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Photoluminescence Study on Coarsening of Self-Assembled InAlAs Quantum Dots on GaAs (001)

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Red-emission at ~640 nm from self-assembled $In_{0.55}Al_{0.45}As/Al_{0.5}Ga_{0.5}As$ quantum dots grown on GaAs substrate by molecular beam epitaxy (MBE) has been demonstrated. We obtained a double-peak structure of photoluminescence (PL) spectra from quantum dots. An atomic force micrograph (AFM) image for uncapped sample also shows a bimodal distribution of dot sizes. From the temperature and excitation intensity dependence of PL spectra, we found that the double-peak structure of PL spectra from quantum dots was strongly correlated to the two predominant quantum dot families. Taking into account quantum-size effect on the peak energy, we propose that the high (low) energy peak results from a smaller (larger) dot family, and this result is identical with the statistical distribution of dot lateral size from the AFM image.

Key words: Bimodal distribution, photoluminescence (PL), quantum-size effect

INTRODUCTION

Recent progress of epitaxial growth in the coherent island Stranski-Krastanow (SK) mode growth allowed the fabrication of nanometerscale quantum dots, such quantum dots were formed directly by molecular beam epitaxy (MBE) in highly strained semiconductor materials.[1–4] The research on semiconductor nanostructures with size-dependent optical and electronic properties is motivated by fundamental physics and potential applications, which include quantum-dot lasers and high-speed nonlinear optical switches.[5] In particular, AlInAs/AIGaAs material should be permitted the realization of red-emitting semiconductor quantum dots lasers due to having a larger band gap.[6] For most III-V materials system studied, emission from self-assembled quantum dots was in the infrared (IR), radiative recombination in the red part of the visible spectrum was a few reported.

In this paper, we report the $In_{0.55}Al_{0.45}As/Al_{0.5}Ga_{0.5}As$ quantum dots emitting around ~ 1.941 ev (640 nm), grown by MBE. A bimodal distribution of quantum dot sizes has been demonstrated by atomic force microscopy (AFM), the double-peak structure of photoluminescence (PL) spectrum of quantum dots was observed. From the temperature and excitation intensity dependence of PL spectra, we found that the double-peak of PL spectrum was strongly correlated to the bimodal size distribution of quantum dots.

DISCUSSION

The samples were grown with SK growth mode on semi-insulating (001) GaAs substrate in a Riber 32P MBE system. An As_4 partial pressure of 6.5×10^{-6} Torr was used during the growth. The substrate temperature (T_s) was measured by a thermocouple which was calibrated using the substrate surface oxide desorption temperature (580°C). The substrate temperature was kept at 600°C, except for the growth of $Al_{0.5}Ga_{0.5}As$ where the temperature was up to 700°C. A sample structure used in this study consists of the following layers: a 200 nm thick undoped GaAs buffer, followed by a 80 nm $Al_{0.5}Ga_{0.5}As$ cladding layer, nominal 10 ML $In_{0.55}Al_{0.45}As$ layer, and finally 75 nm $Al_{0.5}Ga_{0.5}As$ and 5 nm GaAs capping layer. The gauge of forming 3D island was monitored by the change of reflection high energy electron diffraction (RHEED) pattern from streak to spotty during growing $In_{0.55}Al_{0.45}As$

