Efficiency of spin injection in novel InAs quantum dot structures: exciton vs. free carrier injection

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Abstract. Unambiguous experimental evidence for a significant difference in efficiency of excitonic vs. free carrier spin injection is provided in novel laterally arranged self-assembled InAs/GaAs quantum dot structures, from optical orientation and tunable laser spectroscopy. A lower efficiency of exciton spin injection as compared to free carrier spin injection from wetting layers into QDs results in a distinct feature in luminescence polarization of the QDs as a function of excitation photon energy. It is shown that this difference is not related to carrier density and state-filling effects arising from the difference in optical absorption efficiency between the excitons and free carriers. Rather, it is a genuine property for exciton spin injection that suffers stronger spin relaxation due to Coulomb exchange interaction.

1. Introduction

Three-dimensionally confined carriers in quantum dots (QDs) are a topic of great research interest at present due to their extremely long spin lifetimes [1]. In(Ga)As QDs are a model system for spin-functionality-enabled quantum devices as they can be controllably positioned [2] and embedded into active device structures [3]. At the basis of these spintronic applications is the ability to prepare the confined carriers in a defined spin state. Regardless of whether this is accomplished electrically or optically, the carriers are usually injected non-resonantly, i.e. in one of the surrounding layers and energetically well above their final ground state. During their transfer and energy relaxation, they are subject to various spin relaxation processes which depend on the exact injection mechanism, being either individual carrier or exciton injection [4]. Previously, we showed results evidencing lower spin injection efficiency under resonant optical orientation of free excitons (FE) compared to free carriers of the wetting layer (WL) and interpreted this as a result of accelerated spin relaxation due to exchange interaction inside the injected exciton [5]. The aim of the present work is to investigate some aspects of injected carrier density on the spin injection efficiency into the QDs.

2. Materials and Methods

Several InAs/GaAs-based QD structures have been studied, such as self-assembled single QDs (denoted by SQD) and lateral QD rings (denoted by QDR), consisting of five to seven dots per ring. The nominally undoped structures were grown in Stranski-Krastanov growth mode by molecular beam epitaxy on (001) semi-insulating GaAs substrate. Details on the growth process can be found in [6].
To investigate spin injection into the QDs from the InGaAs WL, we used optical pumping of spin-polarized carriers in the WL and monitored the circular polarization of QD luminescence. A tunable Ti:sapphire laser was used as an excitation source, whose circular polarization could be controlled by a rotatable quarter-wave plate. The samples were kept at a temperature of either 6 K or 10 K for the measurements to be presented here. Polarization-resolved detection of the emitted photoluminescence (PL) was achieved by a rotatable quarter-wave plate in conjunction with a linear polarizer in front of a grating monochromator, equipped either with a liquid-nitrogen-cooled Ge-detector in case of power-dependent PL measurements or with a liquid-nitrogen-cooled infra-red-enhanced Si-CCD camera for all other measurements. The degree of luminescence circular polarization was defined as

\[ P_\alpha = \frac{(I_\alpha^+ - I_\alpha^-)}{(I_\alpha^+ + I_\alpha^-)}, \]

where \( I_\alpha^+ \) and \( I_\alpha^- \) denote the PL intensities of \( \sigma^+ \) and \( \sigma^- \) polarized luminescence, respectively. The excitation polarization state \( \alpha \) was adjusted to either \( \sigma^+ \) or \( \sigma^- \). Excitation power was varied by inserting neutral density filters in the excitation light path.

Optical selection rules in the WL, where both strain and quantum confinement split heavy- and light-hole valence band states, allow creation of spin-polarized carriers/excitons by circularly polarized optical excitation along the direction normal to the WL plane. The degree of luminescence circular polarization can be used as a measure of spin conservation during injection and energy relaxation within the QD as well as spin relaxation in the QD ground state, due to the validity of the same optical selection rules inside the QDs.

3. Results and Discussion

Figure 1 shows typical PL spectra from the studied QD structures, which are dominated by the exciton emission associated with the QD ground state. With increasing excitation power, PL emissions from the QD excited states gain strength due to the well-known state-filling effect in QDs. A close-up PL/PLE map of the ground state PL under excitation nearby the WL absorption edge, taken the QDR sample as an example, is presented in figure 2(a). Similar PL/PLE maps were observed in all studied samples. A PLE cross-section through these maps at the QD ground state PL maximum, shown in figure 2(b), exhibits a distinct peak arising from the heavy-hole (hh) FE absorption in the WL (denoted by XH), followed at shorter excitation wavelengths by an absorption continuum due to band-to-band transitions between the hh valence band and the conduction band. Figures 2(c) and 2(d) show the corresponding PL polarization under circularly polarized excitation over the same excitation and detection wavelength ranges as that in figures 2(a) and 2(b). Excitation of free carriers from the WL hh VB results in QD ground state PL polarization of up to almost 15%. On tuning the excitation wavelength to the WL hh FE resonance, the PL polarization noticeably drops down to 7%.

