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Effects of a longitudinal magnetic field on spin injection and detection in InAs/GaAs quantum dot structures

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Abstract. Effects of a longitudinal magnetic field on optical spin injection and detection in InAs/GaAs quantum dot (QD) structures are investigated by optical orientation spectroscopy. An increase in optical and spin polarization of the QDs is observed with increasing magnetic field in the range of 0–2 T, and is attributed to suppression of exciton spin depolarization within the QDs that is promoted by hyperfine interaction and anisotropic electron-hole exchange interaction. This leads to a corresponding enhancement in spin detection efficiency of the QDs by a factor of up to 2.5. At higher magnetic fields when these spin depolarization processes are quenched, electron spin polarization in anisotropic QD structures (such as double QDs that are preferably aligned along a specific crystallographic axis) still exhibits rather strong field dependence under non-resonant excitation. In contrast, such field dependence is practically absent in more “isotropic” QD structures (e.g. single QDs). We attribute the observed effect to stronger electron spin relaxation in the spin injectors (i.e. wetting layer and GaAs barriers) of the lower-symmetry QD structures, which also explains the lower spin injection efficiency observed in these structures.

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1. Introduction

Semiconductor quantum dot (QD) structures are currently under intense investigations in view of their potential applications in spin-based quantum information technology and nanospintronic, as the three-dimensional carrier confinement in these structures promises suppression of many spin relaxation mechanisms that are predominant in semiconductors of higher dimensionality [1]. In three- and two-dimensional structures, the most important spin relaxation mechanisms such as the D’yakonov-Perel and Elliott-Yafet mechanisms are promoted by the motion of carriers. For confined carriers in QDs, the lack of carrier motion efficiently deactivates these spin relaxation mechanisms. Furthermore, their atomic-like, discrete energy level structure restricts efficiency of phonon-induced inelastic scattering processes. Hyperfine interactions of confined carriers with nuclear spins of host lattice atoms within QDs [2, 3] and carrier-carrier exchange interaction [4] become the dominant spin relaxation mechanisms at low temperatures. The expected extended carrier spin lifetimes have been confirmed experimentally [5–8], and efficient room-temperature spin detection has also recently been demonstrated [9], which is encouraging for proposals of QDs in applications of quantum computing.
[10] and spintronic devices such as spin light-emitting devices [11]. Multiple QD structures have been proposed for creating entangled photon pairs, due to their separated biexciton state [12], which may simplify separate extraction of the two entangled photons. Entangled photon pairs are essential in quantum information technology for transmission and encryption. The tunable electronic and spin interaction between carriers in interacting QDs furthermore enables the vision of quantum computation using single carrier or exciton spins in these structures.

For all these applications, a good understanding and control of relevant spin-dependent processes determining spin generation, transport and detection in these nanostructures are necessary. Though spin dynamics of carriers confined within QDs have been investigated in detail at low temperatures, studies of spin injection processes from adjacent layers into QDs remain few. Spin injection and detection processes, whether they are carried out electrically or optically, are essential to future spintronic devices based on QD structures because they provide an effective means to initialize and readout spin information.

In this work, we investigate efficiency of optical spin injection into InAs/GaAs QDs from a surrounding thin wetting layer (WL) and a GaAs barrier by optical orientation and magneto-optical spectroscopy. Based on effects of an applied longitudinal magnetic field, we aim to obtain in-depth information on the dominant physical processes that govern efficiency of spin injection and detection. Effects of geometrical structure of the QDs will be closely examined by studies of different sample structures featuring single and lateral double QD patterns, which have not been investigated in earlier studies.

2. Samples and Experimental Details

A set of InAs based QD samples was studied. They were grown by gas source molecular beam epitaxy (MBE) on a GaAs substrate under different conditions to yield structures of isolated single QDs (SQD) or structures composed of two close-by QDs (denoted by DQD1 and DQD2). In the DQD structures, all QD pairs are aligned along the [1 1 0] crystallographic direction. Following a 300 nm GaAs buffer layer that was grown at 580 °C, 1.8 monolayers (ML) of InAs were deposited at 500 °C for the SQD sample, resulting in randomly positioned single QDs with a density of ~6×10^9 cm^-2 on a thin WL. A typical dot height is around 12 nm with a dot diameter of 40-60 nm. For the DQD1 sample the aforementioned growth procedure continued by depositing 6 ML GaAs and a further 0.6 ML InAs at 470 °C. The thin GaAs capping process led to development of uniaxial strain, resulting in growth of double dots aligned along the [1 1 0] crystallographic direction upon InAs regrowth. The total QD density was increased to around 1.1×10^11 cm^-2 and the QD size was reduced to around 4 nm in height and roughly half the diameter of the original SQDs. For the DQD2 sample, the InAs deposition rate during the regrowth was increased by five times, leading to pairs of QDs that are slightly larger in diameter (around 30 nm) but lower in height (around 3 nm). For each sample, the growth was completed by a 150 nm GaAs cap layer and the repetition of the respective QD structure growth to yield a similar QD layer exposed on the surface for imaging by atomic force microscopy (AFM). AFM images of the structures and more details on the growth procedure can be found in references [13, 14] and [15].

Optical orientation measurements were conducted using a continuous-wave (cw) tunable Ti:Sapphire laser as an excitation source. Its polarization state was controlled by a broad-band quarter-wave-plate, rotatable with respect to the axis of a linear polarizer, to achieve both circular polarization orientations σ⁺ and σ⁻ as well as linear polarization σ±. In some cases, the broad-band quarter-wave-plate was replaced by a photoelastic modulator (PEM) to provide excitation with alternating σ⁺ and σ⁻ polarization at a frequency of 50 kHz. The samples were kept at a temperature of 10 K in a cryostat equipped with a split-coil superconducting magnet (up to 10 T). Excitation and detection were carried out in a back-scattering geometry in the Faraday configuration, i.e. with both excitation beam and an applied magnetic field being oriented along the direction normal to the sample surface. A PEM, in conjunction with an appropriately oriented linear polarizer, was employed to analyze photoluminescence (PL) circular polarization. A grating monochromator connected to a liquid-nitrogen cooled Ge-detector was used for spectrally resolved detection of the PL signal. PL circular polarization degree \( P_\alpha = (I_\alpha^{\sigma^+} - I_\alpha^{\sigma^-})/(I_\alpha^{\sigma^+} + I_\alpha^{\sigma^-}) \) under a specific excitation polarization \( \alpha \) was
calculated by dividing the detector signal at the PEM-frequency (proportional to $I_{a}^{\sigma+} - I_{a}^{\sigma-}$) by the simultaneously measured total PL signal using an appropriate calibration constant for the entire detection system.

