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Triggered single photon emission and cross-correlation properties in InAlAs quantum dot

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Abstract

Triggered single-photon generation from InAlAs quantum dot (QD) was demonstrated for the first time. Emitted photon energy coincides with high detection efficiency range of Si single-photon detectors, which is highly suitable for free-space communication. Single-QD spectroscopy and crossed photon correlation measurements unambiguously revealed that several emitting lines observed in a single mesa structure originated from the identical QD, and two temporary competing decay processes associated with neutral states and charged states were identified. Presence of the competing process is also inferred from an analysis of steady-state photoluminescence intensities. Formation process of charged exciton in QD is also discussed.

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1. Introduction

Highly secure communication is attracting much attention, and single-photon emitters (SPEs) based on semiconductor quantum dot (QD) have been intensively studied as a light source for quantum key distribution (QKD) [1–9]. SPE is an indispensable key element for QKD, allowing for an efficient unconditional secret key distribution [10]. Fiber-based QKD is a main stream for this direction; however, free-space QKD [11] will play a complementary role for flexible QKD networks. Since whole bit-rate of QKD is determined by a product of SPE's efficiency and detection efficiency of single-photon detector, fabrication of SPE whose emitting photon wavelength matches well with the high efficiency range of Si single-photon detector (600–800 nm) is essential for the realization of highly efficient free-space QKD [11]. However, most studies were focused on In(Ga)As-based materials or wideband- gap materials, and as far as we know, very few materials applicable for this purpose are available [4]. In this paper, single InAlAs QD is prepared which can emit photons at around 780nm and triggered single-photon generation from InAlAs QD is demonstrated for the first time. Moreover, sequential photon pair generation and competing decay processes are identified in terms of crossed photon correlation measurements. These results ensure that cross-correlation measurement is a direct monitor of specific single biexciton decay, and that neutral and charged excitons are exclusively formed in a same QD. Formation process of charged exciton in QD will be also discussed.

2. Experimental

A QDs sample was grown on a (001) GaAs substrate by molecular-beam epitaxy. It had two-stacks of QDs layer made of $In_{0.75}Al_{0.25}As$ and $In_{0.7}Ga_{0.3}As$ separated with an $Al_{0.3}Ga_{0.7}As$ layer but the coupling effects between the two QDs layers were negligible in this sample. This work was focused on the $In_{0.75}Al_{0.25}As$ QDs layer. These QDs layers were grown in a Stranski-Krastanow mode and were sandwiched by $Al_{0.3}Ga_{0.7}As$ layers. After the growth, mesa structures were formed by electron-beam lithography and wet chemical etching. Further details of this sample were described in Ref. 12 and 18. All the optical measurements were carried out at 22K. A CW He-Ne laser and a mode-locked pulsed Ti:Sapphire laser were employed for above-band and wetting layer (WL) excitation, respectively. The laser beam was focused on one of the mesa structures by employing a x50 objective lens and luminescence was collected through the same lens. Collected luminescence was dispersed by a monochromator and was detected by a Si-charge-coupled device detector or by a streak camera for spectrum measurements and time-resolved measurements, respectively. Verification of single photon emission was performed with a Hanbury-Brown and Twiss correlation measurement setup [13]. In this case, emitted light was split by a 50/50 beamsplitter into separate arms and directed toward two 0.2-m monochromators for spectral filtering before being detected by Si-single photon counting avalanche photodiodes (APDs). Second-order photon correlation functions, $g^{(2)}(\tau)$, were measured for both auto- and cross-correlation configurations. Here τ denotes a delay time between two photon-emission events. The outputs of APDs served as start and stop signals for photon correlation measurement. The typical total count rate was ~1.5×10^4 cps.

3. Results and Discussion

Figure 1(a) shows a streak image obtained from a single mesa structure under WL excitation. Time-integrated PL spectrum (solid line) and steady-state PL spectrum (dotted line) are illustrated in fig. 1(b). Several sharp emission lines labeled L1-L4 were resolved. Linear (bi-linear) increase of PL intensity with excitation power for L1 (L3) was observed as shown in the inset of Fig. 1, which ensures that L1 (L3) is attributed to neutral exciton X^0 (biexciton XX^0) emission.

