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Manipulating the spin polarization of excitons in a single quantum dot by optical means

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Circular polarization studies of photoluminescence from the neutral (X^0) and the positively charged (X^+) excitons are reported for individual InAs/GaAs quantum dots (QDs). High polarization degrees, 60% for X^0 and 73% for X^+, were recorded without any external magnetic field applied. These studies show that the QD polarization and population dynamics are controllable either by varying the photoexcitation intensity or by using a second IR laser excitation. © *2011 American Institute of Physics*. [doi:10.1063/1.3554422]

Studies of spin preservation of a single carrier localized in a semiconductor quantum dot (QD) are of increasingly interest during the past decade due to the opening of fascinating physics and due to the potential applications in spin-based nanoscale devices for quantum computer operations.[1] The spin of an electron is the best candidate for these applications because classical spin relaxation mechanisms are canceled for an electron confined inside a QD.[2] The spin state 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 (X^0) in QDs, ρ_c at zero external magnetic field (B_{ext}) is expected to be negligible due to the strong electron-hole anisotropic exchange interaction (ω_{ex}).[3–7] Conversely, for charged exciton complexes, an essential ρ_c is expected and has been measured since ω_{ex} is suppressed.[5,8,9] Recently, however, a nonzero ρ_c of X^0 in InGaAs QDs was observed.[10] In Ref. 10, the PL spectra involve both X^0 and the positively charged exciton (X^+) but it is the presence of X^+ that leads to the appearance of nuclear polarization, while X^0 simply experiences the resulting nuclear polarization field (B_N). However, in the present study, we propose that solely X^0 is sufficient to create B_N.

In this letter, ρ_c of X^0 in individual InAs/GaAs QDs is monitored and manipulated by pure optical means. A surprisingly high ρ_c (60%) is demonstrated for X^0 at $B_{ext}=0$, which is explained in terms of the generation of B_N in the QD by spin-polarized electrons. This field stabilizes the electron spin by suppressing ω_{ex} and, hence, plays a similar role as B_{ext} employed by others[6,7] to "restore" ρ_c of X^0. The ability to build up B_N for X^0 even at $B_{ext}=0$ is indeed surprising (see, e.g., Ref. 9) but has been ascribed to the faster capture of electrons as compared to holes into the QD providing a time interval, when the QD is populated by a sole electron, which can then polarize the QD lattice nuclei.[11,12] In contrast to single laser excitation, which only results in X^0, when adding a second infrared (IR) laser excitation X^+ is formed, as was demonstrated by us earlier.[13] An even higher polarization is measured for X^+, $\rho_c=73\%$, which is considered to be an upper bound for spin polarization of electrons captured into the QD from the wetting layer (WL).

The sample under study was grown by molecular beam epitaxy and consists of 1.7 monolayers of InAs constituting the WL with a low density of InAs QDs ($\approx10^6$ cm^{-2}) positioned between GaAs barriers. To excite and collect the PL, a conventional microphotoluminescence (μ-PL) setup, operating at $T=3.9$ K, was used. A Ti:sapphire laser with excitation energy $h\nu_{exc}=1.465$ eV (slightly higher than the WL band gap) and a semiconductor laser with excitation energy $h\nu_{IR}=1.17$ eV (smaller than any QD related interband transition) were used and focused to a diameter of ≈2 μm on the sample surface. Detailed descriptions of the sample and of the experimental setup are given elsewhere.[11–13] ρ_c was determined as $\rho_c=(I_{co}-I_{cross})/(I_{co}+I_{cross})$, where I_{co} (I_{cross}) corresponds to the spectrally integrated PL signal of the co- (cross-)circular components.

