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Spin polarization of neutral excitons in quantum dots: the role of the carrier

collection area

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A high degree ($\approx 55\%$) of circular polarization has been observed for the neutral exciton in InAs/GaAs quantum dots (QDs). The possibility to record non-zero polarization of the neutral exciton is explained in terms of different capture times of the light electron compared with the heavier holes into the QDs from the wetting layer. This interpretation is supported by the progressive reduction of the polarization degree with increasing QDs density, but also with increasing temperature.

1. Introduction

The increasing interest to manipulate the spin of the carriers in semiconductor quantum dots (QDs) is stimulated, to a large extent, by the hopes that electron/exciton spins could serve as a building block for quantum computing which eventually should develop a new spin-based electronics; so called “spintronics” [1]. Of particular interest are the conditions required to achieve long time scale preservation of carrier spin at zero external magnetic field (\mathbf{B}_{ext}). The degree of circular polarization (ρ_c) of the photo-luminescence (PL) directly reflects the carrier spin projection onto the observation axis. While a high degree (up to 95%) of polarization has been reported for the case when QDs are populated with charged exciton complexes [2, 3], an almost absent polarization was recorded for neutral excitons at the same experimental conditions, i.e. at $\mathbf{B}_{ext} = 0$. This was explained in terms of the destructive role played by the anisotropic electron-hole exchange interaction, which vanishes for the case of charged exciton complexes [2-4]. It should be noted that a recent interesting observation of a non-zero ρ_c for the neutral exciton at $\mathbf{B}_{ext} = 0$ was detected in single QDs (SQDs) in a case when the PL spectrum consists of not only the neutral exciton, but also the positively charged exciton complex [5]. The explanation to the observed non-zero ρ_c of the neutral exciton was a dynamic nuclear polarization created by the positively charged exciton, while the neutral exciton serves only as a monitor of the built up nuclear magnetic field (\mathbf{B}_N).

In our previous micro-photoluminescence (μ -PL) study of individual InAs/GaAs QDs, a high degree (up to 60 %) of polarization, ρ_c , was recorded for the PL emitted by neutral excitons [6]. This phenomenon was explained in terms of a rather strong (up to 1.4 T) \mathbf{B}_N built up to “stabilize” the electron spin, i.e. suppressing the anisotropic electron-hole exchange interaction. The mechanism for \mathbf{B}_N to build up even for the case of the neutral exciton in individual QDs

(SQD) was suggested to be due to the difference in capture times of electrons and holes, $\Delta\tau_{e-h}$, from the wetting layer (WL) into the SQD [6]. As a result, during the time $\Delta\tau_{e-h}$, the SQD is occupied with only an electron, which results in a dynamical nuclear polarization by spin-oriented electrons [7]. In SQD experiments (with only one QD located within the area of the laser spot), $\Delta\tau_{e-h}$ could be rather large (on the order of 30 ps).

In the present Letter, the crucial role of $\Delta\tau_{e-h}$ for the dynamical nuclear polarization is experimentally verified. The degree of ρ_c has been found to progressively decrease with increasing QDs density. This is explained by the fact that the typical size of the collection area for each QD within a multiple QDs (MQDs) ensemble of a given QD density is not determined by the size of the laser spot (as in case of SQD) but rather by the averaged inter-dot spatial separation. In addition, an increase of the temperature (T) from 4 K up to 90 K for excitation above the WL band gap has revealed a drop of ρ_c for the SQD from 55% down to zero already at $T \approx 30$ K, while for the case of MQDs ρ_c was decreased only slightly (by only ≈ 2 -3 times). These experimental findings are explained by the fact of considerable loss of spin orientation by the carriers propagating in the plane of the WL, prior to their capture into the QDs. This effect was directly demonstrated in our study by monitoring ρ_c of the PL emitted from the WL. Indeed, the efficiency of the temperature induced loss of spin orientation directly depends on the time which the carrier spends in the WL before capture into the QDs. This time is evidently decreasing with increasing QDs density.