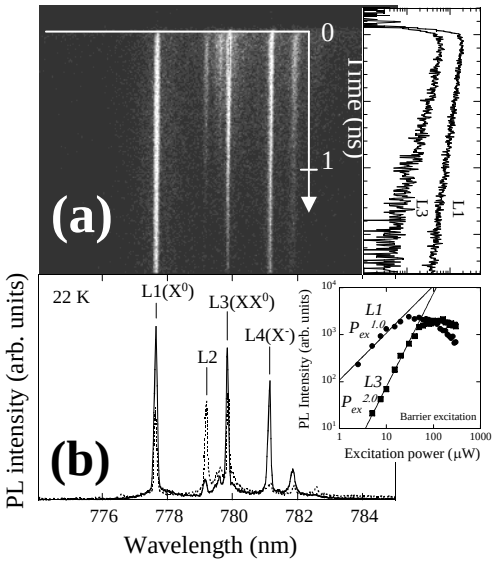

Fig. 1 (a) Streak image of emission lines from single mesa structure under WL excitation. (b) Time-integrated PL spectrum corresponds to the streak image (solid line) and above-band excited PL spectrum (dotted line). Excitation power dependence is also shown for L1 and L3 lines.

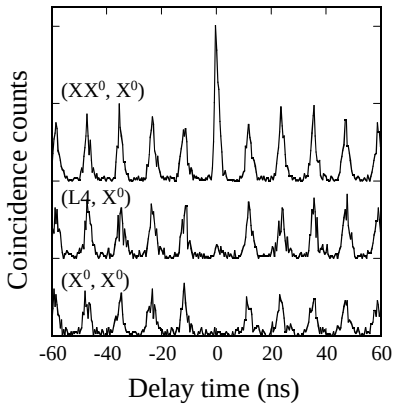

Fig. 2 Normalized coincidence counts for three configurations; (start, stop) = (XX^0,X^0), (L4, X^0) and (X^0,X^0).

In order to investigate a photon statistics of emitted photons from X^0, photon correlation measurements were performed and result is shown in Fig. 2. Clear antibunching at $\tau = 0$ is observed,

which indicates highly pure triggered single photon generation from the InAlAs QD. Emitted photon wavelength coincides well with the high detection efficiency range of Si-single photon detector, which is quite promising for the highly efficient free-space QKD.

Cross-correlation measurements under (start, stop) = (XX^0, X^0) configuration showed clear photon bunching, which indicates triggered XX^0 emission instantaneously followed by the X^0 emission. In order to assign the L4 line, cross-correlation measurement under (start, stop) = (L4, X^0) configuration was carried out. Clear antibunching was observed as shown in Fig. 2. This result is interpreted that X^0 and L4 belong to exclusive decay processes, thus simultaneous decay at one pulse excitation event hardly takes place. This is a clear evidence that the two emission lines are associated with two competing quantum systems exclusively formed in the same QD, which indicates charged excitonic origin (X*) of L4 line. It should be stressed that cross-correlation measurements can also provide information on spatial origin of observed transition lines [14]. The obtained correlation relations in both (XX^0, X^0) and (L4, X^0) configurations revealed that X^0, XX^0 and charged exciton emissions are originated from the same QD, otherwise normalized coincidence count will exceed 1/2.

The competing process observed in cross-correlation measurements is also reflected on steady-state luminescence intensity. Figure 3(a) shows PL intensities for each emission lines over a wide range of excitation powers, which were analyzed by applying a ladder model [15] composing 0-3 exciton states as depicted in Fig. 3(b). By solving the rate equations constructed from Fig. 3(b), steady-state PL intensities I_x and I_{xx} are expressed as:

$$I_X \propto \frac{p_1}{\tau_1} = \frac{1}{\tau_1}\left(1+\frac{1}{G\tau_1}+\frac{G\tau_2}{2}+\frac{G^2\tau_2\tau_3}{2}\right)^{-1} \quad , \qquad (1a)$$

$$I_{XX} \propto \frac{p_2}{\tau_2} = \frac{1}{\tau_2}\left(1+\frac{2}{G\tau_2}+\frac{2}{G^2\tau_1\tau_2}+G\tau_3\right)^{-1} \quad . \qquad (1b)$$

