Spin polarization of the neutral exciton in a single InAs quantum dot at zero magnetic field

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A high degree of spin polarization for the neutral exciton in individual quantum dots, at zero external magnetic field, is monitored. While a high polarization degree is commonly observed for the charged exciton, a negligible polarization has been predicted for the neutral exciton. The exceptionally high polarization (>60%) observed here is explained in terms of a dynamical nuclear polarization field, stabilizing the electron spin. Such polarization of the quantum dot nuclei, in case of the neutral exciton, is possible due to unequal capture time of electrons and holes.

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The spin of a single carrier localized in a semiconductor quantum dot (QD) has been suggested as a building block for future memory and quantum computer operation.[1] In particular, the spin of an electron confined in a QD is a good candidate for these applications because of cancellation of classical spin-relaxation mechanisms.[2] The state of the spin of recombining particles can be directly measured by monitoring the degree of circular polarization (ρ_c) in photoluminescence (PL) experiments. For the case of neutral excitons in QDs, a negligible ρ_c has been predicted at zero external magnetic field ($\boldsymbol{B_{ext}}$) due to strong anisotropic electron (e)-hole (h) exchange interaction.[3–7] Conversely, for the case of charged excitons, the anisotropic e-h exchange interaction is suppressed[5,8] and an essential ρ_c is expected and has been confirmed in experiments on individual In(Ga)As/GaAs QDs. Up to now, a very low ρ_c were recorded at $\boldsymbol{B_{ext}}=0$ for the neutral exciton in experiments with QD ensembles[6,9] or individual QDs.[3,8,10–12]

In the present report, a high degree ($\rho_c \approx 60\%$) of spin polarization is achieved for the neutral exciton at $\boldsymbol{B_{ext}}=0$ in individual InAs QDs, studied by micro-photoluminescence (μ-PL). The anomalously high ρ_c is explained by a separate capture of e's and h's into the QD. This provides a time interval, when the QD is occupied with solely an electron, which can polarize the lattice nuclei. In turn, a nuclear magnetic field ($\boldsymbol{B_N}$), called the Overhauser field, acting upon electrons will effectively stabilize the electron spin and result in the high ρ_c observed in the experiments. Further, the electron magnetic field ($\boldsymbol{B_e}$), the Knight field, acting upon the nuclei in the QD has been directly measured to be ≈ 13 mT. These experiments demonstrate a possible way to bypass the anisotropic e-h exchange interaction in QDs and thereby allowing spin conservation of the neutral exciton

The sample studied was grown by MBE with self-assembled InAs QDs positioned between GaAs barriers.[13] To create spin-polarized carriers, the sample was excited with circular-polarized light (σ^+ and/or σ^-). The degree of circular polarization of the PL is given by: $\rho_c=(I_{co}-I_{cross})/(I_{co}+I_{cross})$, where I_{co} (I_{cross}) is the spectrally integrated PL component of co(cross-)circular polarization, with respect to the helicity of the excitation light.

Figure 1(a) shows a μ-PL spectrum of the wetting layer (WL) emission. In the same panel, ρ_c of the neutral exciton as a function of excitation energy ($h\nu_{ex}$) is presented. It should be stressed that for all $h\nu_{ex}$, the excitation power was tuned in order to maintain at the same integrated PL intensity from the QD (to compensate for the change in sample absorption). When $h\nu_{ex}$ is varied, ρ_c remains positive [Fig. 1(a)], in striking contrast to negatively charged excitons in