3. Results

3.1. PL, PLE and PL polarization at zero magnetic field

Representative PL and PLE spectra from the studied samples in the absence of an external magnetic field are shown in figure 1, taking as examples SQD and DQD1. The PL spectra, shown in the lower part of the right panels in figure 1(a)-(b), are dominated by a PL band at the lower energy that originates from the QD ground states. An additional PL band (shoulder) can also be observed on the higher energy side of the PL spectrum for the SQD (DQD1) sample, which is assigned to emissions from the first excited state by state-filling spectroscopy [16]. Though the displayed spectra were taken under photoexcitation above the bandgap energy of the GaAs barrier, similar spectra were obtained under photoexcitation with photon energy below the GaAs bandgap energy except of a large difference in excitation efficiency. The PL excitation (PLE) spectra, shown in the lower part of the left panels in figure 1(a)-(b), were obtained by detecting at the peak position of the QD ground state PL. They show generation of non-equilibrium carriers in the QDs via optical absorption in three distinctly different regions of the structures: (1) the GaAs barrier at the highest energies (above the GaAs free exciton XG); (2) the WL at the intermediate energies between XG and the WL heavy-hole (hh) free exciton XH, and (3) quasi-resonant excitation within the QDs at the lowest energies below XH. A step-like increase in PL intensity can clearly be seen whenever the excitation photon energy crosses the bandgap energies of the WL and GaAs, which corresponds to an increasing density of states available for optical absorption. For the WL, two steps were observed in the PLE spectra. One corresponds to the band-to-band (BB) transitions from the hh valence band (VB) to the conduction band (CB), denoted as hh-e, and was observed over the spectral range between XH and XL [WL light-hole (lh) free exciton]. Above the XL energy, an additional BB transition from the lh VB to the CB (denoted as lh-e) provides another step in the PLE spectra. In addition, free excitons arising from GaAs (XG) and the WL (XL and XH) can also clearly be seen as strong PLE peaks due to their further increased densities of states as compared to that of free carriers in the respective bands.

In the upper parts of figure 1(a)-(b), we show PL circular polarization degrees of the QDs as a function of detection (the right panels) and excitation (the left panels) photon energy under circularly and linearly polarized photoexcitation. Under circularly polarized excitation, the QD PL exhibits circular polarization that is co-polarized with the excitation light. The exact degree of the polarization critically depends on excitation photon energy and can reach up to 10% under excitation only within the hh-e part of the WL and in GaAs. When both lh-e and hh-e transitions of the WL simultaneously occur (i.e. over the range between XL and XG), however, the observed PL polarization degree is strongly reduced. In contrast to the case under circularly polarized photoexcitation, no PL circular polarization can be observed under linearly polarized excitation regardless of excitation energy as shown in figure 1.

3.2. PL, PLE and PL polarization in a longitudinal magnetic field

Representative PL and PLE spectra from the studied QD structures in an external longitudinal magnetic field are shown in figure 2, taken as an example the SQD sample. Though the PL spectrum practically remains the same in magnetic fields, a new series of peaks appears in the PLE spectrum. The energy positions of these peaks vary with field strength, and are displayed in figure 3 within the spectral range of the BB transitions in the WL. The new series of peaks exhibits a large blue shift that is linear with the field, and is attributed to the Landau levels (LLs) of the hh-e BB transition in the WL. The notation "n-n" denotes the optical excitation from the nth LL of the hh VB band to the nth LL of the CB. In comparison, XH and XL only show a small and quadratic shift towards higher energies with increasing field, confirming their excitonic character.[17] The energy distance between the merging of the LLs at zero field and the exciton line XH yields an exciton binding energy of 3.5 meV.

From the LL transitions, we deduce an in-plane reduced effective mass of the electron-hole pair involving the hh VB of the WL to be 0.059 $m_0$, 0.052 $m_0$ and 0.058 $m_0$ for SQD, DQD1 and DQD2, respectively. These values are comparable with the reported values in strained InGaAs/GaAs quantum
wells [18, 19]. Due to strong overlap between the hh-e and lh-e BB transitions and their lower oscillator strength, no lh-e related LLs could be resolved at excitation photon energy above XL.

Despite that no significant change was observed in the PL spectrum from the QD ground state, an increase in its circular polarization degree can be observed in applied magnetic fields, e.g. from a maximum degree of 10% at 0 T [figure 1(a)] to 15% at 5 T (figure 2) judging from the excitation energy dependence of the QD PL polarization. For the PL polarization of the QD excited state, in addition to an increase in the maximum value, a derivative lineshape was observed. This can be explained by a larger Zeeman splitting expected for the two p-shell states [20]. A variation of the QD PL polarization can also be seen whenever the excitation photon energy crosses an LL transition, see the insert of figure 2 for a close-up of the 1-1 hh-e transition in the WL. It finds a natural explanation in the Zeeman splitting of the LL spin states. It is well known that, in addition to confining the carriers into LLs, the magnetic field also splits spin states of the electrons and holes participating in the LL transitions of the WL by a Zeeman energy determined by their total effective g-factor $g_{e-h}^{WL}$. Though such spin splitting is too small to be resolved in the PLE spectra by monitoring the PL intensity, the change in the photogenerated spin population of carriers when the excitation photon energy is in resonance with one of the spin levels is sufficient to lead to an observable corresponding change in spin polarization of carriers injected into the QDs. This gives rise to a variation of the QD PL polarization degree while sweeping the excitation photon energy across the LLs at a fixed magnetic field, as illustrated in figure 2.

