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Ultrafast Cathodoluminesence Studies of Colloidal Nanocrystals
LAZARO PADILHA, WAN KI BAE, VICTOR KLIMOV, JEFFREY PIETRYGA, Los Alamos National Laboratory, RICHARD SCHALLER, Argonne National Laboratory, Northwestern University — Despite possessing numerous desirable properties, including excellent photostability, high stopping-power, and fast, efficient fluorescence, semiconductor nanocrystal quantum dots are only mediocre gamma-ray scintillator materials. Efforts to improve performance are forestalled by a lack of understanding of how those materials respond to high energy radiation, which in turn comes from a lack of appropriate ultrafast tools for examining the relaxation of gamma-excited quantum dots. To this end, we have developed a 20keV as a surrogate for spontaneous gamma irradiation. We apply this technique to study the time-resolved response of thin films of CdSe/ZnS core/shell quantum dots. Energetic excitation produces a variety of excited states that can be separately resolved by consideration of their established energies and relaxation dynamics. We analyze the relative branching ratios of single excitonic, multi-excitonic and charged excitonic states to derive surprising conclusions regarding the physics of highly-excited quantum dots, as well as the probable source of poor gamma-scinitillating performance.

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