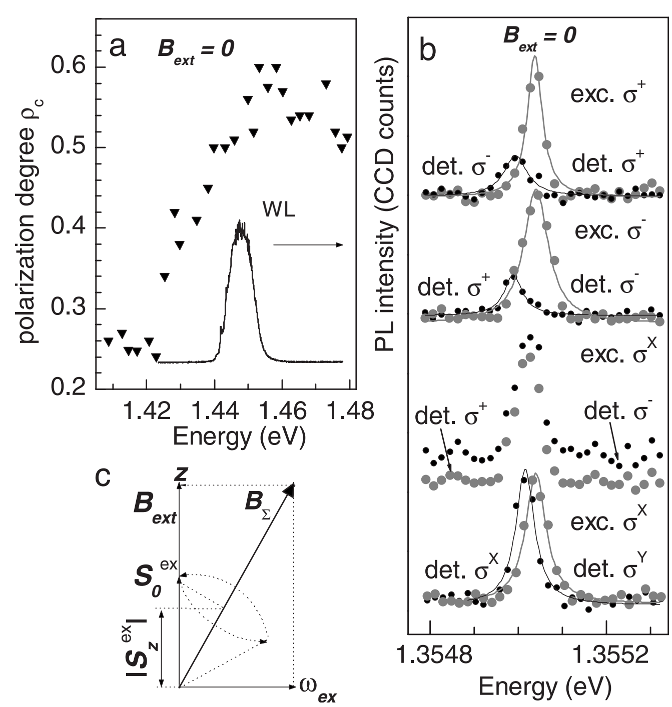

FIG. 1. (a) The symbols show ρ_c as a function of excitation energy measured at σ^+ cw excitation, with $P_{ex}=2$ μW for $h\nu_{ex} > E_{\mathrm{WL}}$ and with increased P_{ex} for $h\nu_{ex} < E_{\mathrm{WL}}$ (see text). The line shows a μ-PL spectrum of the WL measured at $h\nu_{ex}=1.684$ eV and at $P_{ex}=40$ nW. (b) μ-PL spectra (symbols) of a QD measured at $h\nu_{ex}=1.463$ eV, $P_{ex}=2$ μW and at different polarization configurations in excitation and detections paths as indicated in the figure. The solid lines are fit with Lorentzian curves to the measured μ-PL spectra. (c) A schematic illustration of the pseudospin model of a neutral exciton exposed to an external magnetic field, as explained in the text. All data in (a) and (b) were recorded at $T=4.2$ K and $\boldsymbol{B_{ext}}=0$.

QDs, which exhibit a negative ρ_c.[9,12,14–17] A set of typical μ-PL spectra of the neutral exciton[18] for $h\nu_{ex}$ in the range of the WL emission energy (E_{WL}), and for different polarization configurations, is shown in Fig. 1(b). At circular-polarized excitation, the integrated PL intensity of the cocircular-polarized component appears much stronger than the cross-circular one, illustrating a high and positive degree of ρ_c.

The high ρ_c observed here is remarkable, since the bright states ($|+1\rangle$ and $|-1\rangle$) of the heavy-hole neutral exciton is known to be mixed due to the in-plane asymmetry of a QD ($|+1\rangle$ and $|-1\rangle$ correspond to spin projections onto the z axis, which are chosen along the growth axis of the sample). This mixing creates two linearly polarized dipoles $|\mathrm{X}\rangle = 2^{-1/2}(|+1\rangle + |-1\rangle)$ and $|\mathrm{Y}\rangle = 2^{-1/2}(|+1\rangle - |-1\rangle)/i$, which in case of InAs/GaAs QDs emit light along the $\langle 110\rangle$ and $\langle 1\underline{1}0\rangle$ crystallographic directions.[19,20] $|\mathrm{X}\rangle$ and $|\mathrm{Y}\rangle$ are split by the anisotropic e-h exchange energy ($\hbar\omega_{ex}$) (Ref. 21) and the bright states are separated in energy from the dark states. Thus, at $\boldsymbol{B_{ext}}=0$ the neutral exciton is expected to reveal two orthogonal linearly polarized components and only at an elevated external magnetic field ($\boldsymbol{B_{ext}} \| z$), the mixed states $|\mathrm{X}\rangle$ and $|\mathrm{Y}\rangle$ will transform into "pure" $|+1\rangle$ and $|-1\rangle$ states, giving rise to circular-polarized emission. Upon excitation with linearly polarized light (σ^{X}) at $\boldsymbol{B_{ext}}=0$, two orthogonal and linearly polarized PL lines, separated by $\hbar\omega_{ex} \approx 25$ μeV, are indeed monitored [Fig. 1(b)].