As shown by experiments[7,8] and theory[9], the ground state of neutral excitons in QDs at low temperature is characterized by two linearly polarized eigenstates, due to an anisotropic electron-hole exchange interaction. A likely explanation of the observed circular polarization is the observation of trion (charged exciton) luminescence[10], originating from unintentional doping during MBE growth. In trions, exchange interaction is cancelled[11], leading to circularly polarized ground states. As trion and neutral exciton luminescence cannot be distinguished in ensemble measurements due to their small energetic difference[12], the ground state PL band may contain PL of both species and thus

Figure 1. PL spectra (PL intensity \( I \) over detection wavelength \( \lambda_{det} \)) of the studied samples as a function of excitation power: SQD (a) and QDR (b), taken at 6 K under above-barrier excitation (514 nm).
show a non-vanishing circular polarization. Furthermore, the presence of trions opens the possibility of dynamic nuclear polarization (DNP) of the lattice atoms forming the QDs via the hyperfine interaction between the spin-polarized electrons in trions and the nuclei\cite{13,14}. The long-lived DNP will act as an effective magnetic (Overhauser) field, possibly suppressing the effect of anisotropic exchange interaction, which will lead to restoration of circularly polarized eigenstates for the neutral exciton and the observation of corresponding circularly polarized luminescence. Additionally the Overhauser field may also contribute to an increase in carrier spin relaxation time, again assisting the circular polarization of QD luminescence for both neutral and charged excitons. It must be pointed out that all of the aforementioned mechanisms for the observation of the circularly polarized QD PL originate from electron spin polarization in the QDs that is generated by spin injection from, in our case, the WL. Therefore, the degree of the QD PL polarization can be taken as a measure of spin injection efficiency from the WL. The observed decrease in QD PL polarization upon spin injection from the WL hh excitons, shown in figure 2, is thus an indication that the efficiency of exciton spin injection is lower than that of uncorrelated free-carrier spin injection.

It should be noted that the observed XH peak in PLE implies a higher concentration of injected carriers, leading to a higher concentration of QD ground state excitons. Such a change in the QD exciton density can, in principle, affect the QD PL polarization via several mechanisms. First, due to Pauli blocking, the QD ground state can only be occupied by two carriers of opposite spin\cite{15} at the same time. Thus, with increasing injected carrier density, we can expect the QD ground state luminescence to show weaker polarization, as the second (counter-oriented) spin-state starts to be filled by carriers having undergone spin flips during injection and energy relaxation into the QD ground state. Second, an increase in spin-polarized electron injection into the QDs may lead to an increase in dynamic nuclear polarization in the QDs\cite{14} and thus to an increasing degree of luminescence circular polarization. The observed increasing carrier density at resonant excitation of the WL hh exciton may be expected to lead to either rising or falling PL polarization, depending on the relative importance of state filling vs. DNP effects. To exclude such carrier-density-related effects from our measurements, we conducted PL polarization measurements at several excitation wavelengths around the WL XH resonance, while adjusting the incident laser power to ensure identical PL intensity of the QD ground state. This way, the carrier density in the QDs was kept constant and the corresponding effects on luminescence polarization were eliminated. The results of these measurements are presented in figure 3.

From the PL/PLE polarization map in figure 3(d) and its corresponding cross-section (PLE polarization) at the ground state PL peak position in figure 3(c), it is clear that QD ground state luminescence polarization decreases at resonant XH excitation. As carrier density effects no longer play a role here, we therefore conclude that this effect must be due to a lower efficiency in exciton XH spin injection into the QDs as compared with spin injection of free carriers. We attribute the origin of this reduction to more efficient spin relaxation of correlated carriers within the excitons, promoted by the Coulomb exchange interaction. As a direct consequence, the spin polarization of the electrons in the QDs should decrease upon exciton spin injection from the WL. The lower electron spin
polarization may subsequently lead to a weaker DNP effect and a reduced Overhauser field, resulting in increasing importance of the anisotropic exchange interaction of the neutral excitons and stronger electron spin relaxation in the QDs. All these effects lead to a decreasing PL polarization of the trions and neutral excitons in the QD, as observed in our case.

4. Conclusion

In conclusion, we have demonstrated the significance of a distinction between excitonic and free carrier spin injection from an InGaAs WL into novel InAs multiple-QD structures. Reduced PL polarization of the QD ground state under injection of resonantly created heavy hole excitons in the WL is confirmed to be a specific property of exciton injection and is not related to the differing absorption efficiency of WL free carrier continuum and WL free exciton resonance leading to differing carrier densities in the QD structures. Thus we attribute this effect to accelerated spin relaxation of the excitons promoted by the Coulomb exchange interaction between the carriers.

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