Upon excitation with linearly polarized light (σ^X), two mutually orthogonally linearly polarized components (π_X and π_Y) are recorded, since X^0 is split into a pair of bright states by the anisotropic electron-hole exchange energy, $\Delta E_{ex}=\hbar\omega_{ex}=23$ μeV, as inferred from Fig. 1(a).[14] When instead detecting the circularly polarized σ^+ and σ^- PL components, no splitting of X^0 is expected nor is observed [Fig. 1(a)], since both the σ^+ and σ^- components represent a coherent mixture of the two linear states. However, upon excitation with σ^+-light, two circularly polarized components, exhibiting an essential spectral shift, $\Delta E_\Sigma=52$ μeV, are observed [Fig. 1(a)]. The remarkable fact that $\Delta E_\Sigma > \Delta E_{ex}$ implies the existence of an effective magnetic field (B_{eff}) in the sample, resulting in an additional Zeeman splitting between the two exciton components. This field has been generated by the excitation since $B_{ext}=0$ in the present study. The origin of B_{eff} is attributed to B_N, which is generated in the QD. We stress that the generation of B_N at $B_{ext}=0$ is only possible in case of an essential electron Knight field experienced by the nuclei.[9] Upon excitation with σ^-, the spectral positions of the σ^+ and σ^- PL components are reversed (not shown here). This fact supports the identification of B_{eff} as B_N because the orientation of the nuclear spin is determined by the direction of the optically generated electron spin. Along with the large splitting, ΔE_Σ, the lower pair of spectra in Fig. 1(a) also show a large difference in amplitude, which corresponds to a high ρ_c (60%). This experimental observation appears surprising at a first glance, since ρ_c is predicted to be negligible

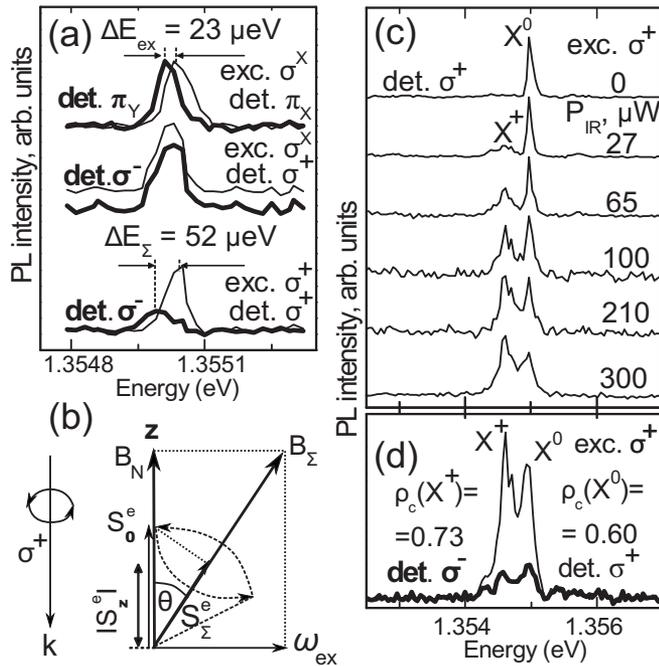a)Electronic mail: alarsson@ifm.liu.se.

FIG. 1. (a) μ-PL spectra of a QD measured at different polarization configurations in the excitation and detection paths as indicated in the figure. (b) A schematic illustration of the vector model of an electron spin exposed to a nuclear magnetic field and to the effective magnetic field of the anisotropic electron-hole exchange interaction. (c) μ-PL spectra of a QD measured at dual laser excitation and for different P_{IR} as indicated in the figure. (d) μ-PL spectra of a QD measured at dual laser excitation (with $P_{IR} = 300$ μW), at σ^+-polarized excitation, and σ^+- (thin) and σ^-- (thick) polarized detections. $P_{ex} = P_{ex}^* = 3$ μW in (a), (c), and (d).

for X^0 in QDs at conditions when no charged exciton is observed.[3–7,10]

