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Spectral and Dynamical Characterization of Multiexcitons in CdSe/CdS/CdZnS Colloidal Quantum Dots

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The time-resolved photoluminescence triggered by a ~30 ps long exciting pulse centered at the time 154 ps was investigated. The use of long pulses allows one not only to trace the decay of multi-exciton complexes, but also to study the process of their formation and, thus, to give a more complete picture of the physical processes occurring in the system. In addition, we excite electron-hole pairs by a laser directly near the p-states.

We obtained a series of transient photoluminescence spectra extracted at different delays relative to the center of the exciting laser pulse using a 50 ps integration window. The asymmetric fitting function can be presented as a sum of three Gaussian components centered at $\varepsilon = \varepsilon_i$ ($i = x, xx, xxx$) and some background constant. We associate the broad peak in each transient PL spectrum described by the Gaussian function with a maximum at $\varepsilon_{xxx} = 2.1$ eV with the radiative recombination of e-h pairs in p-states. It quickly disappears, but since its width is large. The high peak at $\varepsilon_x = 1.98$ eV we associate it with the decay of single excitons. And, finally, we explain the peak at $\varepsilon_{xx} = 1.92$ eV, which first arises and then disappears, as the decay of biexcitons. This peak is relatively low and is strongly overlapped by the other peaks. Comparing the spectral dependences we see that the parameters of the fitting function change with time. To find out how this occurs, let us study the temporal dependences of the amplitudes of Gaussian functions amplitudes of Gaussian functions and their widths.