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Breakdown of Volume Scaling in Auger Recombination in CdSe/CdS Heteronanocrystals: The Role of the Core-Shell Interface SERGIO BROVELLI, FLORENCIO GARCÍA-SANTAMARÍA, RANJANI VISWANATHA, HAN HTOON, SCOTT CROOKER, VICTOR I. KLIMOV — Spatial confinement of electronic excitations in semiconductor nanocrystals (NCs) results in a significant enhancement in nonradiative Auger recombination (AR) of multiexcitons, which is detrimental to promising NC lasing applications. In standard NCs, AR times scale linearly with NC volume. We investigate multiexciton dynamics in NCs composed of CdSe cores and CdS shells of tunable thickness. Thicker shells dramatically reduce AR, particularly during initial shell growth, which cannot be explained by traditional volume scaling alone. Rather, low-temperature fluorescence-line-narrowing studies strongly suggest that suppressed AR derives primarily from the formation of an alloy layer at the CdSe/CdS interface, and a corresponding "smoothing" of the confinement potential (CP). These findings support the recent theory, which predicts that the change from abrupt to smoothly-varying CPs reduces the high-spatial-frequency Fourier components of the exciton wave function, thereby minimizing overlap with the high-energy states involved in nonradiative Auger decay. Our results highlight the importance of NC interfacial structure in the AR process in zero-dimensional NCs and provide general guidelines for the design of new nanostructures with suppressed AR for future lasing applications.

Sergio Brovelli
Chemistry Division, Los Alamos National Laboratory,
Los Alamos, New Mexico, USA

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