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Toward Single-Exciton Lasing through Engineered Exciton-Exciton Interactions JAGJIT NANDA, SERGEI IVANOV, HAN HTOON, ILYA BEZEL, ANDREI PIRYANTINSKI, SERGEI TRETIAK, VICTOR KLIMOV, Los Alamos National Laboratory, Los Alamos, NM-87545 — We synthesize and study “inverted,” core-shell nanocrystals (NCs), in which a core of a wide-gap semiconductor (ZnSe) is overcoated with a shell of a narrower gap material (CdSe). Compared to monocomponent or traditional, “noninverted” hetero-NCs, these novel, “inverted” structures provide new capabilities for controlling their functionalities via a direct control of the spatial distribution of electron and hole wavefunctions. Specifically, by increasing the thickness of the NC shell (for a fixed NC core size), one can continuously tune the carrier localization regime from type-I for thin shells to type-II for intermediate shells and finally back to type-I for thick shells. We show that these heterostructures can be used to significantly increase absorption cross sections and simultaneously decrease the efficiency of Auger recombination compared to monocomponent NCs emitting at the same wavelength. Furthermore, appropriately designed hetero-NCs can exhibit repulsive exciton-exciton interactions that lead to reduced excited-state absorption associated with singly-excited NCs. This effect leads to reduced optical-gain thresholds and can potentially allow lasing in the single-exciton regime, for which Auger recombination is inactive. We use these hetero-NCs to demonstrate efficient light amplification that is tunable across a “difficult” range of green and blue colors.

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