Notations are given in a figure caption of Fig. 3. Here, the biexciton excitation rate was assumed to be 1/2*G* by taking into account a spin-selective exciton capture for XX^0. For this analysis, lifetimes of X^0 (1.02 ns) and XX^0 (0.55 ns) were directly measured from Fig. 1(a) and a *p*-shell decay time was separately obtained to be ~0.3 ns (not shown). Resultant I_x and I_{xx} are given in the solid lines in Fig. 3(a). It is obvious that the present Ladder model fits well for the combined intensities of X^0+X* (solid squares) and XX^0+L2 (solid circles) instead of single emission line intensities. Since the two temporary competing neutral/ charged processes are averaged in the steady-state luminescence, it is likely that the combined intensities of X^0+X* and XX^0+XX* (XX*: charged biexciton) will obey the above model. Therefore, the observed L2 line can be possibly attributed to XX*, which implies negative binding energy with respect to X*.

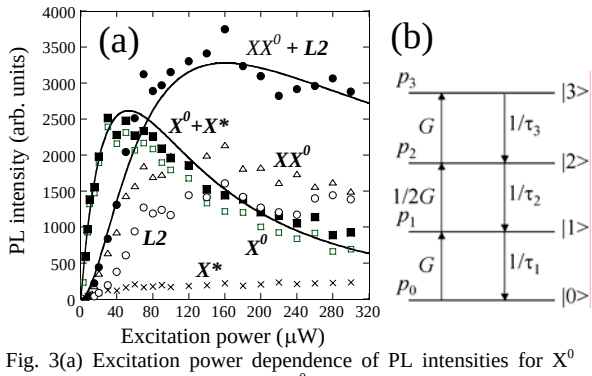

Fig. 3(a) Excitation power dependence of PL intensities for X^0 (open squares), X* (crosses), XX^0 (open triangles), and L2 lines (open circles). Summed intensities of X^0+ X* (closed circles) and XX^0 + L2 (closed squares) are also shown. (b) Schematic picture of 4-level Ladder model. *G* is an excitation rate, τ_i' is radiative lifetime of *i*-th state, and p_i is the probability of finding the *i*-th excitonic multiplex in the QD. Steady-state PL intensities for both excitonic and biexcitonic lines are shown in solid lines in (a). Excitation power of 38 μW corresponds to unit excitation rate.

Since specific emission line X^0 is filtered for avoiding multi-photon emission, the observed competing processes between neutral and charged exciton states can be a drawback the for high bit-rate SPE and also spoil the triggered photon emission nature. Thus, it is beneficial to understand the mechanism of charged exciton formation in QD. For this purpose, excitation energy dependence of PL spectrum was investigated under weak excitation power before the onset of XX^0 emission and results are shown in Fig. 4. Distinct contrast between two excitation energies was observed, *i.e.*, X^0 line is accompanied by charged exciton emission under WL excitation, while the charged exciton emission was almost absent under barrier excitation. Further study on this behavior is now under way, but one possible

interpretation for this observation is higher electron mobility in WL and a hindrance of hole capture into QD due to strain-induced potential barrier in a valence band [16]. For the barrier excitation, the potential barrier becomes less important and photogenerated electron-hole pair will be equally captured into QD, which will result in suppressed charged exciton formation. Therefore, it is suggested that observed X* and XX* are tentatively assigned to negatively charged exciton (X^-) and biexciton (XX^-), respectively. Redshift of X* line relative to the X^0 is also consistent with the present assignment [17].

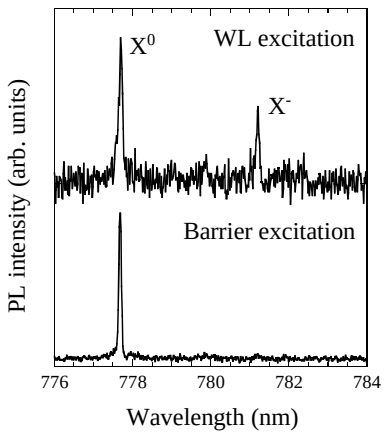

Fig. 4 PL spectra for WL excitation (upper panel) and barrier excitation (lower panel) under weak excitation condition before an onset of biexciton transition.

4. Conclusions

Triggered single photon generation from InAlAs QD is demonstrated for the first time. By employing the crossed photon correlation measurements, it is directly confirmed that the competing decay processes associated with neutral and charged systems are exclusively formed in the same QD.

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