

Abstract Submitted
for the MAR11 Meeting of
The American Physical Society

Copper-doped core-shell ZnSe/CdSe nanocrystals with efficient and widely tunable photoluminescence RANJANI VISWANATHA, SERGIO BROVELLI, VICTOR I. KLIMOV, Chemistry Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545 — We report synthesis and spectroscopic studies of Cu-doped ZnSe/CdSe nanocrystals (NCs) with a wide range of shell thicknesses. Incorporation of copper ions into the NCs introduces an atomic-like state within the NC band gap. This results in a three-level system in which emission occurs due to the transition coupling the NC lowest-energy conduction-band level to the localized hole state associated with the Cu ions. Cu is incorporated into the NC core while the shell remains nominally undoped, which allows us to manipulate the shell thickness (and thus emission color) without affecting the overall level of NC doping. We demonstrate a wide-range spectral tunability of photoluminescence (PL) (from 3.1 eV to 1.25 eV), and “giant” Stokes shifts (~ 0.8 eV), which reduce emission losses due to reabsorption. We show that hole trapping at the Cu sites occurs primarily from the top of the valence band (i.e., no hot-hole transfer processes are detected) and that the main nonradiative mechanism is electron trapping at surface defects, which can be suppressed at cryogenic temperatures, resulting in PL quantum yields of $\sim 40\%$. High intrinsic emission efficiencies, wide-spectral tunability, and a large Stokes shift make these novel NCs attractive candidates for radiation detection, light-emitting diodes, and lasers.

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Date submitted: 23 Nov 2010

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