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Probing carrier separation in a novel series of infrared type-II nanoheterostructures JEFFREY M. PIETRYGA, DOH C. LEE, ISTVAN ROBEL, VICTOR I. KLIMOV, Los Alamos National Laboratory — Three-dimensional confinement of carriers within semiconductor nanocrystals gives rise to novel characteristics such as size-controlled energy gaps, but also increased Coulomb interactions between charge carriers that can hamper spatial separation of electrons and holes, such as in photovoltaic devices. One approach to efficient "splitting" of photogenerated excitons is through introduction of appropriate energy gradients within a heterostructured nanocrystal. Specifically, a type-II staggered alignment of electronic states can provide nearly complete separation of electrons and holes between different spatial regions of a nanoheterostructure. In this work, we use static and transient photoluminescence spectroscopies to examine the evolution of type-II behavior in PbSe/CdSe/CdS core/shell/shell heterostructured nanocrystals. We observe a marked red shift and a dramatic increase in radiative lifetimes (to >10 μs) with increasing CdS shell thickness, indicative of type-II carrier separation, at effective band gaps of 0.95 eV, the narrowest band gap of any such system to date. We will compare these measurement to those of a homologous "tetrapod" system that exhibits surprisingly high emission quantum yields, to further elucidate the effect of structure on carrier separation.

Istvan Robel
Los Alamos National Laboratory

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