Emission characteristics for a single CdSe quantum dot on an optical nanofiber at cryogenic temperatures

K Muhammed Shafi, Wei Luo, Ramachandrarao Yalla and Kohzo Hakuta
Center for Photonic Innovations, University of Electro-Communications, Chofu, Tokyo 182-8585, Japan
E-mail: shafi@cpi.uec.ac.jp

Abstract. We investigate the emission characteristics of a single CdSe quantum dot on an optical nanofiber at cryogenic temperatures from the viewpoint of quantum photonics. We show that the charged exciton (trion) of the quantum dot is a promising quantum emitter for both aspects of spectral and temporal characteristics. The nanofiber/quantum-dot system may give a promising work bench for the future quantum photonic network.

1. Introduction
In recent years, various quantum nanophotonic systems with quantum emitters have been developed in the context of single photon manipulation. A key point of such systems is to confine the light field modes to a tiny area around λ^2, where λ is the wavelength of the emitterd photons. In this view, one promising approach would be to use optical nanofibers (ONFs), tapered optical fibers with sub-wavelength waist diameters. In ONFs, guided modes at the nanofiber waist can be confined to an area with a size of wavelength. Various novel quantum optical processes have been demonstrated so far using hybrid systems of ONFs and quantum emitters [1].

Regarding quantum emitters, one promising candidate would be core/shell quantum dots (QDs) in colloidal solutions, which can realize wide tunability with high quantum efficiency (QE). Although it has been recognized that one weak point of such QDs is a rather long exciton decay time [2], the weak point can be lifted by using thick shell or bulky gradient-shell structures for the CdSe core/shell QDs. Using such QD structures at cryogenic temperatures, it is shown that the QDs are observed not only in neutral conditions, but also in charged conditions [3], where a charged exciton (trion) transition reveals a one order shorter radiative decay time than that for the neutral exciton transition.

So far, however, all above works have been carried out under microscopes for QDs cast on a glass plate. Such situation is far from real photonic applications. In this paper, we extend the work to a hybrid system of an ONF and a single thick shell CdSe QD, which is operated at cryogenic temperatures. This situation can be readily extended to photonic applications in fiber networks. By observing emitted photons through the fiber guided modes, we systematically investigate emission characteristics for one specific QD deposited on a nanofiber for exciton and trion transitions. We show that the trion at cryogenic temperatures would be a promising candidate as a quantum emitter for the ONF and QD hybrid system.
2. Experimental setup
ONFs were fabricated by tapering single mode optical fibers using a heat and pull technique. The nanofiber waist diameter was set to 320 nm so that fluorescence photons from an emitter on the ONF surface can be efficiently channeled into the fiber guided modes with an efficiency around 20% \[4\]. We used thick shell CdSe/ZnS core/shell QDs in toluene solution synthesized by NS Materials Inc. Quantum efficiency (QE) of the QD in solution was measured to be 85%. A single QD was deposited on an ONF using a needle-dispenser with the QD solution \[5\].

Regarding the cryogenic system, we used a custom designed optical cryostat with a pulse-tube refrigerator. The ONF with a single QD was installed in a sample chamber, which was fixed to a cold plate at the bottom of the cryostat. The QD on a nanofiber was cooled down to 3.7 K through buffer-gas cooling by filling the sample chamber with He-gas. Optical transmission of ONF did not show any drop by lowering the temperature to cryogenic temperatures.

Figure 1(a) illustrates a measurement scheme for the emission characteristics of a single QD on a nanofiber. The QD was excited perpendicularly to the nanofiber with a cw or pulsed laser at a wavelength of 532 nm. Emissions were measured through optical fibers. One side of the ONF was introduced to an optical multichannel analyzer (OMA) spectrometer with two gratings of 300 and 1,800 lines/mm. The other side of the ONF was used for photon counting/correlation measurements with two single-photon counting modules (SPCMs). The photon arrival times at the two SPCMs were recorded using a time tagging module (Pico-Harp 300) to obtain photon counting-rates and photon correlations. We measured the photon correlation function \(g_2(\tau)\) of the QD on the nanofiber to confirm the single QD emission. The measured \(g_2(\tau)\) is displayed in Fig. 1(b), revealing a \(g_2(0)\) value close to zero.

3. Results and discussion
Figure 2(a) displays the QD emission spectrum at 3.7 K measured at an excitation intensity of 30 W/cm\(^2\) with an integration time of 60 s. Well resolved four peaks are observed. The observed energy separation between peaks 1 and 2 was 15.2 meV (5.0 nm in wavelength), leading to the assignment of the two peaks as zero phonon lines (ZPLs) for neutral exciton and charged exciton (trion), respectively \[6\]. Spectral width of each peak was 1.3 nm FWHM. Peaks 3’ and 3 were the longitudinal optical phonon (LO-phonon) replica of the exciton and trion ZPL, respectively. The spectrum in the top and bottom of Fig. 2(a) are observed at a different time. It is observed that occurrence probability of the exciton and trion changes by the observation timing. For the top case, neutral condition is much more probable than the charged condition.

