Abstract Submitted
for the MAR05 Meeting of
The American Physical Society

The Structure of Excited-State Transitions in Individual Nanocrystals Probed by Photoluminescence Excitation Spectroscopy HAN HTOON, P.J. COX, V.I. KLIMOV, Chemistry Division, Los Alamos National Laboratory, Los Alamos, NM 87545 — We study the structure of excited-state transitions in CdSe nanocrystals (NCs) using our newly developed, single-NC, photoluminescence excitation (PLE) technique that mitigates the problem of blinking and spectral wandering of NC emission. The low- temperature (4K) PLE spectra of individual NCs show a few sharp band-edge peaks with 3 to 4 meV linewidths. The higher energy transitions, however, are not manifested as narrow peaks but they rather merge into a dense quasi-continuum. These spectroscopic observations can be rationalized based on results of intraband hole relaxation studies. Specifically, these studies indicate that the energy relaxation rate is very large (1 – 2 eV/ps) at high spectral energies, leading to significant lifetime broadening of optical transitions that merge into a structureless quasi-continuum. The relaxation rate sharply drops near the band-edge, which is consistent with our observations of the reduced density of states at low spectral energies. Overall, our studies indicate that, while the idealized description of NC excited-state transitions in terms of sharp atomic-like resonances may be applicable at near-band-edge spectral energies, this description is invalid at high spectral energies where optical transitions are significantly broadened because of ultrafast intraband relaxation.

Han Htoon
Chemistry Division, Los Alamos National Laboratory
Los Alamos, NM 87545

Date submitted: 30 Nov 2004 Electronic form version 1.4