To explain the high ρ_c observed here, we adopt the vector model for the exciton pseudospin developed in Refs. 4 and 7 [see Fig. 1(c)]. Here the anisotropic e-h exchange interaction is viewed as an in-plane magnetic field ($\boldsymbol{\omega_{ex}}$) and the vector $\boldsymbol{S_0^{ex}}$ corresponds to the initial exciton spin. Since the precession time [$\tau_b = 2\pi/\omega_{ex} \approx 165$ ps (Ref. 6)] of $\boldsymbol{S_0^{ex}}$ around $\boldsymbol{\omega_{ex}}$ is smaller than the exciton decay time [$\tau_d \approx 800$ ps (Ref. 19)], the exciton spin ($\boldsymbol{S_0^{ex}}$) will accomplish many turns around $\boldsymbol{\omega_{ex}}$ before recombination.[20] Hence ρ_c (proportional to the projection of the exciton pseudospin onto the z axis, $|\boldsymbol{S_z^{ex}}|$) is predicted to be negligible.[4,6,7] However, as stated above, an external magnetic field of sufficient strength $|\boldsymbol{B_{ext}}|\mu_B g_{ex} > \hbar\omega_{ex}$ ($\mu_B \approx 58$ μeV/T is the Bohr magneton and g_{ex} is the neutral exciton g factor) applied in Faraday geometry ($\boldsymbol{B_{ext}} \| z$) "restores" the polarization of the neutral exciton because of decoupling of the $|+1\rangle$ and $|-1\rangle$ states.[6,7] At increasing $|\boldsymbol{B_{ext}}|$, the $|+1\rangle$ and $|-1\rangle$ states are separated in energy by $\Delta E = \{(|\boldsymbol{B_{ext}}|\mu_B g_{ex})^2 + (\hbar\omega_{ex})^2\}^{1/2}$,[21] which gives rise to a nonvanishing value of ρ_c. In the vector model, an application of $\boldsymbol{B_{ext}}$ initiates the precession of the exciton pseudospin around the total magnetic field $\boldsymbol{B_\Sigma} = \boldsymbol{B_{ext}} + \boldsymbol{\omega_{ex}}$ leading to a nonzero value of $|\boldsymbol{S_z^{ex}}|$ and hence of ρ_c.

From the μ-PL spectra [Fig. 1(b)], an obvious energy separation (≈ 50 μeV) is recorded between the σ^- and σ^+ components at circular-polarized excitation despite $\boldsymbol{B_{ext}}=0$. This is the signature of an effective magnetic field ($\boldsymbol{B_N}$) in the sample with a projection ($\boldsymbol{B_N^z}$) onto the z axis. When the excitation helicity is reversed, the polarization-resolved PL components exchange their spectral positions, clearly demonstrating that $\boldsymbol{B_N^z}$ has reversed its direction. The contribution of $|\boldsymbol{B_N^z}|$ to ΔE, the Overhauser shift (OHS), can be estimated from $\mathrm{OHS} = \{(\Delta E)^2 - (\hbar\omega_{ex})^2\}^{1/2} \approx 42$ μeV.

