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Author(s)	Kobayashi, H.; Kumano, H.; Endo, M.; Jo, M.; Suemune, I.; Sasakura, H.; Adachi, S.; Muto, S.
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Highly circular-polarized single photon generation from a single quantum dot at zero magnetic field

H. Kobayashi^a, H. Kumano^{a,b,*}, M. Endo^a, M. Jo^{a,c}, I. Suemune^{a,b}

H. Sasakura^b, S. Adachi^{b,d} and S. Muto^{b,d}

^a RIES, Hokkaido University, Sapporo 001-0021, Japan

^b Japan Science and Technology Corporation (CREST), Saitama 332-0012, Japan

^c JSPS

^d Graduate School of Engineering, Hokkaido University

Abstract

Origin of sharp photoluminescence lines observed from an InAlAs quantum dot was identified with the measurements of excitation-power dependences and polarization correlations, together with photon correlation measurements. Single photon emission with high degree of circular polarization (DCP) up to 60% was observed from a positively charged exciton (trion) state in the single quantum dot under non-resonant excitation at zero magnetic field.

Keywords: single quantum dot; single photon; charged exciton; circular polarization; spin flip

1. Introduction

In recent years, quantum cryptography is attracting much more attention due to its highly secure nature [1]. Since it is based on physical principles such as indivisibility and non-cloning of single photons instead of mathematical complexity, completely secure communication which is free from eavesdropping will be possible in principle. One of the most important protocols for implementing the quantum cryptography is BB84 [2]. Since the BB84 is based on photon polarization states belonging to two non-orthogonal bases, generation of single photons from a quantum dot (QD) with arbitrary polarization states is prerequisite. However, in most cases, neutral exciton emission shows only energy-split linearly polarized photon emissions due to the fine structure splitting (FSS) of exciton states in elongated QDs [3-5], where the polarization information leaks to the photon energy. On the other hand, according to Kramers theorem, the eigenstates of the singly-charged exciton (trion) are degenerate in the absence of a magnetic field [6,7]. Therefore, spin states of single trion will be directly transferred into photon circular polarization through a recombination

* Corresponding author. Tel & Fax: +81-11-706-9334.
E-mail address: kumano@es.hokudai.ac.jp (H. Kumano)

process [8,9].

In this paper, the origin of luminescence peaks observed from a single InAlAs QD was identified and their FSS was studied. Positively charged exciton was confirmed to be free from FSS, and single photon emission with the high degree of circular polarization (DCP) up to 60% was demonstrated at zero magnetic field under non-resonant excitation.

2. Sample preparation and measurement setup

The QDs sample was grown on a (001) GaAs substrate by molecular-beam epitaxy. The QDs were prepared in Stranski-Krastanow (S-K) growth mode on $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layers and were sandwiched with $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layers. The topmost surface was terminated with a GaAs cap layer. After the growth, the sample was etched into mesa structures with diameters of ~ 150 nm for isolating single QD. For a single-dot spectroscopy, the sample was held in a closed-loop He cryostat and was kept at 22K. A continuous-wave (CW) Ti: sapphire laser at the wavelength of 730 nm was used as a right-circularly polarized (σ^+) excitation source using a quarter-wave plate. An objective lens with the numerical aperture (NA) of 0.42 focused the laser beam on one of the mesa structures and collected luminescence emitted from several QDs in the mesa. Collected luminescence was dispersed by a 0.64-m triple monochromator, and the photoluminescence (PL) with the σ^+ polarization or the left-circular (σ^-) one was analyzed by a set of quarter-wave plate and a fixed Glan-Thomson linear polarizer in front of the monochromator equipped with a Si charge-coupled-device detector in order to avoid any detection artifacts due to the anisotropic response function of our setup.

3. Results and discussion

Figure 1 shows a PL spectrum measured from an InAlAs single QD studied in this work. Apparently three sharp emission lines designated as L1, L2, and L3 were observed, where the L3 peak was superposed with the sharp line designated as L4. The energy separation between the L3 and L4 peaks was about 150 μeV and these two peaks could be resolved only with the high-resolution measurements. The origin of these peaks was studied with the excitation power dependence [10]. The L2 peak showed the excitation power dependence proportional to $(P_{\text{exc}})^2$ where P_{exc} is the excitation power, and the L1 and L3 (including L4) peaks showed the linear dependence on P_{exc} . Therefore the L2 peak will be of biexcitonic nature while the L1 and L3 peaks will be of excitonic one. Time-resolved measurements were also performed on the L2 and L3 (also

including L4) peaks with a streak camera and the recombination lifetimes of 0.55 ns and 1.02 ns were estimated, respectively [11]. This shorter lifetime for the L2 peak is consistent with the assignment of the biexcitonic nature.