To further investigate effects of magnetic field on spin injection and detection, intensity and polarization of the QD ground state have closely been examined in a sweeping magnetic field while keeping photoexcitation at a fixed energy within the range of the hh-e transition in the WL. As expected from the fan diagram shown in figure 3, such optical excitation can be brought into resonance in energy with WL LLs one by one in the order of decreasing index numbers with increasing magnetic field, which is manifested by the appearance of a series of PL intensity peaks shown by the lowest curve in figure 4. Again, the spin splitting at each LL gives rise to a corresponding variation in the PL polarization. In addition to the spin splitting, two more features unrelated to the LLs can be observed from the QD PL polarization as a function of external longitudinal magnetic field (see figure 4). The first feature is that the PL polarization degree under circularly polarized excitation increases rather drastically as soon as the magnetic field moves away from zero regardless of the field direction, giving rise to a dip around $B = 0 \, \text{T}$ in a field scan of the PL polarization $P(B)$. This effect is found to be common for all of the studied QD structures, see figure 5, consistent with the results of the field-induced increase in the QD PL polarization shown in figures 1 and 2 and discussed above. Such a polarization increase with increasing magnetic field is, however, absent under linearly polarized excitation as can be seen in figure 5 where a slight step-like component of the PL polarization around $B = 0 \, \text{T}$ is observed. The second extra feature in figure 4 and figure 5 is a slope in the QD PL polarization as a function of magnetic field, i.e. a nearly linear dependence on field, which can be observed under both circularly and linearly polarized excitation. This feature is found to be more pronounced in the DQD samples as compared with the SQD sample.

4. Discussion

4.1. Origin of the QD PL and PL polarization

The most common origins of PL emissions from semiconductor QDs have in the past been shown to be recombination of neutral and charged excitons,[1] distinguished by the number and nature of the charge carriers constituting the excitons and their corresponding optical polarization. As displayed in figures 1 and 2, our studies show that the ground state PL of all QD structures studied in this work exhibits co-circular polarization with the excitation light. This rules out the possibility that recombination of negatively charged excitons ($X^-$) is responsible for the observed PL. This is because earlier studies [21] have shown that $X^-$ should give rise to PL with counter-circular polarization, i.e. opposite to the helicity of the excitation light, under non-resonant excitation conditions (e.g. excitation above the bandgap energy of WL or GaAs barrier). In case of neutral excitons ($X^0$) in QDs, it has been shown both experimentally [5, 22] and theoretically [23] that the eigenstates of the optically active bright exciton states in zero magnetic field generally are $|X\rangle$ and $|Y\rangle$, split under influence of an
anisotropic electron-hole exchange interaction (AEI) due to anisotropy of the QD originating from e.g. in-plane QD elongation and/or interface anisotropy. Here, $|X\rangle = (|+1\rangle + |-1\rangle)/\sqrt{2}$ and $|Y\rangle = (|+1\rangle - |-1\rangle)/i\sqrt{2}$ where $|+1\rangle$ and $|-1\rangle$ are the $\sigma^+$ and $\sigma^-$ active states with pure spin orientations. Consequently, recombination of neutral excitons in the $|X\rangle$ and $|Y\rangle$ states should give rise to linearly polarized emissions that are orthogonal to each other. In principle, no circularly polarized emission should be observed when no external magnetic field is applied. However, it was recently shown that this restriction can be relaxed and PL from $X^0$ can become partially circularly polarized [24, 25] when an effective magnetic field (i.e. Overhauser field) is created via dynamic polarization of nuclear spins of lattice atoms within QDs under cw excitation by circularly polarized light. Such a collective nuclear field counteracts the AEI effect and can restore the $|+1\rangle$ and $|-1\rangle$ character of $X^0$. For singly positive charged excitons ($X^+$) in QDs, on the other hand, the exchange interactions cancel out in the ground state due to pair-off of total angular momentum of the two holes [26]. Consequently, PL from $X^+$ should be circularly polarized with the same helicity as the circularly polarized excitation light [27]. Thus we conclude that in our QD structures both $X^0$ and $X^+$ can contribute to the observed PL at low temperatures. From the dependence of PL polarization on applied magnetic field, $X^+$ is believed to be the dominant contribution (to be discussed in detail below). The presence of positively charged dots in our structures is facilitated by background p-type doping due to carbon contamination occurring during MBE growth, which has been confirmed by low-temperature PL results showing a free-to-bound transition involving a shallow acceptor in the GaAs barrier.

The broad spectral width of the QD PL ground state, as seen in figures 1-2, is a result of inhomogeneous broadening due to a large variation in the size of the monitored QDs and also in the strain field they experience. The polarization degree of the QD PL is found to increase with increasing emission energy, which can be attributed to the following effects. Firstly, the exciton lifetime of the higher-energy QDs is commonly observed to be shorter due to energy transfer to the lower energy QDs that reduces the extent of spin relaxation [28]. Secondly, a contribution of unpolarized exciton species like $X^0$ and biexcitons $XX$ can vary among the QDs with different energies. For example, a stronger contribution of $XX$ due to filling of extra carriers is expected for the lower energy QDs with a longer exciton lifetime as compared with the higher energy QDs.

The fact that the QD PL circular polarization follows the excitation polarization state and also depends on the excitation energy, as shown in figure 1, suggests that the observed polarization is due to spin injection when the excitation photon energy is above the XH, i.e. due to generation and transfer of spin-oriented carriers from the WL and GaAs into the QDs where they subsequently recombine. For excitation involving only the hh-e BB transition of the WL, optical selection rules [1] predict complete spin polarization of the excited carriers. Hole spin relaxation in two-dimensional structures is known to be relatively fast as compared with electron spin relaxation. Hence, we will assume in the following discussion that the spin information is carried into the QDs by the injected electrons. Upon recombination of injected electrons and holes in the forms of $X^0$ and $X^+$ in the QDs, light of the same polarization as the excitation light is emitted due to the validity of the same optical selection rules as in the WL. For BB excitation involving only the lh-e transition of the WL, on the other hand, the spin orientation of the photo-generated electrons is opposite to the one involving the hh-e transition under the same excitation polarization. Based on the 1:3 ratio in the density of states between the lh and hh states, one would thus expected 50% reduction in electron spin polarization and thus PL polarization when both hh-e and lh-e BB transitions are involved under photexcitation above XL. Our experimental finding of nearly vanishing PL circular polarization degree[13] under such excitation, shown in figure 1, reveals a strongly decreased spin polarization degree of the total electron population in the QDs. This could indicate different spin injection efficiency between the optical spin orientation conditions involving the hh-e and lh-e BB transitions in the WL, of which the exact origin is still not fully understood and is beyond the scope of the present work.

