Optical Inhomogeneity from 2D Spectra vs. Static Size Dispersion in an Ensemble of PbSe Nanocrystals

Samuel D. Park, Dmitry Baranov, Jisu Ryu, and David M. Jonas*

Department of Chemistry and Biochemistry, University of Colorado, 215 UCB, Boulder, CO 80309 USA, *David.Jonas@colorado.edu

The optical inhomogeneity of a PbSe QD sample is determined using 2DFT spectroscopy with the nodal line slope method and compared to size dispersions measured with TEM. Comparison between the two suggests that either TEM overestimates the size dispersion or the optical inhomogeneity does not directly reflect the size dispersion.

Size dispersion is a fundamental property of an ensemble of quantum dots (QDs), and it is an important parameter in understanding the carrier dynamics because optical inhomogeneity broadens linewidths through the size dependent bandgap. A standard method for quantifying the size dispersions of a QD sample is by transmission electron microscopy (TEM) [1]. In this work, we utilize 2DFT spectroscopy in the short-wave infrared region to determine the ensemble optical inhomogeneity (Fig. 1) in a sample of colloidal PbSe QDs and compare it to the static size dispersion obtained from TEM images of the same sample.

The 2D spectra at a waiting time of T = 1 ps of PbSe QDs contain a combination of signals that create a nodal line, in which the nodal line slope uniquely depends on the degree of inhomogeneity. Using the nodal line slope method [2], we determine the optical inhomogeneity upper bound to be 85 ± 5 meV (FWHM). Static size dispersions were obtained by analyzing bright field TEM images and dark field scanning TEM images collected at the Colorado School of Mines and CAMCOR user facility at the University of Oregon, respectively, following published procedure [3], and were found to agree between the two imaging modes and facilities. The apparent size dispersion was converted to energy bandgap dispersion by using literature sizing curves [3], giving 110-145 meV (FWHM), which significantly exceeds the optical inhomogeneity determined from 2D spectra. Based on the comparison between 2D spectra and TEM, it appears that either TEM overestimates the size dispersion or the optical inhomogeneity does not measure or directly reflect the size dispersion.

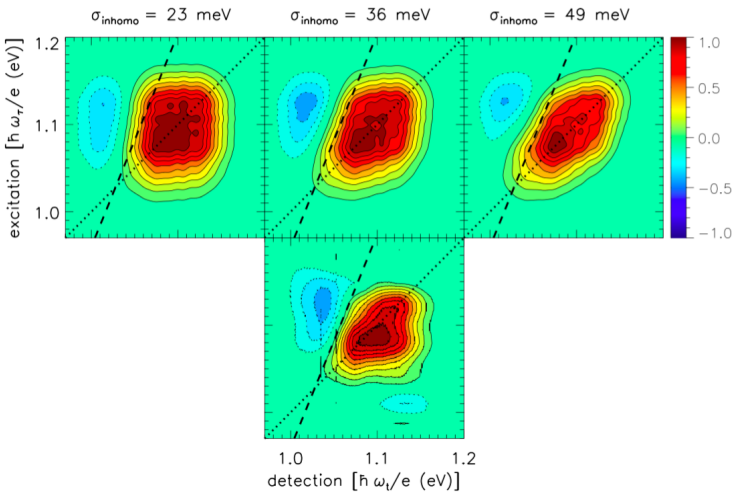

Fig.1 Simulated (top row) and experimental (bottom) 2D spectra of PbSe QDs at T = 1 ps. The top row shows the simulated results for the inhomogeneous linewidth standard deviations labeled on the top of the spectra. The dotted line shows the diagonal and the dashed line shows the slope of the nodal line from the experimental 2D spectrum for comparison.

[1] W. D. Pyrz and D. J. Buttrey, Langmuir **24**, 11350-11360 (2008)
[2] S. D. Moran *et al*., PNAS **109**(9), 3329-3334
[3] D. Segets et al, ACS Nano, **6**(10), 9021-9032 (2012)