2. Sample and Experimental setup

The sample under study was grown by Molecular Beam Epitaxy and consists of one InAs wetting layer (WL) of a nominal thickness of 1.7 InAs monolayers (0.5 nm) with InAs quantum dots of different density (from $\approx 10^6$ up to $\approx 10^9$ cm⁻²) positioned between two GaAs barriers (for

a detailed sample description, see Ref. 8). A conventional diffraction-limited micro-photoluminescence (μ -PL) setup allows studies of both SQDs and MQDs ensembles of different density. A Ti:Sapphire laser beam was focused down to a spot diameter of 2 μm on the sample surface by means of a micro-objective. The excitation energy of the laser ($h\nu_{ex}$) could be tuned in the range from 1.23 to 1.77 eV with a maximum excitation power (P_{ex}) of 3 mW as measured after the micro-objective. The sample was positioned inside a continuous-flow cryostat with a variable temperature range from 3.8 up to 300 K.

To excite the sample with circularly polarized light (σ^+ and/or σ^-), the originally linearly polarized laser beam was passed through a quarter-wave plate (Berek compensator) providing a high degree (≈ 98 -99 %) of circular polarization. The PL signal, collected through the same micro-objective, passed through another quarter wave plate combined with a Glan-Thomson linear polarizer (to analyze the PL signal with respect to its circular polarization) positioned before the entrance slits of the spectrometer. The degree of circular polarization, ρ_c of the PL was determined from $\rho_c = (I_{co} - I_{cross}) / (I_{co} + I_{cross})$, where I_{co} (I_{cross}) stands for the spectrally integrated PL signal of co- (cross -) circular polarization, with respect to the helicity of the excitation light. Seven individual QDs were inspected in the present study, which all showed similar characteristics. In this Letter, experimental data obtained for only one representative SQD is presented. 21 different sample spots with higher dot density were analyzed with respect to the degree of circular polarization.

3. Experimental Results and Discussion

Low-temperature μ -PL spectra of two sample spots, with only a SQD and a MQDs ensemble, respectively, are shown in Fig. 1. In the case of the SQD (Fig. 1 a), the μ -PL spectrum is dominated by the WL emission (peaked at an energy $E_{WL} \approx 1.45$ eV), while for the MQDs

(Fig. 1 b), the spectrally integrated intensities of the WL (I_{WL}) and of the QDs (I_{QD}) are of the same order, directly reflecting the increased QDs density. The single emission line from the SQD (Fig. 1 a) has earlier been identified [6] as the neutral exciton PL.

For excitation with circularly polarized light and with $h\nu_{ex}$ still above but very close to E_{WL} the PL spectrum of a SQD no longer consists of a single line, but rather of two circularly polarized components spectrally shifted with respect to each other by $\approx 50 \mu\text{eV}$ (inset in Fig. 1 a). In addition, a high degree ($\approx 55 \%$) of polarization, ρ_c for the PL of the neutral exciton can be evaluated, which at a first glance contradicts to the widely accepted concept of the destructive role played by the anisotropic exchange interaction on the spin preservation of the electrons and the holes prior to their recombination [2-4]. However, the anisotropic exchange interaction is “switched on” at a time instant at which both the electron and the hole have already been captured into the SQD [2-4].

It has earlier been suggested that electrons and holes are captured from the WL into the SQD on a different time scale [6], providing a time interval $\Delta\tau_{e-h}$ during which the SQD is populated with only an electron, i.e. without hole and hence no anisotropic exchange interaction should be considered within $\Delta\tau_{e-h}$. Accordingly, this spin-polarized electron can, with a certain probability, polarize the nuclei located within the volume of the SQD during $\Delta\tau_{e-h}$ via the dynamical nuclear polarization effect [7]. This results in the appearance of nuclear field \mathbf{B}_N acting upon carriers in the SQD and suppressing the anisotropic exchange interaction, when the hole is captured into the SQD. The evidence for the existence of such a field, \mathbf{B}_N could be inferred from the detected spectral shift between the σ^+ and σ^- components (see inset in Fig. 1 a). In addition, the change of the helicity of the exciting light (say, from σ^+ to σ^-) changes the direction of the electron spin into the opposite one. This should inevitably result in reversed

direction of \mathbf{B}_N , which in turn should lead to an exchange of the spectral positions of the two circularly resolved PL components. This experimental fact (inset in Fig. 1 a) totally supports the suggested model accounting for the existence of high ρ_c for the case of the neutral exciton in a SQD at $\mathbf{B}_{ext} = 0$.