In sharp contrast to the optical spin orientation involving the lh-e transition of the WL, the electron spin and PL polarization of the QDs upon optical spin injection from the GaAs barrier is significantly higher than 50% of that due to the spin injection only under the hh-e excitation from the WL. This is despite the fact that both hh-e and lh-e BB transitions are involved here due to the degeneracy of the hh-lh states in the three-dimensional GaAs barrier. For the DQD1 sample, shown in
figure 1(b), the polarization under excitation in the WL can even be lower than that under excitation above GaAs. As the same spin detector (i.e. the QDs) was employed under both excitation conditions in each sample, the observed lower-than-expected degrees of the QD PL and electron spin polarization under the WL hh-e excitation is likely a consequence of lower spin injection efficiency due to e.g. relatively stronger electron spin relaxation in the WL and/or slower spin injection from the WL into the QDs. This suggestion is consistent with the results in the magnetic field, to be discussed below.

4.2. Effects of a longitudinal magnetic field on spin injection and spin detection

The effects of a longitudinal magnetic field on PL polarization of the studied QDs can be characterized by three main features, namely, (1) a dip in PL polarization degree $P(B)$ around zero field under circularly polarized optical excitation; (2) a slope in $P(B)$ that can be observed under both circular and linear excitation; and (3) variations in PL polarization degree in the vicinity of the LLs. The third feature can be well accounted for by spin splitting of each LL, which gives rise to a difference in the spin-dependent density of states and a corresponding change in the generation of electrons with different spin orientations. Below, we shall focus our discussion on the physical processes responsible for the first two features and their impact on spin injection and spin detection in the QD structures.

4.2.1. Suppression of spin depolarization and enhancement of spin detection efficiency of the QDs.

The first feature, i.e., a dip in PL polarization degree around zero field under circularly polarized optical excitation, can be observed in all of the QD structures as shown in figure 5. To examine if the feature arises from physical processes occurring within the QDs or from the surrounding layers, we have performed similar experiments under optical excitation above the GaAs bandgap as well as under quasi-resonant excitation of the QDs. In the former case, photoexcitation can create carriers in all regions of the structures including GaAs, WL and QDs, whereas in the latter case carriers are created only within the QDs which excludes contributions from GaAs and WL. A representative $P(B)$ is shown in Figure 5(a) for the SQD sample. The fact that the dip can be observed even under excitation of the QDs alone provides compelling evidence that it must originate from physical processes inside the QDs.

To understand the physical mechanism responsible for the observed dip, i.e. suppression of optical and electron spin polarization close to zero magnetic field, we have closely examined the field dependence of the PL polarization of the QD ground state under cw (i.e. fixed polarization) and also under alternating circularly polarized excitation by employing a PEM. Typical results are shown in figure 6(a), taking the SQD sample as an example. We find an enhancement of the zero-field polarization dip under the PEM excitation (the open circles) that is confined to the low-field region, whereas at higher fields the curve is identical to that obtained under cw excitation. From this we conclude that the experimental $P(B)$ curves should consist of two components, i.e. one narrow and one broad dip. Whereas the narrow dip can be affected by the excitation conditions of fixed or alternating circular polarization, the broad dip remains unaffected.

Under cw excitation with a fixed helicity of circularly polarized light, the narrow dip becomes shallower and at the same time shifts away from zero field. This is characteristic for the effect of dynamic nuclear polarization (DNP) under optical spin orientation, which can align nuclear spins of the host atoms in the QDs via hyperfine interaction with spin-polarized electrons in the QDs. The alignment of nuclear spins introduces an Overhauser field acting in turn to stabilize the electron spins by suppressing spin dephasing and spin relaxation under the influence of random and time-varying fluctuations of nuclear spins via hyperfine interaction. This leads to the observed increase of the PL polarization degree at zero magnetic field, and can at the same time give rise to the observed shift of the narrow dip from zero field to a field that cancels out the Overhauser field [see figure 6(a)].

To confirm the effect of DNP, we carried out optical orientation experiments under alternating circularly polarized excitation at a speed exceeding the response time of the DNP (on the order of ms) [29] such that buildup of DNP is prevented. Indeed, by switching the excitation circular polarization (and thus the excited electron spin polarization) with a frequency of 50 kHz (using a PEM), a deeper polarization dip (i.e. a lower electron spin polarization degree) can be observed as shown by the open circles in figure 6(a). This conclusion is further supported by the observed increase of the PL polarization with increasing excitation density under cw optical orientation that facilitates DNP, see
regions I-II in figure 7. Such increase was absent under PEM excitation when DNP is not expected to occur. It should be noted that the observed decrease of polarization degree at even higher excitation densities shown in region III of figure 7 is due to double occupancy of the QD ground state [30], leading to pair off of electron spins.

The aforementioned hyperfine-interaction induced dip is found to be similar in width for all investigated samples, in the range of 60-80 mT, indicating similar fluctuations of the effective hyperfine field. On the other hand, the extent of the shift of the dip from zero field under cw circularly polarized excitation was strongly reduced (down to a few mT) in the DQD samples as compared to 20 mT in the SQD sample. The observed weaker DNP effect in the DQDs could be attributed to weaker electron spin polarization (due to lower spin injection efficiency as will be discussed below) and a relatively shorter time when a QD is occupied by a spin-polarized electrons with the same excitation density (due to a larger QD density). For magnetic field strengths exceeding the dispersion of the nuclear hyperfine field (typically of the order of a few tens of mT) [3], i.e. beyond the range of the narrow dip, the electron spin dephasing due to the hyperfine interaction is quenched [27].

