

Photon Antibunching in a Cluster of Giant CdSe/CdS Nanocrystals

Bihu Lv,^{1†} Huichao Zhang,^{1,3†} Lipeng Wang,¹ Chunfeng Zhang,¹ Xiaoyong Wang,^{1*} Jiayu Zhang,^{2*} and Min Xiao^{1,4*}

¹*National Laboratory of Solid State Microstructures, School of Physics, and Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing 210093, China*

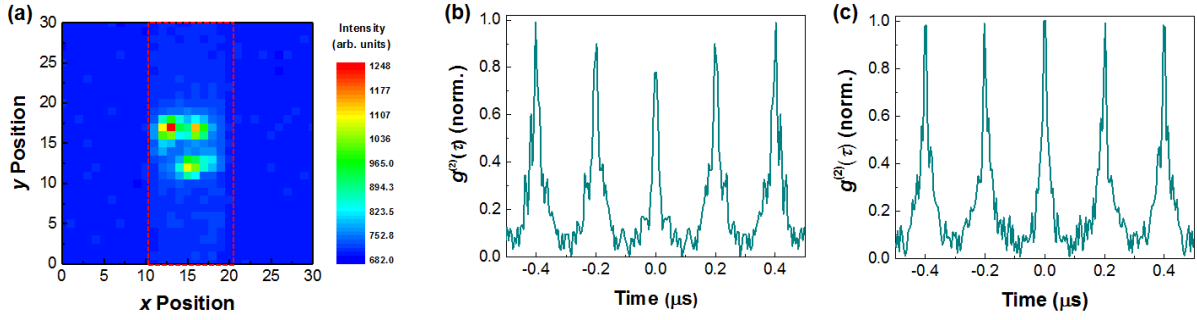
²*Advanced Photonics Center, School of Electronic Science and Engineering, Southeast University, Nanjing 210096, China*

³*College of Electronics and Information, Hangzhou Dianzi University, Xiasha Campus, Hangzhou 310018, China*

⁴*Department of Physics, University of Arkansas, Fayetteville, Arkansas 72701, USA*

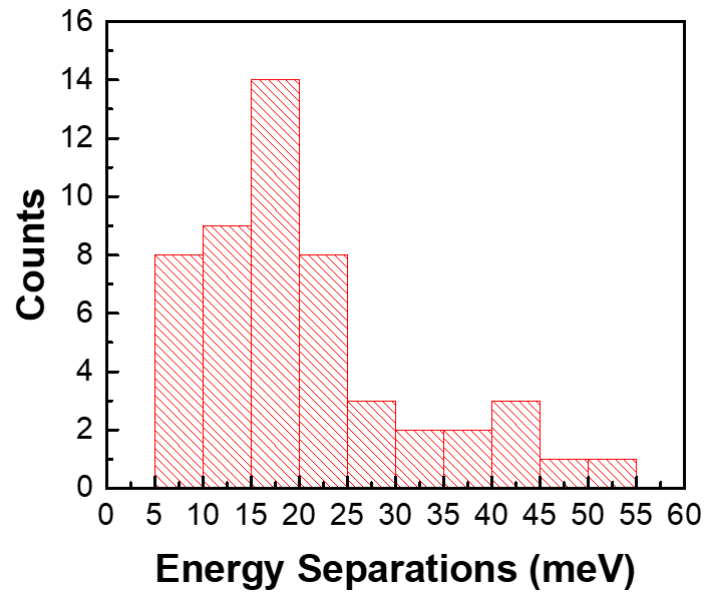
[†]These authors contributed equally to this work.

*Correspondence to X.W. (wxiaoyong@nju.edu.cn), J.Z. (jy Zhang@seu.edu.cn), or M.X. (mxiao@uark.edu).



