Abstract Submitted
for the MAR06 Meeting of
The American Physical Society

Studying Photoluminescence Dynamics of Single Quantum Dots Photon by Photon[1] HAW YANG, KAI ZHANG, AIHUA FU, PAUL ALIVISATOS, UC Berkeley, CARL HAYDEN, Sandia National Laboratory — Colloidal semiconductor nanocrystals, or quantum dots (QDs), have been the focus of much research effort in the past decade. The development of these colloidal dots has allowed the concepts of quantum confinement and dimensional control of electronic and optical properties to find entirely new areas of application, for instance in fluorescent labeling of biological specimens. At the single-particle level, however, colloidal QDs exhibit surprisingly complicated time-dependent behavior in their photoluminescence (PL) characteristics. The PL dynamics of the biologically compatible CdSe/ZnS/streptavidin quantum dots were studied using time-resolved single-molecule spectroscopy. Statistical tests of the photon-counting data suggested that the simple "on/off" discrete state model is inconsistent with experimental results. Instead, a continuous emission state distribution model was found to be more appropriate. Autocorrelation analysis of lifetime and intensity fluctuations showed a nonlinear correlation between them. These results were consistent with the model that charged quantum dots were also emissive, and that time-dependent charge migration gave rise to the observed photo-luminescence dynamics.

[1]Work supported by the US Department of Energy

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Date submitted: 29 Nov 2005 Electronic form version 1.4