Since the spectral profile for the QD emission is governed by the spectral diffusion process, the emission spectrum was measured for shorter integration times to examine the intrinsic spectral width. Fig. 2(b) shows the spectrum observed with an integration time of 1 s at 3.7 K. Although S/N-ratio is not good, one can readily recognize a sharp peak at the trion ZPL wavelength. The observed width is 0.12 nm FWHM (~350 µeV), which is the spectral resolution limit for the
Figure 2. (a) Emission spectrum of single QD at 3.7 K with an integration time of 60 s. Peak 1 and 2 correspond to ZPLs for exciton and trion, respectively. Peak 3' and 3 are the LO-phonon replica of exciton and trion, respectively. Two spectra displayed in top and bottom panels were measured at different timings. (b) Emission spectrum at 3.7 K with an integration time of 1 s. (c) Emission spectrum at 3.7 K with an integration time of 270 s. (d) Total photon count rate measured at room temperature. (e) Total photon count rate measured at 3.7 K

We found that the occurrence probability reaches a stationary condition for integration time longer than 100 s. Fig. 2(c) shows a spectrum measured with an integration time of 270 s at 3.7 K. One can see exciton and trion ZPLs with corresponding LO-phonon replica. Spectral widths are broader than those observed with an integration time of 60 s. From the peak intensity ratio, we estimated the stationary occurrence probability for neutral and charged condition at 3.7 K to be 46% and 54%, respectively. Figs. 2(d, e) display photon count rate measured at the end of ONF for a period of 100 s at room temperature and at cryogenic temperature of 3.7 K, respectively. At room temperature, one can clearly see three steps in the count rate, and the three steps are marked by A, B, and C. Peaks A and B can be attributed to neutral exciton emission and trion emission respectively [7]. Based on the observed step behavior, we estimated the exciton QE to be close to 100% from a fact that the QE for the mixture of exciton and trion is 85%. By lowering the temperature below 50 K, the step behavior in the count rate disappeared and the count rate showed a single step behavior. Measured example at 3.7 K is displayed in Fig. 2(e). Note that the count rate at 3.7 K marked by D is equal to that of exciton at room temperature. It means that the QE of trion increased by lowering the temperature and reached a value close to 100%.

Figures 3(a-c) show temporal behaviours at 3.7 K obtained by measuring temporal correlations between the exciting laser pulse and emitted photons. Repetition rate of the laser pulse was 0.5 MHz and the measurement time was 200 s for each decay measurement. Measurements were carried out using two types of interference filters (IF). One filter was a narrow band filter with an FWHM of 2 nm which could resolve exciton and trion peaks separated by 5.0 nm by adjusting the filter tilt-angle. The other filter which had an FWHM of 10 nm was present OMA spectrometer. It means that the intrinsic spectral width for the QD is much narrower than the observed width.
Figure 3. Semi-log plot of temporal behaviors for a single QD on an ONF measured at 3.7 K. (a) Decay profile measured using a 10 nm IF filter. (b) Exciton decay profile measured using a 2 nm IF filter. (c) Trion decay profile measured using a 2 nm IF filter.

used for measuring emissions from both exciton and trion simultaneously.

Figure 3(a) displays a decay profile measured with the 10 nm FWHM IF filter. One can recognize multiple exponential decay curves. Figs. 3(b, c) display decay profiles measured for exciton and trion separately using the 2 nm FWHM IF filter, respectively. In Fig. 3(b), one can clearly see a double exponential behavior with a fast decay time of 3.2 ns and a slow decay time of 140 ns. The observed double exponential behavior can be well explained by a fine structure model of band edge exciton consisting of two states, termed as dark and bright states [2]. On the other hand, the trion decay profile in Fig. 3(c) shows clearly a single exponential decay process with a decay time of 11 ns. This means that the trion transition can effectively be treated as a simple two level transition.

4. Conclusion
We have investigated photoemission characteristics for a single CdSe QD on an ONF at cryogenic temperatures. From the observed results, we conclude that charged exciton (trion) is superior to the neutral exciton as a quantum emitter. The trion shows a simple two level transition and reveals a more than one order faster response time than the effective exciton response time. Regarding the QE value, although the trion QE is low at room temperature, it can be improved by lowering the temperature to a value close to 100%. Although the occurrence probability of trion is 54% for the present QD, the probability can be improved close to 100% by introducing a photo-assisted technique to the synthesis of QDs as reported in Ref 8. The ONF/QD system with trion may give a promising work bench for the future quantum photonic network.

Acknowledgements
The work was supported by the Japan Science and Technology Agency (JST) through the Strategic Innovation Program. Authors thank NS Materials Inc. for providing the QD samples.

References
[1] Nayak K P, Sadgrove M, Yalla R, Kien F L and Hakuta K 2018 J. Opt. 20 073001
[2] Biadala L, Louyer Y, Tamarat P and Lounis B 2009 Phys. Rev. Lett. 103 037404
[3] Fernée M J, Littleton B N and Dunlop H R 2009 ACS Nano 3 3762
[4] Yalla R, Kien F L, Morinaga M and Hakuta K 2012 Phys. Rev. Lett. 109 063602
[5] Yalla R, Nayak K P and Hakuta K 2012 Opt. Express 20 2932
[6] Louyer Y, Biadala L, Tamarat P and Lounis B 2010 Appl. Phys. Lett. 96 203111
[7] Gomez D E, Embden J, Mulvaney P, Fernée M J and Dunlop H R 2009 ACS Nano 3 2281
[8] Rinehart J D, Schimpf A M, Weaver A L, Cohn A W and Gamelin D R 2013 Am. Chem. Soc. 135 18782