Density of states and photoluminescence spectra in the dense arrays of CdSe/ZnSe quantum dots with Gaussian potential profile.

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We report on the study of dense arrays of ZnCdSe quantum dots (QDs) with the QD density of $10^{11}$-$10^{12}$ cm$^{-2}$ formed by molecular beam epitaxy by inserting a CdSe layer into a ZnSe matrix. The nominal thickness of CdSe layer was varied from 1.4 to 2.7 monolayers. Transmission electron microscopy (TEM) studies established that the lateral sizes of QDs are distributed within the range 2-12 nm with the maximum at 2-4 nm. The photoluminescence (PL) spectroscopy studies of the samples revealed a narrow PL line (FWHM~50 meV), which was red-shifted with increasing the CdSe amount. Such a narrow PL line width contradicts the huge size distribution of QDs, which should result in a much broader emission band, assuming all dots to be emitting. PL excitation (PLE) spectra of the arrays contain two pronounced peaks separated by the 65 - 115 meV gaps. To explain these puzzles we have performed firstly the modeling of the density of ground states of excitons, $I_{DOS(1)}$, in the QDs of different sizes, belonging to these arrays. The TEM histograms of the QD size distributions were used to find the spectra of this density of states. MBE grown QDs possess the maximal Cd content in the center; it is gradually decreased towards periphery. Spatial potential for electrons and holes in such QDs can be well approximated by the potential of Gaussian type $V_{G}(r) = V_{off}(1 - \exp[-r^2/a^2])$, where $r$ is the distance from the center of the QD, $V_{off}$ is the conduction band offset for the electrons or the valence band offset for the holes; characteristic size $a$ is of the order of the QD radius. The energies of both electron and hole levels found numerically and by the variational method turn off to be well consistent. The Coulomb interaction between carriers was estimated within the framework of the first order perturbation theory, as well as the influence of QD anisotropy. For observed QD size distributions, the modeling results in the peak of the excitonic state density located in the region of 2.6.-2.7 eV. This position is considerably higher than the PL peak energy; however, it matches well with the higher-energy peak in the PLE spectra. A possible explanation of these findings is that the small dots, determining this maximum, do not emit by themselves, but transfer the excitation to the radiating dots of larger sizes. Using available data on PL kinetics in such QDs we have performed modeling of the modified density of states, $I_{DOS(2)}$, taking into account the energy transfer process between the dots. This modeling has shown that the optical spectra can be reasonably simulated assuming the fast resonant energy transfer from the ground energy levels of the small QDs towards the excited levels of the large QDs, with consequent energy relaxation of the excitation to their ground levels. The contribution of non-resonant energy transfer involving LO phonons, which can be essential in some cases, is discussed as well.

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