The polarization-selective PL measurements were performed on the L2 and L3 peaks, where the L4 peak was separated with the high-resolution measurements. By the PL measurements with the polarizations perpendicular to each other (called relatively as V- and H-polarizations), the energy splitting of the L2 and L3 peaks became clear as shown in Figs. 2(a) and 2(b), respectively. Especially, the L2 and L3 peaks showed the anti-correlations, i.e., the V-polarized emission showed the blue-shift for the L2 peak and the red-shift for the L3 peak while the H-polarized emission showed the red-shift for the L2 peak and the blue-shift for the L3 peak. This anti-correlated polarizations can be well understood by the spin-splitting of the neutral exciton state [12] in the neutral biexciton-exciton cascade transitions, and therefore the L2 and L3 peaks were attributed to the neutral biexciton (XX^0) and the neutral exciton (X^0), respectively. The L4 peak shown in Fig. 2(b) as well as the L1 peak showed no FSS. Especially the L4 peak showed the very sharp peak whose full width at half maximum (FWHM) was ~ 20 μeV . These observations suggest that the L1 and L4 peaks are charged excitons (trions) since the formation of spin singlet states cancels the electron-hole exchange interactions which induce the FSS [12]. Considering the normal trend that the negatively charged excitons (X^-) and the positively charged ones (X^+) show up in the lower and higher energy sides of the neutral excitons, respectively [13], the L1 and L4 peaks were attributed to the X^- and X^+ charged excitons.

To examine the emission correlations among the various excitons, the photon correlation measurements were performed. It revealed that the sharp L4 emission line attributed to the positive trion (X^+) showed clear photon anti-bunching behavior against the X^0 and X^- emissions when the sharp X^+ emission line was introduced into the so-called HBT setup [14] together with either X^0 or X^- emission lines. This demonstrates that the excitonic emissions of the neutral exciton X^0 and the charged excitons X^+ and X^- are single-photon emission processes delivering single photon to either one of the three excitonic processes under every pulsed photo-excitation. In other words the excitonic single-photon generation process is exclusive to each other originating from the same QD.

When the QD was resonantly excited at the energy one longitudinal optical (LO) phonon energy higher than the neutral exciton emission energy, the PL peaks except for the X^+ charged exciton disappeared as shown in Fig. 1. Since the resonant excitation will generate electron-hole pairs in the QD, this shows that the QD is singly positively charged before the photo-excitation.

PL spectra under the circularly polarized excitation were examined. The expanded spectra at around the X^0 and X^+ emissions are shown in Fig. 3, where the QD was excited via the wetting layer with the σ^+ polarization and the luminescence was analyzed with the σ^+ or σ^- circular polarizations. The striking difference in the X^+ PL intensities between the σ^+ and σ^- polarizations indicates the

efficient spin-state conservation during the overall processes including the capture, energy relaxation, and recombination processes in spite of the non-resonant excitation above the X^+ emission energy by ~ 110 meV. On the other hand, the X^0 emission exhibited little difference between the two circular polarizations as shown in Fig. 3. This is as expected in usual elongated QDs where the e-h exchange interaction spin-splits the exciton states and therefore the linearly-polarized photon emission takes place.

The DCP of the X^+ emission defined by $(I_{\sigma^+} - I_{\sigma^-}) / (I_{\sigma^+} + I_{\sigma^-})$ was examined, where I_{σ^+} and I_{σ^-} are the luminescence intensities with the circular polarizations of σ^+ and σ^- , respectively. The measured luminescence spectra were well fitted by a Lorentzian lineshape function and the I_{σ^+} and I_{σ^-} intensities were carefully evaluated. The resultant DCP measured under the σ^+ -polarized excitation was estimated to be 58.7%. This is the highest DCP value ever reported without applying magnetic field and under the non-resonant excitations.