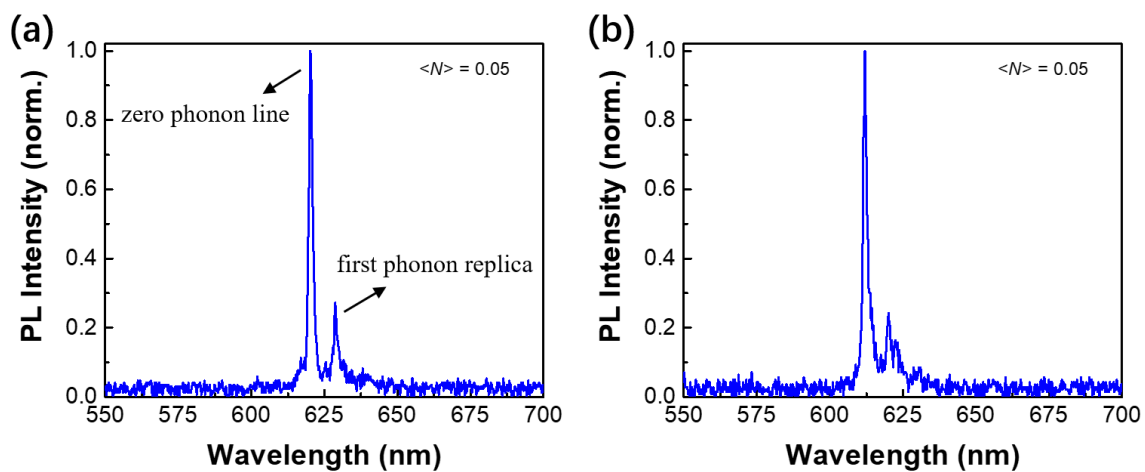
Supplementary Figure 1 | Photon antibunching measurements of several isolated gNCs.

(a) Photoluminescence (PL) image of three isolated CdSe/CdS gNCs excited at $\langle N \rangle = 0.1$ with an acquisition time of 1 s. The red dotted box marks the profile of the spectrometer entrance slit corresponding to a sample region of $3.2 \mu\text{m} \times 9.6 \mu\text{m}$. **(b)** Second-order photon autocorrelation measurement of these three gNCs with a $g^{(2)}(\tau)$ value of 0.78 at the zero-time delay. **(c)** Second-order photon autocorrelation measurement of >5 gNCs with a $g^{(2)}(\tau)$ value approaching one at the zero-time delay. The above optical measurements were performed at the cryogenic temperature of 4 K.

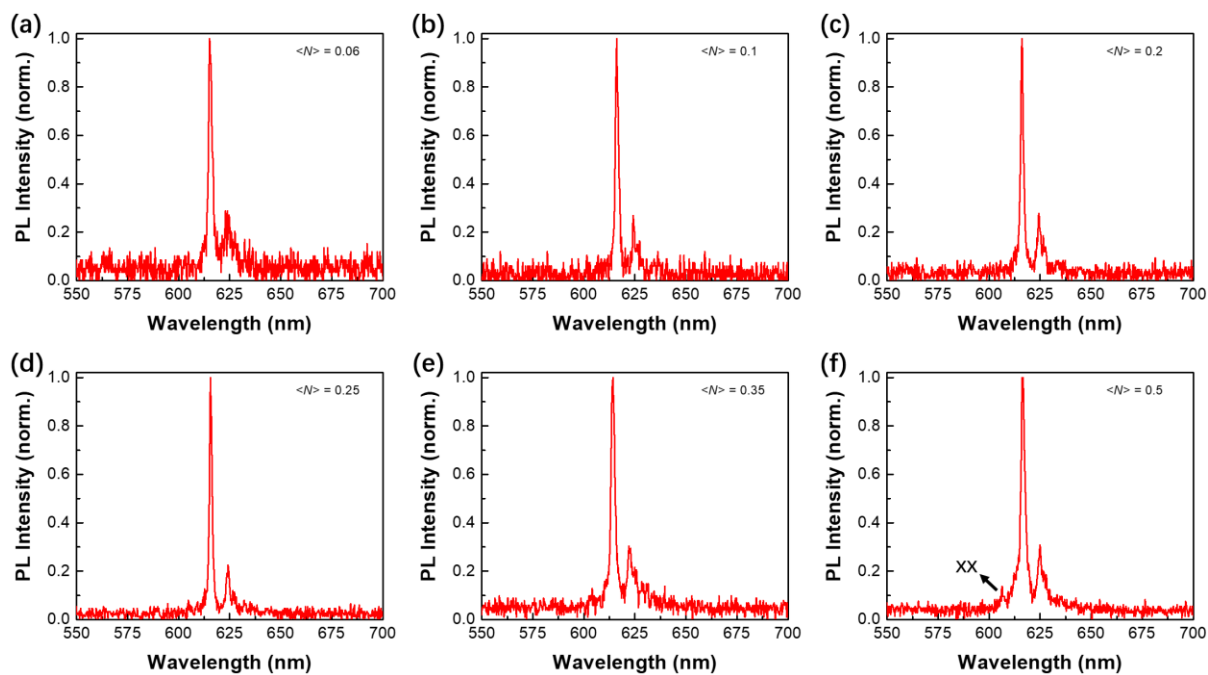


Supplementary Figure 2 | Energy separations of nearby PL peaks in single gNCCs.