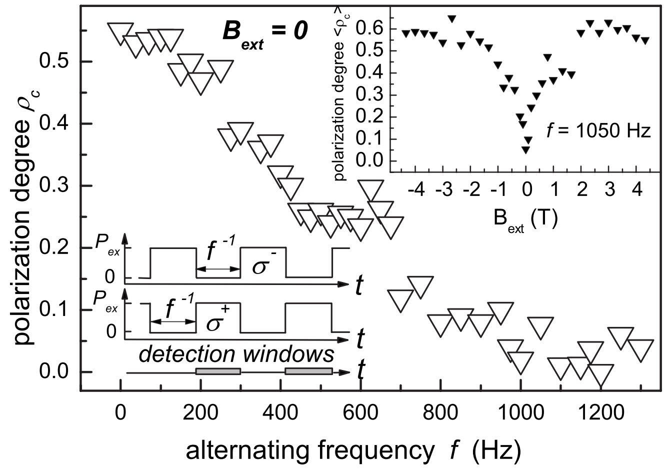

FIG. 2. ρ_c as a function of the alternating frequency, f, measured during the time windows corresponding to an excitation with σ^+, at $P_{ex}=2$ μW, $h\nu_{ex}=1.463$ eV, $T=4.2$ K, and at $\boldsymbol{B_{ext}}=0$. The upper right inset shows $\langle\rho_c\rangle$ as a function of $\boldsymbol{B_{ext}} \| z$ measured at $P_{ex}=2$ μW, $h\nu_{ex}=1.463$ eV, $T=4.2$ K, and at $f=1050$ Hz. The lower left inset illustrates the excitation procedure with light pulses of alternating helicity. The PL signal is only accumulated within the detection time windows corresponding to σ^+ (or σ^-) excitation.

To further elucidate the existence of a magnetic field in the sample at σ^+ and/or σ^- cw excitation and to measure the build-up time of $\boldsymbol{B_N^z}$, the QD was excited by a beam with alternating σ^+ and σ^- polarization with frequency, f. The QD was accordingly exposed to σ^+ light during the time, $\Delta t = f^{-1}$, followed by σ^- light (of the same power). Detection of the two circular-polarized PL components was performed within the time intervals corresponding to only σ^+ (or σ^-) excitation windows (see the lower inset of Fig. 2).

The polarization degree ρ_c, recorded with σ^+ excitation, remains approximately the same for $f < 100$ Hz, but decreases progressively for $f > 100$ Hz to stabilize at a few percent at $f > 1000$ Hz (Fig. 2). $\Delta t \approx 10$ ms is accordingly sufficient for $\boldsymbol{B_N}$ to buildup, while for $\Delta t < 1$ ms, $\boldsymbol{B_N}$ is negligible. The dependence of the averaged polarization degree ($\langle\rho_c\rangle$) (Ref. 22) on the external magnetic field $\boldsymbol{B_{ext}} \| z$ (upper inset in Fig. 2) shows that $\langle\rho_c\rangle$ changes symmetrically to reach about 55% at $|\boldsymbol{B_{ext}}| \approx 1.5 \div 2$ T. Based on this experiment, $|\boldsymbol{B_N^z}|$ determining ρ_c at cw excitation is predicted to be of the same order. To understand the origin of $\boldsymbol{B_N}$, a concept of dynamic polarization of lattice nuclei by optically oriented electrons is employed.[23] This effect originates from the coupling of electron and nuclear spins through the hyperfine Fermi interaction, while the corresponding interaction of nuclear spins with a hole is considerably weaker.[23] Hence, the nuclear-hole interactions will be excluded from further discussion. The interaction between a single electron and a large number (10^4–10^5) of nuclei (N) in the QD results in a dynamical nuclear spin polarization leading to the appearance of spin-oriented nuclei, which is equivalent to building up an effective magnetic field acting upon the electron localized in the QD. Nuclear fields as large as several Tesla have been detected for In(Ga)As/GaAs QDs.[3,24,25] The rise time of the nuclear polarization at $\boldsymbol{B_{ext}}=0$ in In(Ga)As/GaAs QDs has earlier been experimentally determined to be 9.4 ms.[26] This value agrees well with the time scale for the buildup of $\boldsymbol{B_N}$ in our experiments.