4. Conclusions

Single photon emission with high degree of circular polarization at zero magnetic field is demonstrated from a positively charged exciton state under non-resonant excitation. Resultant DCP as high as $\sim 60\%$ indicates that the spin relaxation is strongly suppressed during capture and thermalization processes into a trion ground states in a QD. Detailed mechanism of the spin flip in a QD is now under study, but the phonon-mediated spin flip may be involved [15] and the circumnavigation of the phonon scattering phenomena could be a crucial issue for the further improvement of the DCP.

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Figure captions

Figure 1

PL spectra measured from an InAlAs single QD under the wetting layer excitation and the resonant excitation at the photon energy one LO phonon higher than the L3 peak.

Figure 2

PL spectra measured with no polarization (solid line), vertical (V) polarization (dashed line), and horizontal (H) polarization (dot and dash line) on (a) L2 peak, (b) L3, and L4 peak.

Figure 3

PL spectra measured under the right-circularly polarized (σ^+) excitation. The solid line measured with σ^+ circular polarization and the dashed line with σ^- circular polarizations.

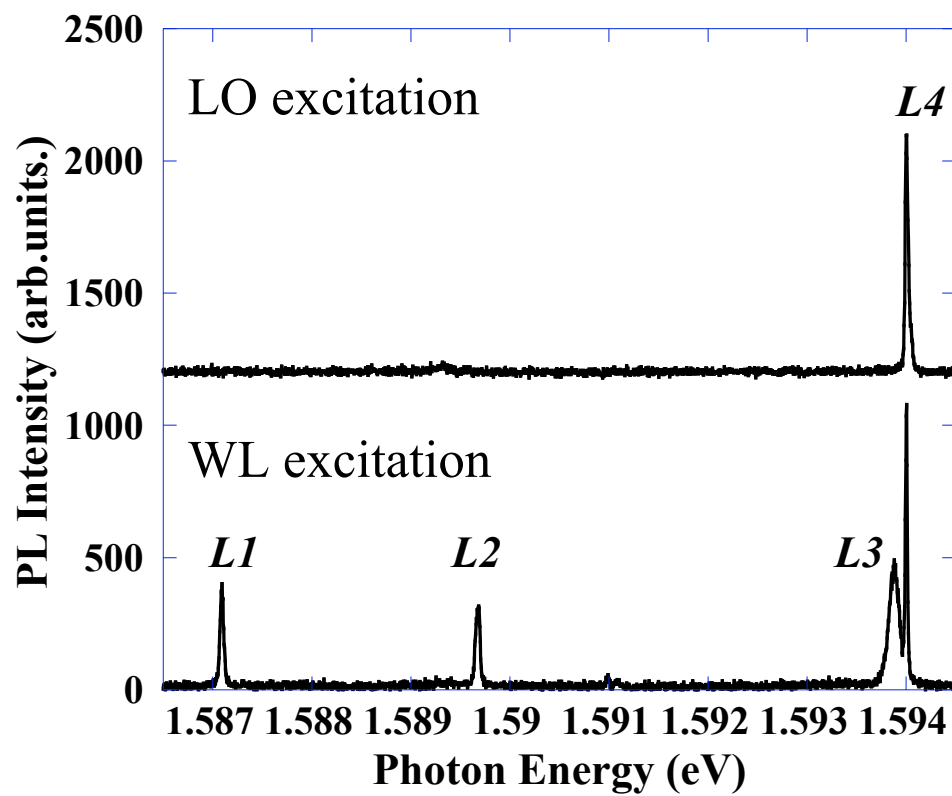


Figure 1, Kobayashi *et al.*

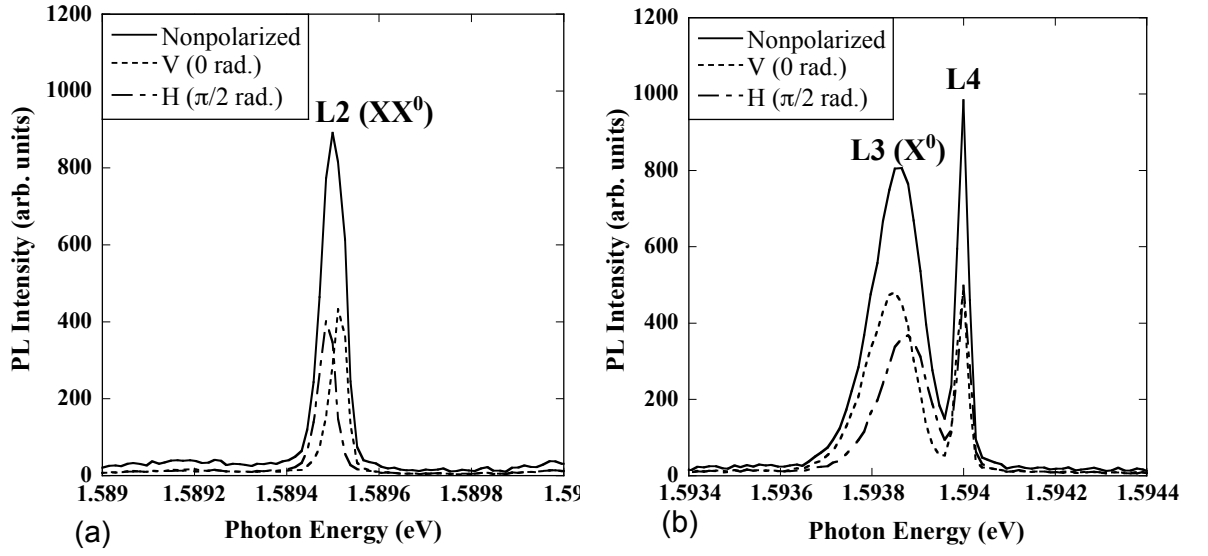


Figure 2, Kobayashi *et al.*

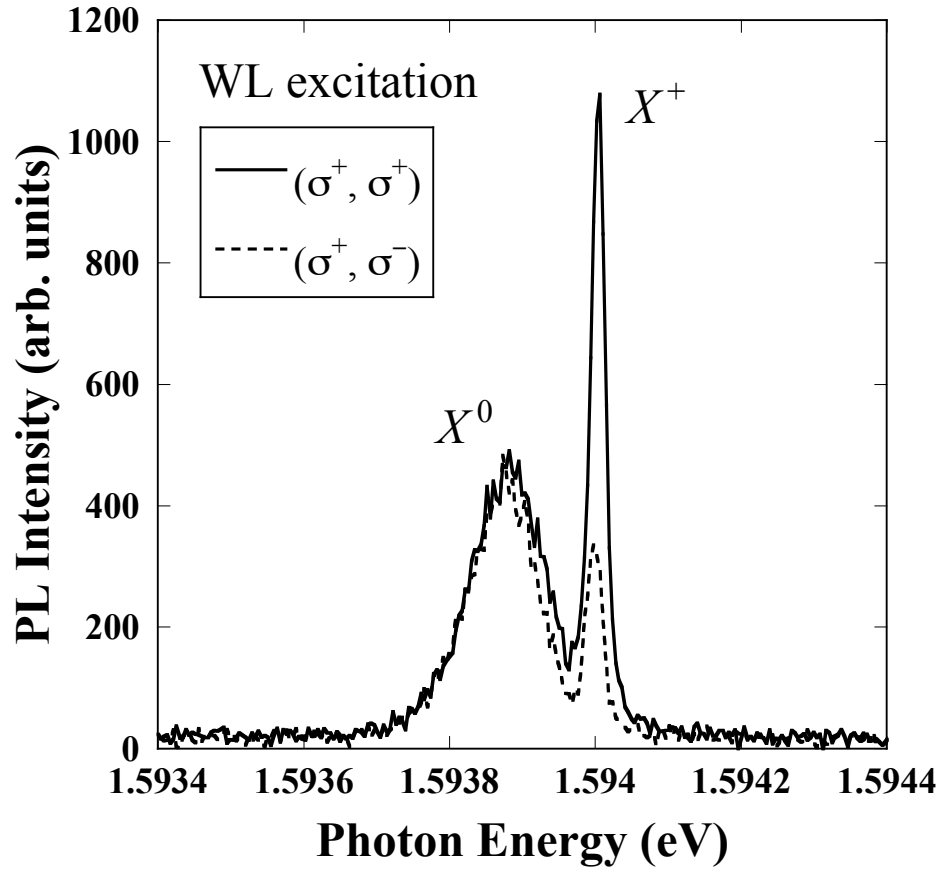


Figure 3, Kobayashi *et al.*