Photoluminescence Efficiency of Germanium Dots Self-Assembled on Oxides

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Carrier localization methods in indirect bandgap materials have led to significantly stronger photoluminescence (PL) from Si/Ge nanostructures [1–3]. Ge nanocrystals generally display PL at ~2 eV, independent of dot size, as well as near infrared PL, which can be size dependent from quantum confinement effects [4, 5]. Under the proviso that the existing tight-binding (TB) [5] and effective mass (EM) [6] theoretical models provide a good description of the Ge dot energy gap versus dot diameter, this work investigates the effect of nanoparticle size and the size distribution on the near infrared PL spectrum obtained from self-assembled Ge dots grown on a thin layer of TiO_2 or SiO_2 on Si.

The 20 nm thick porous TiO_2 films [7] were deposited on standard 5 nm thick SiO_2 thermal oxide films on silicon substrates. The Ge dots were produced on the TiO_2 or SiO_2 films as follows [8]: first, several monolayers of Ge were deposited on the oxide surface at room temperature, and then the sample was annealed while still under vacuum in the deposition chamber at a temperature between 450 and 700 °C for ~30 min thus forming the Ge dots without oxide contamination. The Ge dots grown on SiO_2 were then capped with a 15-nm thick layer of amorphous Si in situ. Dot size distributions were obtained from atomic force microscopy (AFM) and transmission electron microscopy. The PL spectra were measured at 5 K using a Bomem DA3 FTIR spectrometer and excited with ~20 mW of 457.9 nm argon laser light.

For the as-grown samples, the dot PL emission occupies a wide near-infrared band between 0.8 and 1 eV [8]. In Fig. 1 we show the calculated PL efficiency versus dot size for four samples. These curves have been obtained in three steps [8]. Firstly, the PL spectrum was converted to an intensity plot versus dot diameter rather than energy by taking the PL emission from each dot to occur at the dot bandgap calculated using the TB or EM model. Secondly, a numerical form for the physical size distribution of that sample was obtained by performing a least-squares fit of a Gaussian to the dot size distribution measured by AFM. Finally, the PL efficiency versus dot size was calculated using the fitted Gaussian dot size distribution to normalize the PL intensity distribution obtained in the first step. The results obtained using the EM model are very similar to those shown in Fig. 1. Although the absolute intensities of the PL from the samples vary, the calculated curves are all well fitted by straight lines on a log-log plot, with essentially the same slope for all samples and demonstrating that under weak confinement there is a universal power-law increase in PL efficiency with decreasing dot size. In Fig. 2, the size density was calculated by taking the ratio of each sample's PL spectrum with the PL emission efficiency obtained from sample 163. Also shown are the size distribution histograms obtained from AFM data. The PL data predicts very well the small-diameter onset of the distribution (the PL-derived distribution stops above ~20 nm due to the detector cutoff in the PL apparatus).

In summary, an analysis of the PL response with respect to theoretical predictions of the bandgap versus dot size allowed an assessment of the size dependence of the PL quantum efficiency in Ge dots. The reverse process was applied to successfully predict the dot size distribution for independent samples from its PL alone.

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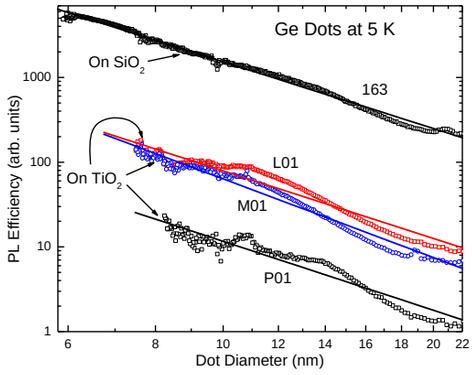

Figure 1. Variation of PL efficiency with Ge dot diameter within the TB model for samples prepared on SiO_2 and TiO_2 substrates. The line shows a linear fit to the curve.

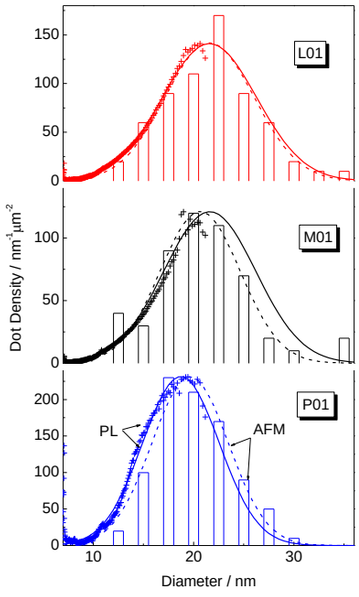

Figure 2. PL-derived dot densities (crosses) versus dot diameter (Gaussian fits – solid lines) for samples grown on TiO_2 compared with the Gaussian distributions (dashed lines) fitted to the AFM data (columns).