Accordingly, the field $\boldsymbol{B_N}$ introduced above is identified as a nuclear field grown up in the QD upon cw excitation with circular-polarized light, which injects spin-oriented electrons into the QD. Consequently, no $\boldsymbol{B_N}$ is predicted in the QD under linearly polarized excitation (i.e., photoexcited electrons appear nonpolarized). Hence, the σ^+ and σ^- PL components are not expected to be split nor exhibit any measurable value of ρ_c, as consistent with our experimental observations [Fig. 1(b)]. Since $\boldsymbol{B_N}$ influences only electrons, the Overhauser shift should be defined as OHS $=|\boldsymbol{B}_N^z|\mu_B|g_e|$,[25,27] where g_e is the electron g factor. Adopting $|g_e|=0.5\div 0.6$ (Refs. 3 and 24) and OHS≈ 42 μeV, one can evaluate $|\boldsymbol{B}_N^z|\approx 1.2\div 1.4$ T, in satisfactory agreement with the predictions made above. The experimentally estimated exciton polarization, ρ_c, is entirely determined by the averaged electron spin ($\boldsymbol{S}$) according to $\rho_c=2|\boldsymbol{S}_z|$,[19] where $\boldsymbol{S}_z$ is the projection of $\boldsymbol{S}$ onto the z axis. This is in agreement with the assumption that, for excitation into the WL, the electron spin is preserved during the capture and relaxation processes in the QD, while the hole spin orientation is lost.[5,9,15] It should be emphasized that a buildup of nuclear polarization has been demonstrated earlier for the case of the neutral exciton.[27] However, this was achieved at a nonzero field, $\boldsymbol{B_{ext}}$, and the nuclear polarization was determined by spin flip assisted radiative recombination of dark excitons.

To explain the experimentally observed fact that a relatively strong nuclear magnetic field ($1.2\div 1.4$ T) is builtup in the QD even for the case of a neutral exciton at $\boldsymbol{B_{ext}}=0$, the preceding step to the formation of the exciton is considered as a process of separate capture of e's and h's as was demonstrated in our previous studies on QDs (Ref. 13) as well as by others (e.g., Refs. 28–30 and references therein). The parameter, $\Delta\tau_{e\text{-}h}$, is the difference in capture times between e's (τ_e) and h's (τ_h) into the QD. Since both e's and h's are excited into the WL, these times could be estimated as $\tau_{e(h)}\approx L_{e(h)}/V_{e(h)}$. Here $L_{e(h)}$ corresponds to the collection length for individual e's (h's) into the QD and $V_{e(h)}$ is the e's (h's) velocity in the WL plane, as was earlier deduced to be 1.6×10^7 (3.1×10^6) cm/s for the same sample.[31] For our experimental conditions (with one QD located within the area of the laser spot) $L_e=L_h\approx 1$ μm can be assumed (i.e., half the diameter of the laser spot) and, hence τ_e $(\tau_h)\approx 6(32)$ ps resulting in $\Delta\tau_{e\text{-}h}=\tau_h-\tau_e\approx 26$ ps. It should be noted that $\Delta\tau_{e\text{-}h}$ represents an expected PL rise time for a QD, experimentally determined to be $30\div 50$ ps (Refs. 9 and 32) in reasonable agreement with our estimate, $\Delta\tau_{e\text{-}h}\approx 26$ ps. Accordingly, before the recombination of an exciton, the QD is assumed to be populated with only an electron for ≈ 26 ps. The fraction of time (Γ_e) with single electron occupancy in the QD, is defined as $\Gamma_e=\Delta\tau_{e\text{-}h}/\tau_r$, where τ_r is the average time between two subsequent exciton formation events. For an excitation power slightly below the biexciton formation level, one can estimate $\tau_d\geq\tau_r$ because to form the biexciton, the formation of a second exciton in the QD is required before the first exciton recombines. Hence, $\tau_r\approx\tau_d\approx 800$ ps is used giving $\Gamma_e\approx 0.0325$ which is in reasonable agreement with other reports.[3,25]