The high value of ρ_c recorded for X^0 in the present study is explained by considering the electron spin (S^e) within a vector model for the exciton pseudospin (S_{ex}) (Refs. 4 and 7) [Fig. 1(b)]. Here, ω_{ex} is considered to behave as an effective magnetic field (ω_{ex}) directed in the plane of the sample, i.e., perpendicularly to the z-axis. If the sample is illuminated with a circularly polarized laser beam, excitons with S_{ex} parallel to z are created. For a neutral exciton captured into a QD, S_{ex} will start to precess around ω_{ex} with the period $\tau_b = 2\pi/\omega_{ex} \approx 180$ ps (for $\hbar\omega_{ex} = 23$ μeV). For the exciton decay time $\tau_d \approx 800$ ps,[15] the time averaged projection of S_{ex} onto the z-axis (and the measured ρ_c) is expected to be zero. If an external field B_{ext} is applied parallel to the z-axis, S_{ex} will instead rotate around the total field $B_{ext} + \omega_{ex}$, which, in turn, will raise the polarization degree ρ_c from zero. In the present case, B_{ext} is replaced by the nuclear field B_N. After excitation, the electrons and holes propagate in the WL plane prior to capture into the QD. The electron spin does not relax during such a capture into the QD, whereas the initial hole spin is lost.[5,16,17] After capture, the spin of the electron in the QD (S_0^e) will rotate around the total magnetic field $B_\Sigma = B_N + \omega_{ex}$ with the average value S_Σ^e directed at an angle θ with respect to the z-axis [Fig. 1(b)]. Hence, S_0^e is influenced by two competing factors: ω_{ex}, which tends to decrease its projection onto the direction of the PL detection, and B_N, acting to preserve it. ρ_c is entirely determined by the projection of S_Σ^e onto the z-axis (S_z^e) and as a result $\rho_c = 2 \cdot |S_z^e|$.[15] It also follows from Fig. 1(b) that $|S_\Sigma^e| = |S_0^e| \cos\theta$ and $|S_z^e| = |S_0^e| \cos^2\theta$.

Thus, if $|S_0^e|$ could be estimated from the experiments, one could estimate ρ_c. In order to evaluate $|S_0^e|$, one needs to "switch off" ω_{ex}, which can be realized experimentally by employing dual laser excitation: a second IR laser was used, which has an insufficient energy to generate electron-hole pairs in the QD, but excites electrons from the valence band into deep levels in the GaAs barriers and thereby creates free holes that can be captured into the QD.[13] Consequently, X^+ is expected in the μ-PL spectra of the QD and in this case, ω_{ex} is switched off and a higher value of ρ_c, as compared with X^0, is predicted.

Figure 1(c) shows the evolution of the μ-PL spectra of the QD recorded at dual laser excitation conditions, obtained for a fixed P_{ex} with increasing excitation power of the IR laser (P_{IR}). Clearly, at $P_{IR} = 0$ (top spectrum), the spectrum only consists of the X^0-line and an increase of P_{IR} results in a progressive redistribution of the μ-PL spectra in favor of the X^+ line, which dominates the spectra at highest P_{IR} used in our experiments. Figure 1(d) shows the σ^+ and σ^- PL components of the μ-PL spectrum obtained at highest $P_{IR} = 300$ μW, which allowed us to derive $\rho_c = 0.73$ and $\rho_c = 0.60$ for the X^+ and X^0 lines, respectively. Taking $\rho_c = 0.73$ as the maximum degree of circular polarization (i.e., when ω_{ex} is canceled) to be achieved for the QD studied and $\cos^2\theta \approx 0.8$ from Fig. 1(b),[18] the expected ρ_c for X^0 can be evaluated to be $0.73 \cos^2\theta \approx 0.59$, which agrees well with the experimentally obtained value of $\rho_c = 0.60$ [Fig. 1(d)].

The spin pumping rate (W_S) from the electrons into the nuclear spin system (which determines ρ_c measured in our experiment) is directly proportional to the fraction of time that the QD is occupied with only an electron (Γ_e) and inversely proportional to the square of the energy separation (E_Z) between the electron levels with spin along and opposite to the z-axis.[3,19–21] Evidently, this fraction of time is $\Gamma_e = \Delta\tau_{e-h}/\tau_r$, where $\Delta\tau_{e-h} \approx 26$ ps is the difference in time between the electron and the hole capture into the QD[11,12] and τ_r is the "recycling" time, i.e., the averaged time between two adjacent events of exciton formation in the QD. To obtain the data in Fig. 1, P_{ex} was adjusted to be slightly smaller than $P_{ex} = 3$ μW (P_{ex}^*) at which the biexciton ($2X^0$) appears in the μ-PL spectra. Upon excitation with P_{ex}^*, one can approximate τ_r as τ_d because the observation of $2X^0$ should require that a second exciton should be formed in the QD before the first exciton recombines. Consequently, one can assume $\tau_r \approx \tau_d \approx 800$ ps (Ref. 15) and hence, $\Gamma_e \approx 0.0325$.

