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Effect of Remote Doping on Photoluminescence and Carrier Spin Dynamics in InGaAs Quantum Dots

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Abstract

We have investigated carrier spin injection and relaxation in self-assembled remote p-doped InGaAs quantum dots (QDs) grown by molecular beam epitaxy (MBE). Fast spin injection within pico-second time range has been observed after laser pulse excitation, which yields appreciably high photoluminescence (PL) circular polarization degree (*CPD*) up to ～60%. The overall PL intensity of QD ensemble becomes stronger with increasing number of residual holes in QD from acceptors, while the *CPD* stays almost the same. This effect is found as caused respectively by the minority holes inside QDs that limits the emission strength and injected majority electrons of high mobility that determines PL *CPD*.

1. Introduction

Semiconductor quantum dots (QDs) have been drawing enormous research interest since its emergence. This is driven by the fact that three dimensional quantum confinement of QDs not only yields sharp and discrete radiative transitions with high quantum efficiency, but also effectively suppresses the carrier spin relaxation process mediated by D'yakonov-Perel and Elliot-Yafet mechnisms [1]. Therefore, optically bright electronic states with long spin lifetime was enabled and observed in QDs, which paves the way for its future application as spin-light emitting and laser diode (spin-LED and LD).

Vital to any spin-based photonic device is functional p and n-doped junctions, where electrons and holes can recombine efficiently. By bringing the spin property of carrier into the context of light emitting, all the optical processes which are common in conventional devices need to be taken great care of and reconsidered. This is because the spin-oriented carriers are susceptible to electrostatic scatterings e.g. by impurities, and therefore can become a central issue for those p and/or n-type doped semiconductors. Open questions still remain as to the influence of doping on spin injection and relaxation in QDs, which holds paramount importance in real device performance.

To understand these unclarified issues, we performed the time-resolved optical spin orientation measurement on remote modulation p-doped InGaAs QDs, where we observed an enhanced PL with higher doping level and a stable *CPD* from QD excited states (ESs). Such effects are induced respectively by hole-limited radiative recombination and electron spin-determined PL circular polarization.

2. Experimental procedure

Crystal growth of modulation p-doped InGaAs QDs

A GaAs (100) substrate was used for the MBE growth of QDs, which is deposited by a 400 nm-thick GaAs buffer layer. Then a beryllium acceptor-doped GaAs layer was grown with a dopant areal density of 6.4×10^{17} cm^{-2}. Seperated by 10 nm intrinsic GaAs spacer from the doped layer, the $In_{0.66}Ga_{0.34}As$ QDs were grown, with a sheet density of 7×10^{10} cm^{-2}, and capped by 50 nm GaAs. For characterization of morphology, another layer of $In_{0.66}Ga_{0.34}As$ QDs was grown on the surface of capping layer. For comparative study of doping effect on QD PL, the thickness of modulation doping layer was varied for 14 and 28 nm respectively, which corresponds approximately to a residual hole number of 6 and 12 per dot, and we label as QD6 and QD12, henceforth. For comparison, we also grow the reference sample without doping and denote it as QD0.

Time-resolved optical spin orientation measurement

Time-resolved optical spin orientation spectroscopy was performed using a Ti: Sapphire pulsed laser as excitation source. Circular-polarized laser was tuned slightly above the bandgap of GaAs barrier to excite spin-polarized electrons and holes. The circular photoluminescence (PL) from QDs are differentiated by optic set of quarter-wave-plate and linear polarizer, and finally detected by streak camera. It should be noted that hole spins are easily depolarized in barrier and QDs due to stronger spin-orbit interaction and intra-heavy and light hole valence band relaxation [2]. As a result, PL circularity is solely determined by electron spin polarization which can be derived from PL circular polarization degree (*CPD*) as,

$$CPD = (I^{\sigma^+} - I^{\sigma-}) / (I^{\sigma^+} + I^{\sigma-}) \qquad (1)$$

here, I^{σ^+} ($I^{\sigma-}$) is PL intensity of σ^+ (σ^-)-polarized component, respectively.