The second component in the field scan of the QD PL polarization, i.e. the broader dip in figure 6, represents a spin dephasing/depolarization mechanism with a larger strength. It thus requires a stronger external field to suppress. AEI has been discussed in the literature to be among the dominant mechanisms for electron spin dephasing and relaxation of neutral excitons in QDs, by providing an effective transverse field and also a randomly fluctuating field. An external longitudinal magnetic field acts to compete with the AEI effect and circularly polarized neutral exciton eigenstates can be recovered as we suppress the AEI [26, 31] Following Dzhioev et al. [32, 33] we can model the evolution of the PL circular polarization degree in the field observed from neutral excitons under circularly polarized excitation by

$$P_c(B_z) = P^0_c \frac{\Omega^2_0}{\Omega^2_1 + \omega^2}.$$  \hspace{1cm} (1)

Here $B_z$ is the strength of the magnetic field along the z-axis (growth axis), $P^0_c$ is a fitting parameter, corresponding to the initial circular polarization degree, without dephasing by the AEI. $\hbar\Omega_1 = g^{\text{QD}}_X \mu_B B_z$ is the exciton Zeeman energy (with $g^{\text{QD}}_X$ being the QD exciton effective g-factor and $\mu_B$ the Bohr magneton) and $\hbar\omega$ the zero field splitting of the QD exciton energy level introduced by the AEI. Using an approximate excitonic g-factor of 1 [34], we obtain AEI splittings in the range of 30-40 µeV by fitting equation (1) to our experimental results from the studied QD samples, which is in good agreement with the AEI values reported in the literature [26, 35, 36]. The fitting curves for the SQD and DQD1 are displayed as the solid lines in figure 6(a)-(b), which satisfactorily describe the broader dip. In principle, DNP can also cause circular polarization of PL from QD neutral excitons at zero field by introducing an effective magnetic field induced by polarized nuclear spins that counteracts AEI. This effect, if sizable, should be accompanied by a shift of the AEI induced polarization dip from zero field. As this shift is not detectable in all of our samples from our fitting of equation (1), likely due to the overall low degree of electron spin polarization and the resulting weaker DNP, we suggest that AEI has lead to a vanishing degree of PL circular polarization and also of electron spin polarization at zero field for neutral excitons. Under such assumption, we can conclude from the contribution of the AEI-induced dip to the total experimental curves that in all of the investigated samples the majority (54%-73%) of the observed PL originates from charged excitons X+.

AEI can also explain the slight step-like component, or S-shape dependence, of the circular polarization curves under linearly polarized excitation, which is noticeable in figure 5 and is also shown as a close-up in figure 6 (b). Such S-shape dependence originates from linear-to-circular polarization conversion due to the AEI and can be modeled by [32, 33]

$$P_c(B_z) = P^0_c \frac{\alpha \Omega^2}{\Omega^2_1 + \omega^2}.$$  \hspace{1cm} (2)
where $P^0_1$ is a fitting parameter, corresponding to the initial linear polarization degree. We find that with the same AEI value excellent agreement can be obtained between the simulated and experimental curves under both circularly and linearly polarized excitation, as shown in figure 6(b). This further confirms the assignment of the broad dip to the AEI-induced spin dephasing.

To summarize, the observed increase of the QD PL circular polarization in an external longitudinal magnetic field under circularly polarized excitation can be explained by field-induced suppression of electron spin dephasing within the QDs. The two dominant physical mechanisms leading to the electron spin dephasing in the studied QDs are attributed to be hyperfine-interaction and AEI. By suppressing these spin dephasing processes, the PL and electron spin polarization can be increased by a factor of up to 2.5 with a given degree of spin polarization of the injected electrons. In other words, the spin detection efficiency of the QDs can be improved by the same factor.

It should be pointed out that, even when the spin dephasing described above becomes nearly completely inactive at a high magnetic field, the maximum PL and electron spin polarization of the QDs still remains around or below 20% for all of the studied QD structures. The main reasons for this limited polarization can be manifold. Firstly, other than the spin dephasing due to the Larmor precession described above, there are spin relaxation processes in the QDs promoted by time-varying perturbations that can lead to spin flips. These processes can in principle be suppressed as well in applied magnetic fields. However, they may follow a field dependence that is different from what is described by equations (1) and (2), such that they are not completely quenched within the field range applied here. Consequently, the spin detection efficiency of the QDs is limited by a factor of $1/(1+\tau/QD/\tau/QD)$ where $\tau/QD$ and $\tau/QD$ are the electron lifetime and spin relaxation time of the QD excitons employed for spin detection. The second reason limiting the optical/spin polarization degree of the QDs can be a reduced spin generation efficiency under optical orientation due to mixing of states. For optical excitation above the WL and GaAs, electron-hole pairs were created at a high momentum. Together with a possible reduced local symmetry due to e.g. in-plane strain anisotropy, mixing of states (spins) can occur that leads to optical generation of both spin orientations. Even under the resonant excitation of the QDs, carriers/excitons were generated at energies much higher than the ground state, resulting in imperfect spin generation due to state-mixing. The third reason for the observed limited QD PL polarization is low spin injection efficiency, due to severe spin relaxation in the spin injectors (i.e. WL and GaAs) and spin loss during carrier transfer from the spin injector to the QDs. The latter involves carrier transport across the interface between the QDs and the spin injector, as well as energy and momentum relaxation of the injected hot carriers within the QDs down to the ground state that was monitored in the experiments. Spin relaxation accompanying energy and momentum relaxation of carriers has in earlier studies been shown to be important in two- and three-dimensional semiconductor structures, which accelerates with increasing excess energy [37, 38]. In the present study, no such correlation between the extent of spin relaxation and excess energy was found. In fact, the SQD structure with the highest excess energy (360 meV), measured between the QD ground state and the WL XH level, shows higher PL (and electron spin) polarization than the DQD structures with an excess energy of 180 meV (DQD1) and 150 meV (DQD2), respectively. Therefore, we believe that spin relaxation accompanying energy and momentum relaxation of injected carriers within the QDs is not the dominant mechanism responsible for the low spin injection efficiency. This is not surprising keeping in mind that such process is expected to be significantly reduced in the QDs due to a lack of carrier motion. This leaves spin relaxation within the spin injector and spin scattering across the interface as the most likely mechanisms leading to the observed low spin injection efficiency. Furthermore, there could be a contribution from QDs occupied by biexcitons XX. Biexciton recombination results in counter-polarized luminescence, thus reducing the ensemble PL polarization degree. Below we will provide experimental evidence that demonstrates the importance of spin relaxation in the spin injector on the QD PL and spin polarization.

