Resonantly Excited Exciton Dynamics and Saturation in Colloidal CdSe/ZnS Quantum Dots

J. H. Kim[1], B. J. Kim[1], S. M. Kim[1], Ho-Soon Yang[1], K. Kyhm*[2]
[1]Department of Physics, [2]Research Center for Dielectric and Advanced Matter Physics, Pusan National University, san 30, Kumjung-gu, Busan, 609-735, Korea
* correspondence: kskyhm@pusan.ac.kr

Recently chemically synthesized nanocrystal (NC) quantum dot(QD)s were introduced as a gain medium to realize quantum dot lasers for their size-tunability and low cost in comparison with epitaxial quantum dots[1]. Quantum confinement in three dimensions results in an enhancement in the density of electronic states, which enables a higher carrier concentration to contribute to the band-edge emission, and the spacing of the energy states increases as the dot size is reduced[2]. Additionally, the electron-hole binding is strongly enhanced as the dot size is comparable with the excitonic Bohr radius. Thus, excitons are expected to play an important role in the emission process. However, the radiative recombination efficiency is reduced with decreasing dot sizes due to surface trapping. Poor surface passivation of NC QDs which leads to large nonradiative carrier loss, and large size distributions hinder the achievement of strong stimulated emission. Thus, well-passivated surface and size uniformity are required in the synthesis process. A colloidal chemical process was known as a way to improve the quality of QDs, resulting in dense and small dots with size uniformity. In this work, high density colloidal CdSe QDs are prepared in a capillary to keep the uniform size distribution and also prevent agglomeration. It is known that overcoat treatment with ZnS improves the surface passivation of QDs, leading to narrower emission spectra for closely-packed solid NC QDs[3,4].

In order to investigate excitonic dynamics in CdSe/ZnS core-shell structures, we performed the degenerate pump-probe experiments under the resonant excitation, where the laser was tuned at exciton energy. As a preliminary work, absorption and photoluminescence spectra are compared; small Stoke shift and narrow photoluminescence spectrum confirm the surface passivation by ZnS overcoating. Normalized Differential Transmission shows two step decays. We conclude the initial fast decay is attributed to the Auger recombination while the later decay is typical exciton radiative recombination process. The appearance of the fast decay emerges at critical excitation power, and the fast decay also has a dependence on the excitation power. This fact indicates the density dependence of the fast decay is most likely related to the Auger process. On the other hand, this critical excitation power coincides with the saturation power of the normalized differential transmission, which was measured at peak rise time. As excitions are resonantly excited, free carriers are prevented so the exciton-Mott transition density can be estimated from the experiments. We found that the saturation density corresponds to the Mott-like transition from excitons to electron-hole plasma, and the Auger recombination is enhanced significantly at this critical transition density.

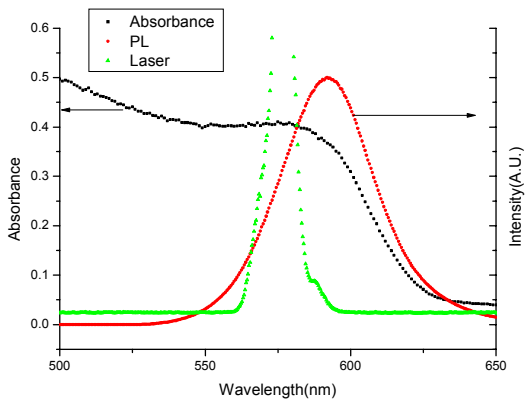

Fig. 1 Absorption and photoluminescence spectrum.

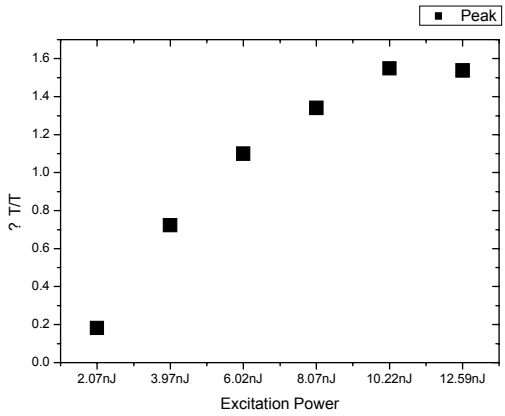

Fig. 2 Saturation of Normalized Differential Transmission for excitation power.

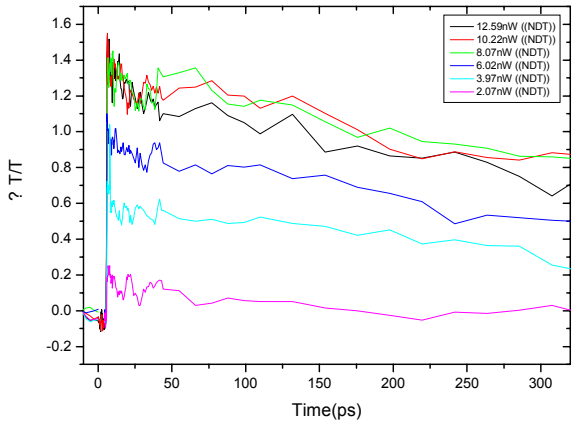

Fig. 3 Differential Transmission as a function of delay time for various excitation powers.

1. L. Pavesi, L. Dal Negro, C. Mazzoleni, G. Franzo, and F. Priolo, Nature 408, 440 (2000)
2. V. I. Klomov et al., Science 290, 314 (2000)
3. J. Jack Li et al., J. Am. Chem. Soc. 125, 12567 (2003)
4. B. O. Dabbousi et al., J. Phys. Chem. 101, 9463 (1997)