3. Results and discussion

Effect of beryllium doping on PL intensity and CPD

We investigated the doping influence on the circular-polarized PL properties of QD emission. Shown in Fig. (a) and (b) are PL spectra measured from QD0 and QD12 at 6K under σ+-laser excitation of GaAs bandgap. Both samples feature broad and strongly co-polarized PL within the spectral range 880-960 nm, which arises from the excited

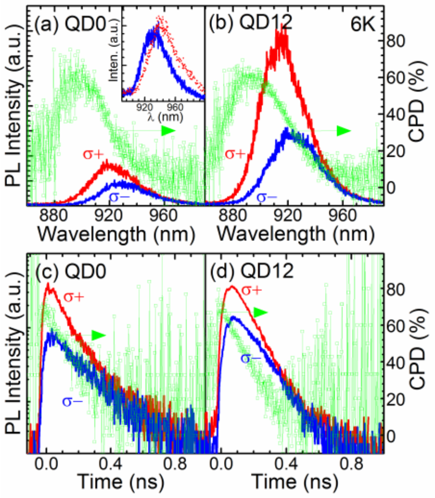

Figure 1. (a)-(b) Circular-polarized PL spectra from QD0 and QD12 at 6 K, the *CPD* is presented as open square. PL emission by unpolarized below-GaAs bandgap laser excitation is also shown for QD0 (solid line) and QD12 (dashed line) in the inset of (a). (c)-(d) the corresponding excited state circular-polarized PL decay spectrally integrated within 890-920 nm, together with transient *CPD* denoted as open square.

states of QD ensemble. The reason why we didn't see ground state emission is that its transition energy falls below the spectral limit of our streak camera, therefore resulting in an insufficient PL detection. It is clearly shown QD12 gives stronger PL intensity than QD0 in both detection polarizations by around four times, implying effect of holes on PL recombination process. Indeed, by looking at carrier migration in GaAs barrier after photo-creation, electrons move faster than holes due to a higher mobility and are injected into QDs prior to the holes. As a consequence, inequilibrium population of carriers is present, where electron is the majority. In such condition, the PL radiative recombination rate will be affected the residual holes provided by acceptor dopants. This explains why QD12 sample with more holes per dot is stronger in PL than that of QD0. To confirm, we measured on the QD6 sample (not shown here) and found its PL intensity is in-between QW0 and QD12. In addition, the un-polarized excitation laser was tuned below GaAs bandgap to resonantly generate electron-hole pairs inside QDs, where electron and hole population are almost same. Indeed, we observed nearly equal PL emission from QD0 (solid line) and QD12 (dashed line) as shown in the inset of Fig. 1(a). All these observation are in support of our conclusion. Then, the *CPD* was calculated based on circular PL to derive the electron spin polarization. As can be seen from open square in Fig. (a) and (b), both samples exhibit same *CPD* value as high as ~60 %, indicating an efficient electron spin injection from barrier to QDs. The same *CPD* value is not hard to understand because the electron spin injection process are the same among all samples. Toward lower energy side, *CPD* shows monotonous decrease. This is caused by the spin flip-induced electron relaxation to lower energy states as commonly observed in QD systems.

Circular-polarized PL transients from QD ES were monitored to derive the temporal evolution of *CPD*. Fig. 1 (c) and (d) display the corresponding PL decays from QD0 and QD12, which are spectrally integrated within 890-920 nm. Both samples show short lifetime less than 400 ps, dominated by carrier energy relaxation to lower-lying states, together with a fast *CPD* decay (open square) reflecting a spin relaxation time ~300 ps, which is close to those reported by others [3]. The fast spin-flip time of QD ES compared with GS (~1 ns) can be accounted by two factors, the first is random nuclear filed induced by hyperfine interaction of electron spin with the surrounding lattice nuclei spin. While the second is larger spin-orbit coupling experienced by ES elelctron due to its extended orbital wavefunction than GS. Therefore, QDs of smaller size are desired for prolonging the electron spin relaxation time.

An interesting aspect of remote p-type doping is that the acceptor can not only be used to enhance QD PL via proving additional holes, but also function as efficient spin injection channel. The acceptor bound excitons (ABX) are formed by trapping free carriers at dopant site. Within quantum-tunneling distance from QDs, the electron and hole spin in ABX complex is able to be transferred to QD immediately without spin loss. Such process renders another way to achieve efficient spin injection from barrier to QDs, which is now also under study.

3. Conclusions

High-density p-type remote modulation-doped InGaAs QDs have been investigated for spin injection process. We demonstrate the PL intensity from QDs can be increased by higher doping level, while the *CPD* is only determined by electron spin polarization. The QD ES exhibits fast spin-flip rate which are affected by nuclear spin fluctuation and stronger spin-orbit interaction. More importantly, the ABX is considered as a promising candidate for efficient and fast spin injection into QDs.

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