Since τ_r^{-1} is directly proportional to P_{ex}, a strong dependence of Γ_e and hence ρ_c on P_{ex} is unambiguously predicted. Indeed, a gradual increase from $\rho_c = 0$ (at minimum P_{ex}) to $\rho_c = 0.60$ measured at $P_{ex} = P_{ex}^*$ is demonstrated (Fig. 2). This behavior is explained in terms of a progressively increasing B_N with increasing P_{ex}. Obviously, this should simultaneously result in an increase of E_Z (contributing to ΔE_Σ), which is indeed in consistence with the experimental results (Fig. 2). The saturation of the parameter ΔE_Σ recorded at $P_{ex} \gtrsim P_{ex}^*$ (Fig. 2) could be understood in terms of a "negative feedback" mechanism[19]: an increase of $|B_N|$ and hence of E_Z should be accompanied by a decrease of W_S due to inverse proportionality of W_S and E_Z^2.[3,19–21] These two competing factors (the increase of Γ_e and W_S with increasing P_{ex} and the decrease of W_S with increasing E_Z^2) would explain the limited value of $|B_N|$ corresponding to only $\approx 15\%$ of polarized nuclei achieved in our experiments.
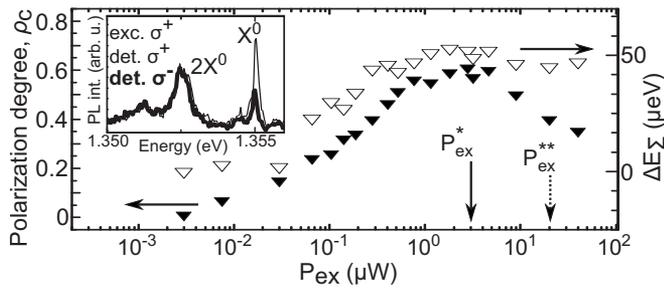
FIG. 2. The dependence of ρ_c (ΔE_Σ) on P_{ex} is shown by filled (open) symbols. The solid (dotted) vertical arrow indicates the excitation power used to obtain the data shown in Fig. 1 (inset in this figure). The inset shows μ-PL spectra of a QD measured at σ^+-polarized excitation, at σ^+-polarized excitation, and at σ^+- (thin) and σ^-- (thick) polarized detections. $P_{ex}=P_{ex}^{**}$ =21 μW in the inset.

An additional remarkable observation is the rather high value of ρ_c (40%) for X^0 recorded at $P_{ex}^{**} \approx 7 \times P_{ex}^*$, while $\rho_c=0$ is detected for $2X^0$ (inset of Fig. 2). This observation is explained by the symmetry of the $2X^0$ ground state, which consists of two electron-hole pairs having zero total spin. Consequently, the $2X^0$ will emit σ^+- and σ^--polarized photons with equal probabilities. The carrier-carrier scattering taking place inside the QD, when it is occupied with more than one electron-hole pair, is believed to explain the strong reduction of ρ_c for X^0 at P_{ex} above P_{ex}^* (Fig. 2).

Finally, the difference between the results presented here and in Ref. 10 should be highlighted; in Ref. 10, the QD nuclei are polarized by an electron in the presence of two holes (i.e., during the radiative lifetime of X^+), while in the present case, the nuclei are polarized by a sole electron (i.e., without a hole) in the QD.[11] When a hole is subsequently captured into the QD, the electron has the possibility to recombine with it, resulting in the X^0 PL line. The larger magnitudes for the nuclear and electron polarizations could be understood in terms of a different QD composition in our case (nominally pure InAs) with respect to the case in Ref. 10 (InGaAs), since B_N depends on the value of the nuclear spin. The spin of Ga and As is only 3/2, while the spin of In is 9/2.

In conclusion, the unexpectedly high degree of polarization ($\rho_c \approx 60\%$) of X^0 is explained in terms of the creation of a nuclear field, B_N, in the QD by spin-polarized electrons. B_N essentially decreases the destructive role played by the anisotropic electron-hole interaction, ω_{ex}, on the electron spin preservation. For the case of X^+, where ω_{ex} is naturally canceled, an even higher degree of polarization ($\rho_c \approx 73\%$) was achieved and taken as the upper bound of the electron spin

for the experimental conditions with optical excitation above the band gap of the WL.

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