\uparrow$) laser polarization. The data in Fig. 2 are very well reproduced using $|P_c^0| = 33\%$ as the only fitting parameter in Eqs. (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 $\delta_n \rightarrow \infty$. 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 from $|X\rangle$ and $|Y\rangle$ [9].

Next we discuss the origin of the DNP that builds up via repeated electron-nuclear spin flip-flops through the fluctuating term $(I_-^k S_+ + I_+^k S_-)$ in Eq. (1) [1,11]. Spin flip is very costly in energy for an electron in a X^0 [4], as the bright and dark states (for example $|\Uparrow, \downarrow\rangle$ and $|\Uparrow, \uparrow\rangle$) are separated by the isotropic CE energy of up to $\delta_0 \simeq 500$ μeV at zero magnetic field in InGaAs dots [22]. As a result the probability for electron-nuclear spin flip-flops is very low. We confirm below that the X^0 is not at the origin of the DNP in our sample, but merely experiences the existing field B_N in the dot.

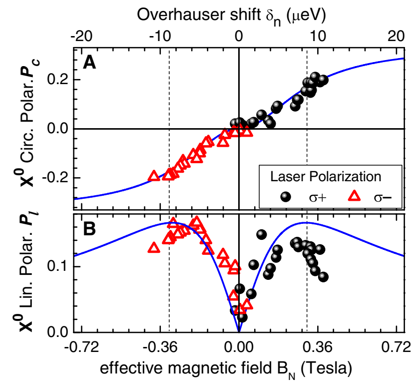

FIG. 2 (color online). QD I. $E_{\rm laser} = 1.44$ eV. $\delta_1 = -9$ μeV. (a) Circular polarization P_c and (b) Linear polarization P_l for X^0 as a function of B_N for σ^+ (σ^-) laser polarization shown as solid circles (hollow triangles) assuming $|g_e| = 0.48$. Solid lines calculated with Eqs. (2) and (3). Dashed vertical line: measured value of $|\delta_1|$.

The PL in Figs. 1(a) and 1(b) shows that the dot is occupied alternately by X^0 and X^+[23]. 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 a hole is trapped for $t \leq \tau_r$, the X^+ exciton is formed, if not, the X^0 will recombine. 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 CE 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 [24], the electron spin of the X^0 experiences the same B_N as the electron in the X^+.

We compare PLE measurements on dot I (Figs. 1 and 2 and upper part of 3) with another dot II with a much larger splitting $\delta_1 = 34$ μeV (see Fig. 3 lower part). P_c in the order of 50% is created for the X^+ in both dots when exciting with $E_{\rm laser} = 1.425$ to 1.465 eV. In the low energy tail of the density of states of the WL at $E_{\rm laser} \simeq 1.41$ eV, the carrier absorption rate is too low to create DNP [11,25]. At $E_{\rm laser} \geq 1.48$ eV the P_c drops in absolute value and even changes sign (δ_n changes sign accordingly) as the light hole transitions in the WL are excited [16]. Comparing Figs. 3(g) and 3(h) for dot II shows clearly that the P_c created for the X^+ is transferred to the nuclear spins.

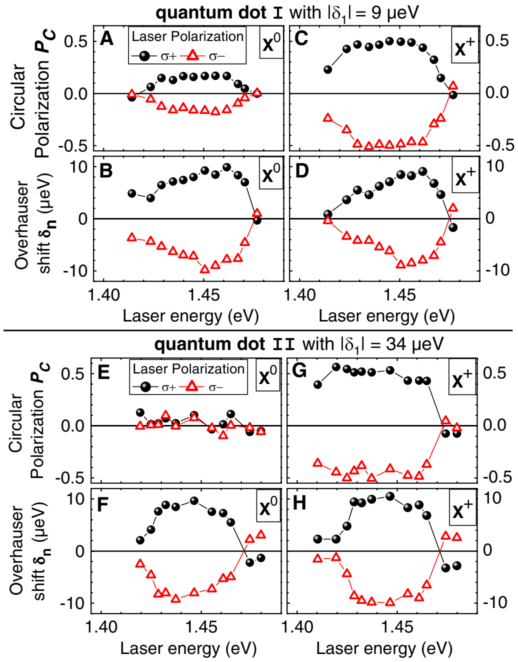

FIG. 3 (color online). Circular polarization P_c and Overhauser shift δ_n as a function of $E_{\rm laser}$ for σ^+ (σ^-) laser polarization shown as solid circles (hollow triangles) for (a),(b) X^0 of dot I, (c),(d) X^+ of dot I, (e),(f) X^0 of dot II and (g),(h) X^+ of dot II. PL detection energy dot I (II) $\simeq$ 1.358 eV ($\simeq$1.342 eV).

In stark contrast, the P_c for the X^0 is on average zero in Fig. 3(e). The X^0 in both dots is subject to a B_N of several hundred mT [Figs. 3(b) and 3(f)], 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. So the circular polarization P_c shown in Fig. 3(a) for dot I is due to the nuclear field B_N present in the dot, and not vice versa.

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 nonresonant 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. The main criteria for the polarization conversion scheme to work are (i) alternating presence (ideally with a deterministic control of the charge state [2]) of the neutral X^0 and charged excitons X^+ (X^- should also be suitable [12]); (ii) the anisotropic exchange splitting δ_1 and maximum Overhauser shift $\delta_n^{\rm MAX}$ should be of similar magnitude; and (iii) the electron spin states should be stable during carrier capture and radiative lifetime. Considering the slow evolution of B_N [24] 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 [26].

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[18] Light hole levels in the dot are several tens of meV away in energy due to strain and are not considered here.
[19] Although resonant PL experiments show that a linearly polarized excitation does result in linearly polarized PL, the coherent superposition between $|\Uparrow, \downarrow\rangle$ and $|\Downarrow, \uparrow\rangle$ created in nonresonant excitation is lost during carrier capture. The spin information is solely carried by the electron and we have found no correlation between the linear polarization of the laser and P_c or P_l of the PL.
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