According to our model, the parameter $\Delta\tau_{e-h}$ should play a crucial role on the dynamical nuclear polarization (and hence on the ρ_c possible to achieve in the experiment). For the case of SQD, the parameter $\Delta\tau_{e-h}$ could be rather large because a SQD is able to collect carriers from the total area of the laser spot (with a radii of $\approx 1 \mu\text{m}$), as has been demonstrated earlier [8]. Conversely, for the case of the MQDs ensembles, the typical size of collection area for each quantum dot within an ensemble of given density is no longer determined by the size of a laser spot, but rather by the averaged inter-dot separation distance. Consequently, to check the suggested model studies of MQDs of different densities are essential.

The inset in Fig. 1 b shows two PL spectra resolved with respect to their circular polarization helicity under excitation of σ^+ polarized light. A circular polarization degree as high as $\approx 20 \%$ is deduced from this data. Apparently, an increasing QDs density leads to a decreasing polarization degree (compare data shown in Fig. 1 a and b). A rough estimate of the QDs density at each sample spot of the QDs ensembles could be deduced from the ratio $R = I_{QD}/I_{WL}$. Data obtained from different sample spots are summarized in Fig. 2. It is important to stress that in order to obtain ρ_c at each particular sample spot, the excitation power P_{ex} was adjusted to have the same PL peak intensity as was detected for the case of SQD. It is seen that ρ_c progressively decreases with increasing R (Fig. 2), an experimental fact, which totally supports the suggested model. It should be noted however, that ρ_c does not vanish (but remains at a level of $\approx 7 \%$) despite of the essential increase of parameter R (up to 4 orders of magnitude), i.e. for conditions

at which the parameter $\Delta\tau_{e-h}$ should be negligible. This fact could be explained by a model developed by others [9], according to which it is possible to polarize the lattice nuclei by spin-flip assisted recombination of dark excitons even for the case of neutral excitons captured into the QD.

Another possible way to investigate the build-up of B_N (and hence an observed non-zero ρ_c) at the capture processes of carriers from the WL into the QDs is to study the temperature dependence. The evolution of ρ_c for the cases of SQD and MQD, respectively, as a function of the temperature is shown in Fig. 3. In the case of the SQD, ρ_c has vanished already at $T = 30$ K (Fig. 3 a) while for the MQD, the polarization decays from 20 % down to only 7 % as the temperature increases from 3.9 up to 90 K (Fig. 3 b). The reason for the decay of the PL polarization degree for both the SQD and the MQD with increasing temperature is the temperature-induced loss of spin orientation of carriers in the WL on their way towards the QDs.

This conclusion immediately stems from the measured dependence of polarization degree of the WL as a function of temperature (inset in Fig. 3 a). The experimentally observed fact that ρ_c decays much faster with the temperature for the case of SQD than for the MQDs (compare data in Fig. 3 a and b) could be explained in terms of a much shorter time for the carriers spending in the WL prior to their capture into the QDs for the case of the MQDs. Hence their spins are less affected by the destructive influence of the sample temperature. It should be emphasized that for carriers confined inside the QDs, the classical spin relaxation mechanisms are suppressed [10] relatively the relaxation in bulk materials. The latter statement finds its direct confirmation in our study: If one compares the dependences of ρ_c on the temperature recorded at $h\nu_{ex} > E_{WL}$ and at $h\nu_{ex} < E_{WL}$, respectively (Fig. 3 a), it is obvious that for the latter case ρ_c is almost temperature-independent in the temperature range of $3.9 \text{ K} < T < 20 \text{ K}$, while in case of

$h\nu_{ex} > E_{WL}$ (filled symbols in Fig. 3 a) ρ_c decreases by ≈ 10 times at the same experimental conditions.