4.2.2. Effect of electron spin relaxation in WL and GaAs barrier on spin injection into QDs.

Another interesting effect of the longitudinal magnetic field on the PL polarization of the QDs is the linear dependence, manifested by a slope in the experimental curves $P(B)$ shown in figures 4 to 6. Unlike the case of the polarization dip around zero field discussed above, this effect persists even
under linearly polarization excitation. A slope identical to that under circularly polarized excitation can be observed, except that it is vertically shifted such that it crosses zero polarization at zero field. In other words, PL (and electron spin) polarization develops in field and switches sign following the direction of the field. This observation cannot be explained by the linear-to-circular polarization conversion because it should have otherwise yielded an S-shape field dependence, returning to zero polarization at high fields, as described above by Eq. (2). As no spin polarization is expected to be generated by the linearly polarized light, the most plausible explanation is that the observed field-induced electron spin imbalance must be caused by spin relaxation from the energetically higher-lying to the lower-lying spin states that have undergone a Zeeman splitting in the magnetic field. This imbalance increases with increasing field strength due to increasing Zeeman splitting, and switches sign when the ordering of the spin states is reversed by reversing the direction of the field. To determine if the effect is only associated with the QDs or it originates from the surrounding layers (WL and GaAs), we have compared the results obtained under optical excitation above the WL or GaAs bandgap energy with that recorded under the quasi-resonant excitation of the QDs. Strikingly, the slope is reduced significantly whenever the excitation photon energy is tuned below XH, as demonstrated in figure 8. As electrons are only photogenerated in the QDs under such excitation condition, the observed reduced slope provides evidence for much weaker spin relaxation in the QDs. We can thus conclude that strong spin relaxation in the WL and GaAs barrier is the main source of the observed slope in the P(B) curves under the excitation above the WL and GaAs bandgap energies. This conclusion is reasonable considering that the spin relaxation time in QDs is typically significantly longer [39] than that in higher-dimensional layers such as the WL and GaAs barrier in our QD structures.

Another striking observation is the missing slope in the SQD sample, see figure 8(a), which is markedly different from the two DQD samples where a sizable slope was observed [figure 8(b)-(c)]. This finding points to stronger spin relaxation in the DQD samples. From figure 8 a clear difference in the slope can also be seen between GaAs and WL excitation in the DQD samples, with the latter showing a steeper slope (roughly doubled). This reveals a stronger spin imbalance in the WL than in GaAs, which could be caused either by a shorter spin relaxation time with respect to spin injection time or by a larger Zeeman splitting of electrons or both. Judging from the reduced spin injection efficiency observed at zero field (thus no Zeeman splitting) from the WL as compared with the GaAs barrier discussed above, we believe that spin relaxation is at least an important factor here. This effect also contributes to the observation of a sizable shift of all PLE polarizations towards positive polarization under excitation in the WL and in a negative magnetic field, shown in figure 9(b). For excitation in the GaAs barrier (at higher energies than XG) there is only a slight shift in the PLE polarization, which is consistent with its reduced slope in P(B) shown in figure 8(c). We note, however, that the shift of the polarization curves under excitation above the GaAs bandgap shown in figure 9(b) is slightly underestimated, considering that the effective excitation density is higher which leads to a reduction in the degree of the slope in P(B). For quasi-resonant excitation directly into the QDs (at lower energies than XH) the PLE polarization under linear excitation (and thus the total polarization shift) is negligible, in good agreement with the vanishing slope in P(B) shown in figure 8(c). This finding further confirms that the observed slope and the associated spin relaxation should mainly occur outside of the QDs. Consistent with the missing slope in the SQD sample shown in figure 8(a), no field-induced shift of the PLE polarization was observed in this sample over the entire spectral range of the excitation light as displayed in figure 9(a). It is interesting to note that the polarization shift observed in the DQD2 sample is very similar between the hh-e and lh-e excitation of the WL with linearly polarized light, see figure 9(b). Since different holes are involved between these two excitation conditions, the Zeeman splittings of holes and also excitons are expected to be markedly different leading to different spin imbalance. The observed similar shift of polarization can thus be explained by assuming that it is governed by spin relaxation between the Zeeman-split spin states of the CB electrons, which are involved in both hh-e and lh-e excitation.

To further illustrate the effect of spin relaxation of the barriers or spin injectors (i.e. WL and GaAs) on spin injection and the resulting QD PL polarization as a function of external longitudinal magnetic field, we employ a set of coupled rate equations modeling both spin relaxation and spin injection processes under optical excitation within the WL and GaAs barrier [figure 10(a)]:
the Zeeman splitting of $n_{\uparrow}^b$ expected since no spin imbalance is created ($\Delta E_c^b = 0$) and are the spin relaxation time and $\tau_c$ of the spin relaxation processes, respectively. For faster spin relaxation in the barrier, e.g. $\tau_c = \tau_{\uparrow}^s \approx \tau_{\downarrow}^s$, the QD PL polarizations under $\sigma^+$ ($G_{\downarrow} > G_{\uparrow}$) and $\sigma^-$ ($G_{\uparrow} > G_{\downarrow}$) excitation show a constant polarization degree that is independent of magnetic field, see the dotted lines in figure 10(b). The sign of the QD PL polarization follows that of the excitation light. The exact degree of the PL polarization critically depends on the efficiency of optical orientation (and spin loss) and importance of spin relaxation in the barrier, which are reflected by the ratios of $G_{\downarrow} / G_{\uparrow}$ and $\tau_{\uparrow}^s / \tau_c$, respectively. Here we have neglected spin relaxation within the QDs, for simplicity, as it is expected to be less important than that in the WL and GaAs.