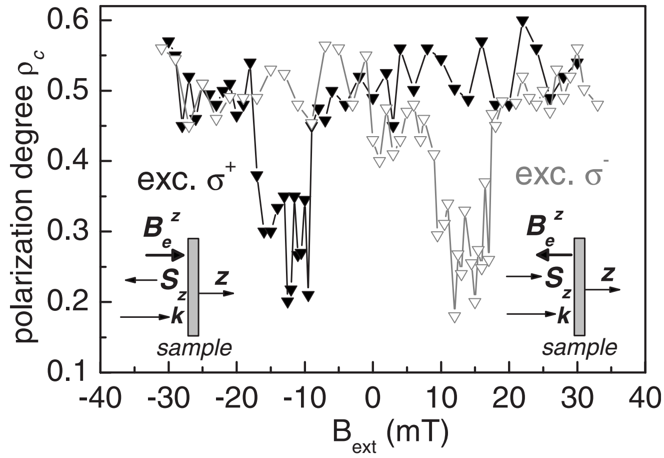

FIG. 3. ρ_c as a function of B_{ext} ($\boldsymbol{B_{ext}}\parallel z$) measured at cw excitation with σ^+ (black) and σ^- (gray) at $P_{ex}=2$ μW, $h\nu_{ex}=1.463$ eV, and $T=4.2$ K. Positive (negative) B_{ext} correspond to $\boldsymbol{B_{ext}}$ parallel (antiparallel) to the direction of the laser beam. The arrows indicate the directions of the vectors $\boldsymbol{k}$, $\boldsymbol{S}_z$, and $\boldsymbol{B}_e^z$.

To check the idea on separate carrier capture times, $\Delta\tau_{e\text{-}h}$, determining the possibility for $\boldsymbol{B_N}$ to buildup, $\Delta\tau_{e\text{-}h}$ is decreased. This is achieved by exciting directly into the QD, i.e., $h\nu_{ex}<E_{\text{WL}}$, resulting in a considerable decrease in the length $L_{e(h)}$. The results demonstrate a gradual reduction in ρ_c, down to ≈ 0.25 (in the range 0.05–0.25 for different QDs) upon decreasing $h\nu_{ex}$ down to ≈ 1.41 eV [Fig. 1(a)]. Additionally, experiments on QD ensembles show a monotonous decrease in ρ_c with an increasing QD density (i.e., when $L_{e(h)}$ is no longer determined by the laser spot size, but rather by the averaged interdot distance). These observations support our model with separate carrier capture into the QD determining the nuclear field buildup.

The nuclear magnetic field $\boldsymbol{B_N}$ acting upon an electron in the QD and the electron field $\boldsymbol{B_e}$ acting upon each nucleus are consequences of the same process of dynamical polarization of nuclei by spin-oriented electrons. This circumstance allows $\boldsymbol{B}_N^z$ to be expressed in the following form:[33–35]

$$\boldsymbol{B}_N^z=\alpha\{(\boldsymbol{B_{ext}}+\boldsymbol{B_e})\cdot\boldsymbol{S}\}\{(\boldsymbol{B_{ext}}+\boldsymbol{B_e})^2+B_L^2\}^{-1}(\boldsymbol{B}_{ext}^z+\boldsymbol{B}_e^z) \quad (1)$$