The relatively high ρ_c ($\approx 40\%$) recorded at $h\nu_{ex} < E_{WL}$ and $T = 3.9$ K contradicts at a first glance to the model suggested: In this case photo-excited carriers do not propagate in the plane of the WL prior to capture into the SDQ and hence the model developed above predicts a negligible value on the parameter $\Delta\tau_{e-h}$ (and, accordingly ρ_c). However, it has to be realized that at excitation below the WL, the laser light is absorbed not only by the volume of the SQD but also by the band tail states (i.e. localized states) of the WL [11], followed by the tunneling of these localized carriers from these localizing potentials into the SQD. Taking into account that electrons are lighter than holes, it is reasonable to assume that electrons tunnel out of the localizing potentials and capture into the SQD prior to the holes, thus providing a non-vanishing value of the parameter $\Delta\tau_{e-h}$ even at $h\nu_{ex} < E_{WL}$.

As a result, upon excitation with $h\nu_{ex} < E_{WL}$, an increased temperature should not affect the hole capture time into the SQD (as long as $T < E_i$, where E_i is ionization energy of hole localized in band tail potential) and hence, ρ_c is predicted to remain unchanged. At the same time, at $T > E_i$ holes will become delocalized, which should result in a sharp decrease of parameter $\Delta\tau_{e-h}$ and, accordingly ρ_c . Typical values of E_i in the sample under study are on the order of 2-3 meV as deduced from our previous studies [12]. As a result, ρ_c is predicted to essentially drop with increasing temperature, when $T > 20-30$ K, in full agreement with our experimental findings (Fig. 3 a).

Finally we would like to comment on the possible reason of temperature induced loss of spin orientation of electrons while they propagate along the WL plane (inset in Fig. 3 a). Three mechanisms are known for spin relaxation of free electrons in non-magnetic zinc-blend

semiconductors [1]. They are Bir, Aronov and Pikus (BAP) [13] mechanism Elliott-Yaffet (EY) [14] mechanism and Dyakonov-Perel (DP) [15] mechanism. BAP is the mechanism of electron spin relaxation due to the exchange interaction between electron and hole spins and is important in p-type semiconductors. As our sample is non-intentionally doped, we omit BAP from further discussion. EY considers electron spin rotations only during the brief acts of electron collisions with impurities or phonons. Thus electron momentum relaxation (with the rate τ_p^{-1}) should be accompanied by a spin relaxation (with the rate τ_s^{-1}) and as a result $\tau_s^{-1} \sim \tau_p^{-1}$ is predicted according to EY. In contrast to the EY, the electron spin rotates in an effective magnetic field not during but between the collisions according to DP and hence, DP predicts $\tau_s^{-1} \sim \tau_p$.

In bulk materials, there is interplay of all these mechanisms, but in quantum wells there is limited evidence that mechanisms other than DP are important [1]. However, a temperature increase should lead to a decreasing τ_p due to both collisions of an electron with phonons and scattering at the boundaries of the InAs/GaAs interfaces. The latter is due to the temperature induced increase of thermal velocity of an electron and becomes the dominant scattering mechanism in ultra-narrow quantum wells (the WL thickness is only 0.5 nm). According to the DP mechanism, a decrease of τ_p should lead to an increasing τ_s , which evidently contradicts to the experimental data shown in the inset in Fig. 3 a. As a result, we consider an increased rate of electron scattering at the interfaces of the WL, which becomes more efficient at higher temperatures, as the most plausible explanation of sharp decrease of PL polarization degree from the WL with increasing temperature (inset in Fig. 3 a).

4. Conclusions

To conclude, we have experimentally demonstrated a progressive decrease of the circular polarization degree of the emitted InAs/GaAs QDs PL with an increasing QDs density. This is

explained in terms of the progressively decreasing $\Delta\tau_{e-h}$ parameter, which plays a crucial role for the build up of the nuclear field and hence, to monitor a vanishing circular polarization of the neutral excitons emission. Our model based on the idea of reduction of the parameter $\Delta\tau_{e-h}$ with increasing QDs density agrees nicely with the experimental results on temperature dependence of ρ_c for both SQD and MQDs. A sharp decay of the circular polarization degree of PL from the WL as a function of temperature suggests that the Elliott-Yaffet mechanism is mainly responsible for the electron spin relaxation in ultra-narrow quantum wells like the WL, rather than the Dyakonov-Perel mechanism widely accepted for quantum wells.