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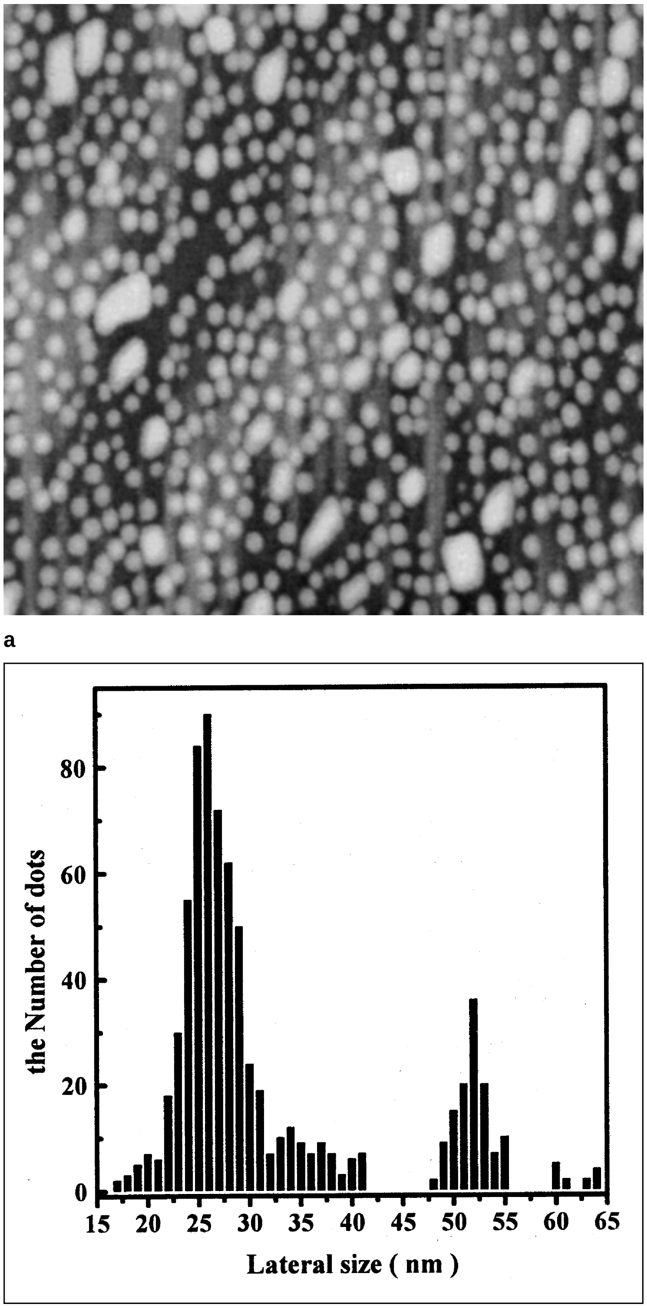


Fig. 1. AFM image of the uncapped nominal 10 ML quantum dots sample. (a) The size of AFM scans was 1 × 1 μm². (b) Histograms of the dots lateral size was created from nearby 1 × 1 μm² AFM images.

layer.[7] A Nanoscope III AFM was used *ex-situ to* investigate the dot lateral size distribution of an uncapped 10 ML sample grown at a same growth condition.

Figure 1 shows the morphology of the uncapped sample with nominal 10 ML $In_{0.55}Al_{0.45}As$ by AFM. A statistical lateral size distribution of quantum dots from the image have been obtained with the use of a public domain image processing software[8] (Scion Image program). Non-regular dots were substituted for a circular, the diameter of circular was estimated by an average diameter of dot on a few directions. The histogram of quantum dots lateral size was created from 1 × 1 μm² AFM image. There are two predominant families of the lateral dot sizes (as shown in Fig. 1). Their average sizes were about 26±5nm and 53±7nm, and their densities were about 5.9×10^{10} cm^{-2} and 1.5×10^{10} cm^{-2}, respectively. The larger dots may result from small dots coalesced partly under our growth condition. The bimodal distribution of dot sizes has been observed for self-assembled Ge quantum dots on Si, but the optical property of the bimodal distribution of dot sizes was not reported by Ross et al.[9]

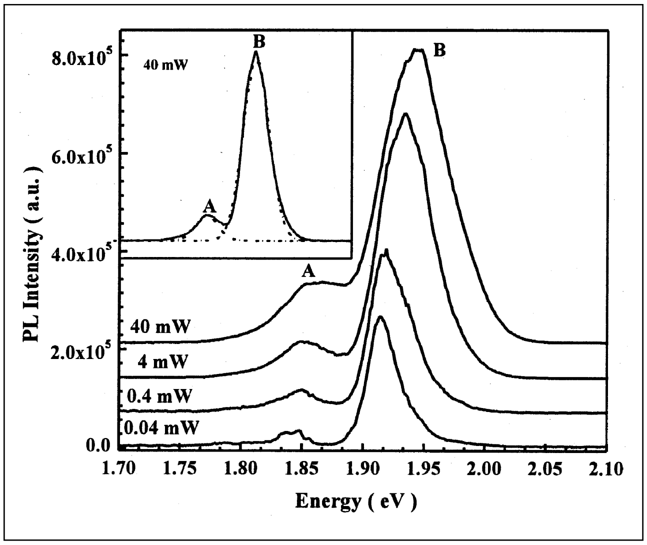
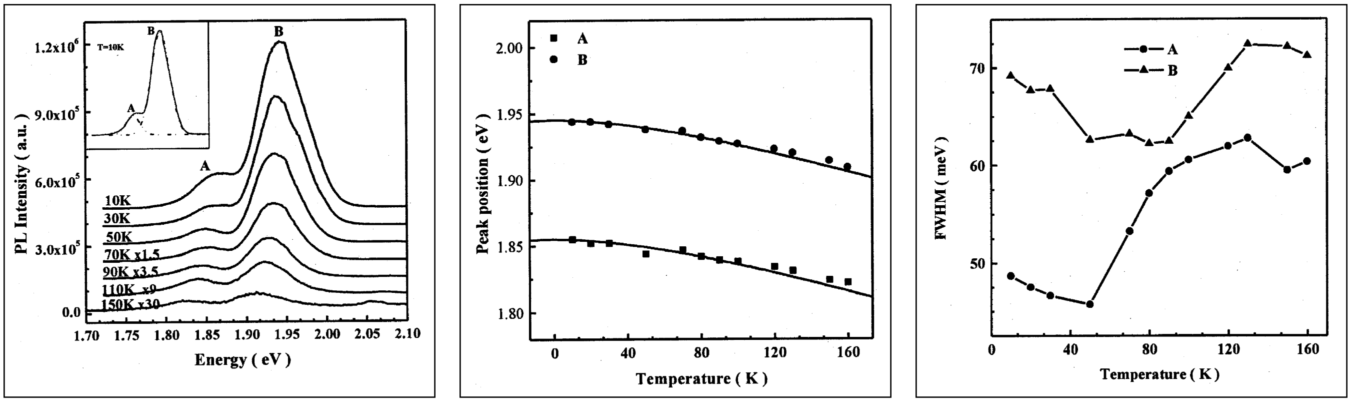