Linearly polarized excitation results in close to zero QD polarization at all fields, as expected since no spin imbalance is created ($G_{\downarrow} = G_{\uparrow}$) and spin relaxation between the Zeeman split spin levels can be neglected in this case. For faster spin relaxation in the barrier, e.g. $\tau_c = \tau_{\downarrow}^b$, however, QD PL polarization is predicted to vary with magnetic field strength regardless of excitation polarization as a result of energetically favorable spin relaxation towards the lower-lying spin level, see the thick solid lines in figure 10(b). The negative sign of the slope is originated from a negative electron g factor used in the simulations, in light of the same sign known for GaAs and InAs.
In addition to the appearance of the slope, we also note that the relative magnitude of polarization, measured as the difference between the polarization degrees under circularly and linearly polarized excitation, reduces as the spin relaxation time in the barrier shortens. This leads to a weaker PL polarization at zero field under circularly polarized excitation, as compared with the case of slower spin relaxation in the barrier [see the dotted lines in figure 10 (b)]. These predictions are in excellent agreement with our experimental finding shown in figure 5, where the extent of the field variation and the absolute degree of the PL polarization are seen to be anti-correlated.

From the simulations, the observed difference between the studied QD samples is attributed to their different ratios of $\tau_s^b / \tau_c$, i.e. $>100$ for the SQD sample, 10 for the DQD1 and 40 for the DQD2 sample. We should point out that these values only serve to provide a qualitative comparison between the samples. There could be a large uncertainty involved in their absolute values due to unknown values of several parameters for the studied structures. The general trend illustrated from our simulations should hold, however, which clearly show that the $\tau_s^b / \tau_c$ ratio is significantly lower in the DQD sample structures than for the SQD sample. This demonstrates drastically increased importance of spin relaxation in the WL and GaAs barrier of the DQD structures. In principle, the lower ratio of $\tau_s^b / \tau_c$ could be caused by either a shortening of spin relaxation time $\tau_s^b$ or a prolonged carrier capture time $\tau_c$ into the QDs from the barriers. A possible factor leading to the former could be the presence of stronger structural and strain anisotropy in the WL of the DQD structures than in the "isotropic" WL of the SQD, i.e. the very factor driving the preferred alignment of the two QDs along $[\overline{1} \overline{1} 0]$. This strain anisotropy could also possibly extend into the immediate surrounding GaAs layers and cause a similar effect there, as the most efficient carrier/spin injection from the GaAs barrier into the QDs mainly occurs within the region closest to the QDs. Structural asymmetry is known to mix spin states and to promote spin relaxation via the D'yakonov-Perel mechanism, thus could enhance spin loss in the DQD structures. We should point out that this anisotropy in the WL and/or GaAs should be distinguished from the one acting inside the QD, causing the AEI, as we do not see any correlation in the strength of the two.

A prolonged carrier capture time $\tau_c$ into the QDs from the barriers, on the other hand, could possibly be a result of trapping due to large potential fluctuations in the WL or near the GaAs-QD interface that is more severe under the specific growth conditions for the DQD structure. If such an extended carrier capture time becomes comparable to the radiative decay time of free excitons, one would expect an increase in the PL intensity arising from the exciton recombination. The absence of the WL emission in both SQD and DQD samples implies that $\tau_c$ is much shorter than the radiative decay time of free excitons in both samples. This means that the slowdown of $\tau_c$ in the DQD samples is either insignificant or not substantial enough to make it comparable to the radiative decay time of free excitons. To examine the significance of a prolonged carrier capture time in the spin injection efficiency, we carried out a study of the $P(B)$ dependence as a function of excitation density. We found that the degree of the slope of the polarization curves in magnetic field can be reduced by increasing excitation power under the WL excitation of the DQD samples, as shown in Figure 11, which corresponds to an increase in the $\tau_s^b / \tau_c$ ratio. We attribute this observation to a shorter spin injection time $\tau_c$, expected under a higher excitation density when the effect of the potential fluctuations is reduced due to filling of localized states (i.e. band tail states) of the WL leading to an increase in carrier mobility.

5. Conclusions

We have studied the effects of a longitudinal magnetic field on spin injection and spin detection in several lateral QD structures, including single and aligned double QD structures. Two main effects were observed and have been attributed to spin depolarization in the QD spin detector and the spin injectors (i.e. WL and GaAs), respectively. The first effect gives rise to a drastic increase in the PL and electron spin polarization of the QDs with increasing field strength regardless of field direction, manifested by a dip around zero field. It has been shown to originate from electron spin depolarization driven by the hyperfine interaction for the $X^+$ excitons and the AEI for the $X^0$ excitons of the QDs.
This depolarization effect can be significantly suppressed in applied magnetic fields, leading to an enhanced spin detection efficiency of the studied QDs by a factor of 2.5. The second effect of the longitudinal magnetic field is a nearly linear field dependence of the QD PL (and electron spin) polarization, observed even under linearly polarized optical excitation when no spin polarization is expected to be generated. We have shown that this effect is caused by spin relaxation in the spin injectors (i.e. WL and GaAs), which induces spin imbalance favoring the lower-lying spin state of electrons undergoing a Zeeman splitting. While the first effect related to spin depolarization within the QDs is similar in all of the studied QD structures, we have found a pronounced impact of the structural types on the second effect. The aligned DQD structures exhibit stronger spin relaxation in both WL and GaAs barrier, which is suggested to be a result of a reduced spin relaxation time due to stronger in-plane anisotropy as well as a prolonged carrier capture time due to larger potential fluctuations, as compared with the "isotropic" SQD structure. Such spin relaxation leads to lower spin injection efficiency, approximately by a factor of 2 judging from the spin polarization of the electrons injected into the QDs and the resulting PL polarization.