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References

- [1] Spin Physics in Semiconductors, 2008, M. I. Dyakonov (ed), Berlin Heidelberg Springer-Verlag.
- [2] Ware M. E., Stinaff E. A., Gammon D., Dotty M. F., Bracker A. S., Gershoni D., Korenev V. L., Bădescu Ș. C., Lyanda-Geller Y. and Reinecke T. L., 2005, Phys. Rev. Lett. **95**, 177403.
- [3] Kaji R., Adachi S., Sasakura H. and Muto S., 2008, Phys. Rev. B **77**, 115345.
- [4] Ivchenko E. L., 1995, Pure Appl. Chem. **67**, 463.
- [5] Belhadj T., Simon C.-M., Amand T, Renucci P., Chatel B., Krebs O., Lemaître A., Voisin P., Marie X. and Urbaszek B., 2009, Phys. Rev. Lett. **103**, 086601.
- [6] Moskalenko E. S., Larsson L. A. and Holtz P. O., 2009 Phys. Rev. B **80**, 193413.
- [7] Dyakonov M. I. and Perel V. I., 1984, Optical Orientation, F. Meier and B. P. Zakharchenya (ed), Amsterdam, North-Holland, Ch. 2.
- [8] Moskalenko E. S., Donchev V., Karlsson K. F., Holtz P. O., Monemar B., Schoenfeld W. V., Garcia J. M. and Petroff P. M., 2003, Phys. Rev. B **68**, 155317.
- [9] Gammon D., Efros Al. L., Kennedy T. A., Rosen M., Katzer D. S., Park D., Brown S. W., Korenev V. L. and Merkulov I. A., 2001, Phys. Rev. Lett. **86**, 5176.
- [10] Khaetskii A. V. and Nazarov Y. V., 2000, Phys. Rev. B **61**, 12639.
- [11] Toda Y., Moriwaki O., Nishioka N. and Arakawa Y., 1999, Phys. Rev. Lett. **82**, 4114.
- [12] Moskalenko E. S., Larsson M., Schoenfeld W. V., Petroff P. M. and Holtz P. O., 2006, Phys. Rev. B **73**, 155336.
- [13] Bir G. L., Aronov A. G. and Pikus G. E., 1976, Sov. Phys. JETP **42**, 705.
- [14] Elliott R. J., 1954, Phys. Rev. **96**, 266.
- [15] D'yakonov M. I., and Perel' V. I., 1971, Sov. Phys. JETP **33**, 1053.

Figure captions.

Fig. 1. μ -PL spectra of two sample spots, (a) SQD and (b) MQDs_1 measured at $T = 3.9$ K, $P_{ex} = 2$ nW, $h\nu_{ex} = 1.767$ eV and with linear polarization of the laser. The inset in a (b) shows μ -PL spectra of two sample spots SQD (MQDs_1) resolved with respect to their σ^+ and σ^- components measured at $T = 3.9$ K, $h\nu_{ex} = 1.465$ eV with σ^+ helicity of the exciting light and $P_{ex} = 1$ (0.4) μ W.

Fig. 2. ρ_c as a function of R measured at $T = 3.9$ K, $h\nu_{ex} = 1.465$ eV with σ^+ helicity of the exciting light. To record all experimental points, P_{ex} was adjusted in such a way that the peak intensity of each of the MQDs PL band had the same peak PL intensity as μ -PL spectrum of the SQD (the inset in Fig. 1 a).

Fig. 3. (a) Filled (open) symbols show ρ_c of the SQD as a function of temperature measured at $h\nu_{ex} = 1.465$ (1.433) eV with σ^+ helicity of the exciting light and $P_{ex} = 1$ (7) μ W. (b) ρ_c of the MQDs_1 as a function of the temperature measured at $h\nu_{ex} = 1.465$ eV with σ^+ helicity of the exciting light and $P_{ex} = 0.4$ μ W. The inset in (a) shows polarization degree of the WL as a function of temperature measured at $h\nu_{ex} = 1.472$ eV with σ^+ helicity of the exciting light and $P_{ex} = 0.2$ μ W.

