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New Developments in Nanocrystal Lasing: Type-II Nanostructures and "Giant" Quantum Dots[1]

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Nanocrystal (NC) quantum dots show high photoluminescence quantum yields and size-dependent emission colors tunable through the quantum-confinement effect. Despite their favorable light-emitting properties, NCs are difficult to use in optical amplification. Because of almost exact balance between absorption and stimulated emission in nanoparticles excited with single excitons, optical gain can only occur due to NCs that contain at least two excitons. A resulting complication is fast optical-gain decay induced by nonradiative Auger recombination, a process in which one exciton recombines by transferring its energy to another. In this talk, I will discuss two approaches for resolving the problem of ultrafast Auger recombination in NCs. In one approach, we utilize core/shell hetero-NCs engineered in such a way as to spatially separate electrons and holes between the core and the shell (type-II heterostructures). The resulting imbalance between negative and positive charges produces a strong local electric field, which induces a large, $\sim$100 meV transient Stark shift of the absorption spectrum with respect to the luminescence band. This effect breaks the exact balance between absorption and stimulated emission and allows us to demonstrate optical amplification in the single-exciton regime when Auger recombination is simply inactive. In another approach, we use recently developed "giant" dots that comprise a small emitting CdSe core overcoated with a thick shell (up to 20 monolayers) of a wider-gap CdS. These nanostructures produce a peculiar quasi-type-II localization regime, which develops as a result of a significant difference in effective volumes of the electron and hole wave functions. These structures show greatly suppressed Auger recombination, which allows us to realize broadband optical gain (extends over $>$ 400 meV) due to multiexcitons of various orders with an excitation threshold, which is at least a thousand times lower than in regular CdSe NCs.

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