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Impact of surface charge on spectrally diffusive photoluminescence in GaAs quantum dots grown by droplet epitaxy

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Introduction Various single quantum dot (QD) spectroscopy studies have confirmed significant line broadening in the photoluminescence (PL) spectra. The line broadening mechanism is commonly attributed to spectral diffusion, however, we still lack a global understanding of the microscopic mechanism of spectral diffusion. In this study, we experimentally analyze the dependence of QD morphology on environment-mediated spectral broadening. The use of QDs with a controlled height variation and a fixed lateral size enables us to attribute spectral diffusion to surface charge fluctuations [1].

Experimental GaAs QDs were self-assembly grown in $Al_{0.3}Ga_{0.7}As$ by droplet epitaxy on semi-insulating GaAs (100) substrates [2]. After the growth of a 100 nm $Al_{0.3}Ga_{0.7}As$ layer, different amounts of gallium with 1.5, 2, 3, 5, 7.5, or 10 monolayers (ML) were supplied with 0.5 ML/s at 200 °C. Then, an As_4 flux was supplied with 2.5×10^{-4} Torr at 200 °C. Notably, when the amount gallium supply was increased from 1.5 to 10 ML, the dot height increased from 2.3 (±0.5) to 24 (±5) nm, i.e., by a factor of ten, but the base size increased only by as factor of less than two. After the dots were grown, the samples were annealed at 400 °C, and capped by a 20 nm $Al_{0.3}Ga_{0.7}As$ and a 30 nm $Al_{0.3}Ga_{0.7}As$ at 400 °C and 580 °C, respectively. Then, the samples were capped by a 10 nm GaAs layer. The dot morphology was investigated by AFM and cross sectional STM, and the optical properties were analyzed by micro PL.

Results and Discussion Figure 1(a) shows PL spectra detected using a micro objective setup. There are sharp spectral lines at high energies, and relatively broad peaks at low energies. Similar features were also observed in polar nitride QDs [3]. Figure 1(b) compares the measured linewidths (gray points) with the model results (solid lines) as a function of emission energy. In the model we calculated Stark coefficients of GaAs dots with different height, and the magnitude of electric field fluctuation associated with surface charge fluctuation by a Monte Carlo method. The measured dependence is well simulated when we assume a charge density on the order of 10^{11} cm^{-2}, which is consistent with the surface state density of GaAs(100) reported in literature [4].

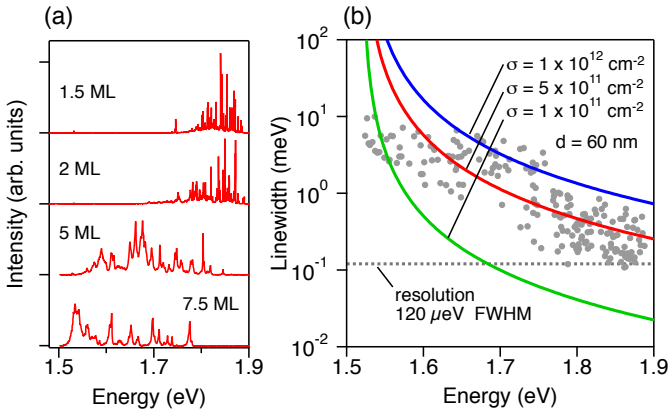

Figure 1(a) PL spectra detected using a micro objective setup. (b) Comparison between measured linewidths (gray points) and the calculated energy fluctuations due to randomly positioned surface charges with different density (solid lines).

References

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