Fig. 1

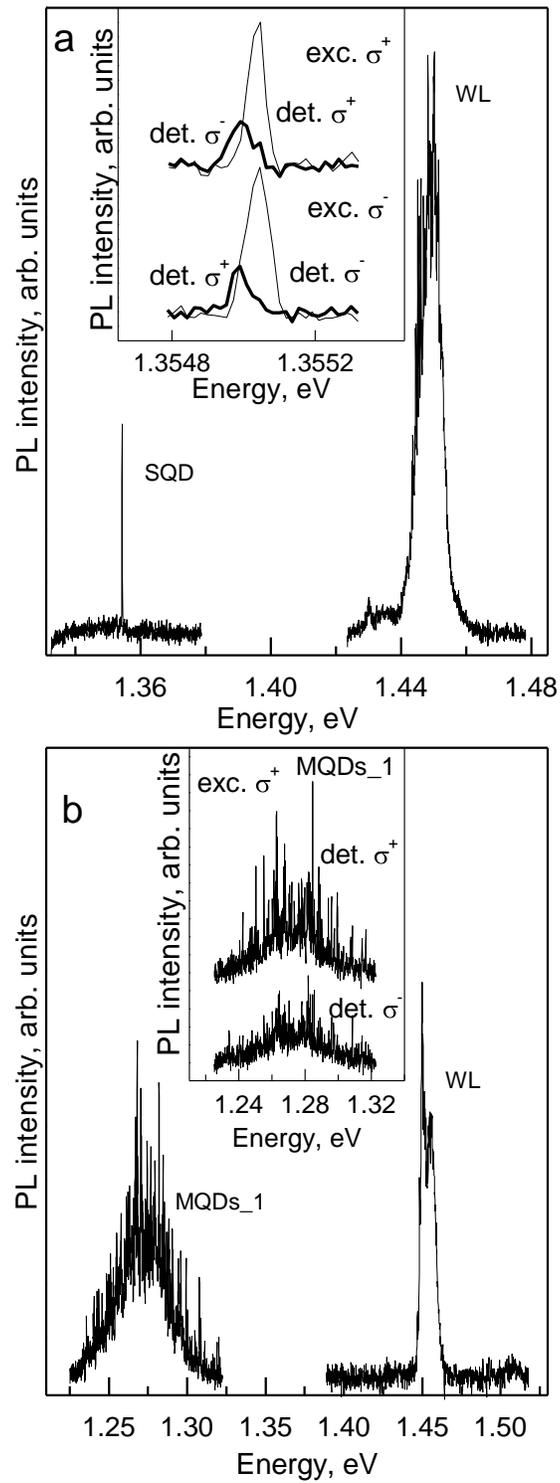


Fig. 2

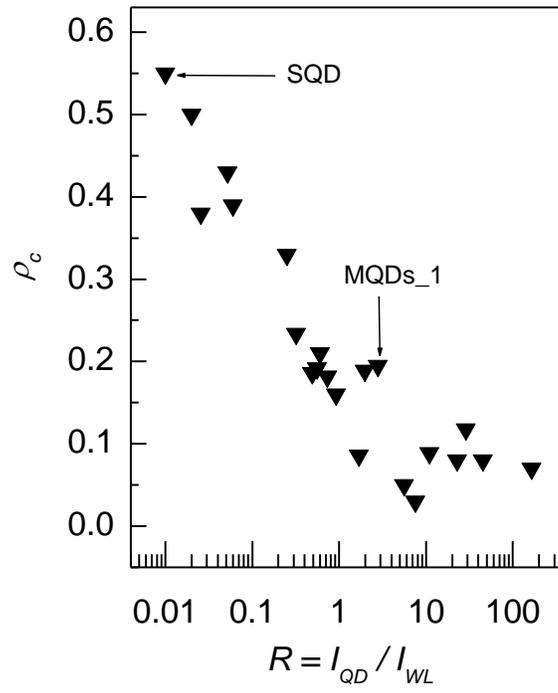


Fig. 3