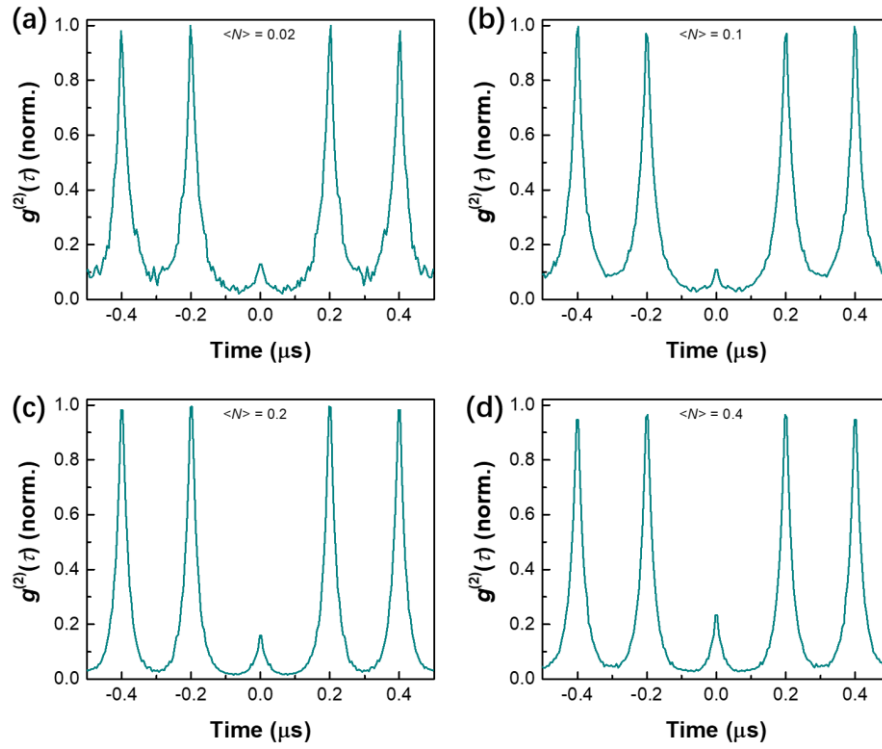
Statistical histogram for the energy separations between all the nearby peaks in the PL spectra of >30 single gNCCs excited at 4 K with $\langle N \rangle = 0.1$.



Supplementary Figure 3 | PL spectra of individual gNCs measured at 4 K. (a), (b) PL spectra measured with an integration time of 1 s for two representative gNCs excited at $\langle N \rangle = 0.05$ with the energy separations of 27.5 and 26.5 meV, respectively, between the zero phonon line and the first phonon replica.



Supplementary Figure 4 | PL spectra of an individual gNC measured at 4 K. PL spectra measured with an integration time of 1 s for a representative gNC excited at (a) $\langle N \rangle = 0.06$, (b) $\langle N \rangle = 0.1$, (c) $\langle N \rangle = 0.2$, (d) $\langle N \rangle = 0.25$, (e) $\langle N \rangle = 0.35$, and (f) $\langle N \rangle = 0.5$, respectively. It should be noted that a very weak peak appears on the blue side of the PL spectrum measured at $\langle N \rangle = 0.5$ and is attributed to the biexciton (XX) recombination with an emission energy 32.1 meV higher than that of the main peak from single excitons.



Supplementary Figure 5 | Photon antibunching of an individual gNCC measured at 4 K.

The laser excitation power corresponds to (a) $\langle N \rangle = 0.02$, (b) $\langle N \rangle = 0.1$, (c) $\langle N \rangle = 0.2$, and (d) $\langle N \rangle = 0.4$, respectively.