where α is a proportionality constant, $\boldsymbol{B_e}=b_e\boldsymbol{S}$, $\boldsymbol{B}_{ext}^z$ ($\boldsymbol{B}_e^z$) is the projection of $\boldsymbol{B_{ext}}$ ($\boldsymbol{B_e}$) on the z axis, b_e is to be evaluated below, and B_L is the effective magnetic field caused by the nuclear spin-spin interactions [estimated to be ≈ 0.3 mT for InAs/GaAs QDs (Ref. 14)]. $\boldsymbol{B_e}$ is related to ρ_c in the following way: $|\boldsymbol{B}_e^z|=|b_e|\cdot|\boldsymbol{S}_z|=1/2|b_e|\cdot\rho_c$. The average interaction energy of an electron spin S with N nuclei of the same species, assuming that their mean spins ($\boldsymbol{I_{av}}$) are equal, is expressed as: $A\boldsymbol{I_{av}}\boldsymbol{S}$,[23] where A is the hyperfine constant. Taking this quantity as the nuclear spin energy in an electron field, $\boldsymbol{B_e}$, one obtains: $A\boldsymbol{I_{av}}\boldsymbol{S}=-N\hbar\gamma\boldsymbol{I_{av}}\boldsymbol{B_e}$ and, hence $\boldsymbol{B_e}=-\boldsymbol{S}A/(N\hbar\gamma)$, where γ is the nuclear gyromagnetic ratio. A/γ is estimated as $1/2\Sigma_j(A_j/\gamma_j)$, where j numerates In and As. The number of nuclei in a QD is assumed to be $N\approx 5\times 10^4$.[3,5,25,26,36] With γ_{In} $(\gamma_{\text{As}})=5.86(4.58)\times 10^7$ rad T^{-1} s^{-1} (Ref. 37) and A_{In} $(A_{\text{As}})=56(46)$ μeV,[3] $b_e\approx -30$ mT is derived. Hence for fully polarized electron spin ($\rho_c=1$) $|\boldsymbol{B}_e^z| \approx 15$ mT and for our experimental conditions ($\rho_c=0.55$) $|\boldsymbol{B}_e^z|$ is estimated as ≈ 8.3 mT.

It follows from Eq. (1) that at $\boldsymbol{B}_{ext}=0$, $|\boldsymbol{B}_e|$ should considerably exceed B_L to achieve a significant value of $|\boldsymbol{B}_N|$. Second it follows that $|\boldsymbol{B}_N^z|$ should vanish (and accordingly ρ_c should decrease) at $\boldsymbol{B}_{ext}^z=-\boldsymbol{B}_e^z$. To check this idea, $\boldsymbol{B}_{ext}\| z$ was applied (see Fig. 3). In this dependence of ρ_c on $|\boldsymbol{B}_{ext}|=B_{\text{ext}}$ for σ^- and σ^+ excitations, distinct minima in ρ_c are monitored at $|\boldsymbol{B}_{ext}|$ from 8 up to 18 mT. Thus, the average value $|\boldsymbol{B}_e^z|=13$ mT is chosen as the strength of the electron field, in satisfactory agreement with the predictions for $|\boldsymbol{B}_e^z|$ made above. It should be noted that the dips in ρ_c are observed at opposite directions of $\boldsymbol{B}_{ext}$ when the excitation is changed from σ^+ to σ^- and that the compensation of $\boldsymbol{B}_e^z$ by $\boldsymbol{B}_{ext}^z$ takes place at the predicted directions of $\boldsymbol{B}_e^z$. The σ^+ (σ^-) excitation creates $\boldsymbol{S}_z$ pointing antiparallel (parallel) to the direction of the laser beam ($\boldsymbol{k}$), accordingly $\boldsymbol{B}_e^z\uparrow\uparrow(\uparrow\downarrow)\boldsymbol{k}$ for these excitation conditions. Finally, the experimental observation that $|\boldsymbol{B}_e^z| \gg B_L$ supports the idea that an essential nuclear magnetic field $\boldsymbol{B}_N$ could be achieved even for zero external magnetic field, as evidenced by the high degree of circular-polarization measured for the neutral exciton.

To conclude, we report on a high degree of circular polarization for the neutral exciton upon circular-polarized excitation, without any external magnetic field applied. This is explained in terms of the buildup of a nuclear magnetic field in the QD, which stabilizes the electron spin. The possibility to polarize the QD nuclei is shown to be due to the unequal capture times of electrons and holes.

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