Our results have demonstrated the need to improve both spin detection and spin injection efficiency in the QD structures. In terms of spin detection by QDs, controlling spin depolarization induced by AEI in neutral QDs and hyperfine interaction in charged QDs is essential. Possible strategies are to explore high-symmetry QDs for suppression of AEI and to carefully prepare nuclear spin environment in QDs by e.g. nuclear field locking [40] to reduce nuclear field fluctuations. Our work also points out the importance of structural design in exploring multiple QD structures for spintronic applications. While multiple QD structures aligned along a preferred direction can provide new opportunities for applications, the very means of achieving this (e.g. in-plane anisotropy) also counteracts their spin-preserving properties. One possible way to circumvent this problem is to improve carrier mobility in the spin injector, i.e. reducing potential fluctuations and trapping near the spin injector/QD interface, thereby reducing the capture times into the QDs. This will limit the extent of spin relaxation experienced in the spin injector before injection of spin polarized electrons into the QDs.

Acknowledgements

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References
[31] Sénes M, Urbaszek B, Marie X, Amand T, Tribollet J, Bernardot F, Testelin C, 
[34] Alon-Braitbart S, Poem E, Fradkin L, Akopian N, Vilan S, Lifshitz E, Ehrenfreund E, 
Gershoni D, Gerardot B, Badolato A et al. 2006 Physica E 32 127
Rev. Lett. 88 223601
Rev. B 69 161301
[37] Chen W M, Buyanova I A, Rudko G Y, Mal'shukov A G, Chao K A, Toropov A A, 
125313
[38] Chen W M, Buyanova I A, Kayanuma K, Chen Z H, Murayama A, Oka Y, Toropov A 
72 1341
459 1105
Figure 1. PL spectra under excitation above the bandgap of the GaAs barrier (the lower curves on the right panels) and PLE spectra of the QD ground state (the lower curves on the left panels) for the SQD (a) and DQD1 (b) sample, obtained at $T = 10$ K. XG, XL, and XH denote the free excitons related to the GaAs, the WL light-hole and the WL heavy-hole band edges, respectively. The upper curves are the corresponding circular polarization degree of the observed luminescence under cw $\sigma^+$, $\sigma^-$ and $\sigma^-$ polarization of the exciting laser beam.
Figure 2. PL spectra under excitation above the bandgap of the GaAs barrier (the lower curves on the right panel) and PLE spectra of the QD ground state (the lower curves on the left panel) for the SQD sample, obtained at $B = -5$ T and $T = 10$ K. The upper curves show the observed optical polarization degree, under the excitation polarization states given for each polarization curve. The labels 1-1 and 2-2 denote the VB and CB LL numbers in the WL hh band between which the excitation transitions occur. A close-up of the PL polarization under $\sigma^-$ polarized excitation in the vicinity of the 1-1 LL transition is shown in the insert. The dashed vertical line marks the position of the 1-1 LL PLE peak. The arrows indicate the ordering in the spin orientations of the photo-generated electrons in the CB of the WL, but their distance does not scale with the actual Zeeman splitting.
Figure 3. (a) Fan diagram of the WL Landau levels and the excitonic lines for the SQD sample, obtained from the PLE spectra of the QD ground state. (b)-(d) PLE spectra of the QD ground state at $B = 3, 5$ and $7$ T, respectively.
Figure 4. QD PL intensity and circular polarization from the DQD2 sample under excitation at a fixed energy within the hh-part of the WL as a function of longitudinal magnetic field. The excitation polarization state for the PL polarization data is as specified. The arrows indicate the electron spin orientations of the spin sublevels of the corresponding CB LL states in the WL.

Figure 5. Circular polarization of the QD PL as a function of longitudinal magnetic field for SQD (a), DQD1 (b) and DQD2 (c). The excitation photon energy was set above the GaAs bandgap for all curves except the one in (a) that is marked by "qr" when the excitation energy was set below the WL heavy-hole exciton yielding quasi-resonant excitation of the QDs. The polarization state of the excitation light for each curve is as indicated.
Figure 6. (a) PL circular polarization from the SQD as a function of longitudinal magnetic field under the GaAs excitation by cw $\sigma^+$ (the filled circles) and alternating circularly polarized light (the open circles). The solid curve shows the contribution from the AEI, fitted using Eq. (1). (b) PL polarization from the DQD1 as a function of magnetic field under cw $\sigma^+$ (the upper curve) and $\sigma^x$ (the lower curve) excitation. The solid lines are fits of Eq. (1) and Eq. (2) to the corresponding data using the same AEI strength $\omega$.

Figure 7. Power dependence of the PL circular polarization from the SQD under cw $\sigma^+$ excitation (the filled symbols) and alternating circularly polarized excitation (the empty symbols). The lines are a guide to the eye.
Figure 8. Circular polarization of the QD PL as a function of longitudinal magnetic field under quasi-resonant excitation of the QDs (the grey curves), excitation in the WL hh band (the curves with open circles) and above the GaAs bandgap (the curves with open triangles).
Figure 9. PLE intensity and polarization under $\sigma^+$, $\sigma^\times$ and $\sigma^-$ excitation from the SQD (a) and DQD2 (b) sample at $B = -7$ T.
Figure 10. (a) Sketch of the processes accounted for in our rate equation modeling. (b) The QD electron spin polarization degree for short ($\tau_s^b = \tau_e$, the thick solid lines) and long ($\tau_s^b = 100 \tau_e$, the thin dashed lines) spin relaxation times in the barrier as calculated by solving the rate equations Eq. (3). In the simulations, $G_\uparrow / G_\downarrow = 1.5$, $G_\uparrow / G_\downarrow = 1.5$ and $G_\downarrow = G_\uparrow$ are assumed for $\sigma^+$, $\sigma^-$ and $\sigma^x$ excitation conditions.

Figure 11. QD PL circular polarization degree of the DQD1 sample as a function of magnetic field under excitation in the WL with high (the empty circles) and low (the solid lines) excitation power. $\sigma^+$, $\sigma^-$ and $\sigma^x$ denote the excitation polarization states employed.