Fig. 2. PL spectra of the capped sample with different excitation power densities. Inset shows the Gaussian fitting of each peak (A and B) with one predominant size in each dot family (dash shot dot).

PL measurements were performed with a 406.7 nm line of Kr⁺ laser, and a GaAs detector by a lock-in technique. PL spectra of the capped sample obtained by excitation from 0.04 to 40 mW at low-temperature (10K) are shown in Fig. 2. The sample shows a low-temperature PL emission in red region, which is typical carriers recombination in InAlAs/AlGaAs quantum dot structures.[10] There were two peaks in all PL spectra. A double-peak structure of InAlAs quantum dots on GaAs has been observed by Tsatual et al.[11] They thought that two peaks stemmed from radiative recombination involving states of different localization energy. However, we found that the PL intensities of two peaks (A and B) for our sample depended linearly on the excitation power density over three orders of magnitude, and the PL intensity of high energy peak was stronger than that of low energy peak at very low excitation intensity. Moreover, the relative PL intensity of high energy peak is decreased with increase excitation intensity. These demonstrated that the high energy peak cannot stem from the excited-state of $In_{0.55}Al_{0.45}As$ quantum dots.

Figure 3a shows PL spectra of the capped sample obtained by excitation with 40 mW at different temperature. For each temperature, the each peak energy and full width at half maximum (FWHM) can be fitted by one Gaussian shape that suggests one



a b c

Fig. 3. (a) PL spectra of the capped sample at different temperatures. Inset shows the Gaussian fitting of each peak (A and B) with one predominant size in each dot family (dash shot dot). Temperature dependence of each peak position, the continuous lines are calculated according to the Varshni law using the parameters of InAlAs and are shifted along the energy axis (b). Temperature dependence of FWHM for each peak (c).

predominant size in each dot family. A long wavelength shift of each peak with increase temperature was due to the temperature-dependent bandgap shrinkage of the InAlAs quantum dots and AlGaAs barrier. We found that the temperature dependence of each peak energy followed Varshni equation (as shown in Fig. 3 b):

$$E_g(T) = E_g(0) - \frac{\alpha T^2}{\beta + T}$$

Where $E_g(0)$ is the band-gap energy at 0K, α is in the electronvolts per degree Kelvin, β is proportional to the Deby temperature (in Kelvin). The values used for the fitness of Fig. 3b are $\alpha = 4.73 \pm 0.3 \times 10^{-4}$ (ev/K) and $\beta = 149 \pm 40$ (K) based Ref. 12. The parameter $E_g(0)$ is determined by fitting. The FWHM of each peak as a function of temperature is also shown in Fig. 3c. At low temperature (from 10 to 100K), the FWHM of each peak was reduced, followed by an increase at higher temperature, this behavior have been observed for InAs and InGaAs quantum dots previously.[13,14] The temperature dependence of FWHM can be explained qualitatively with the thermalized carrier distribution in inhomogeneous quantum dots and the electron-phonon scattering. The PL linewidth of quantum dots is determined by the inhomogeneous distribution of dots size at low temperature. When the temperature increases, the thermalized carriers can be relaxed over a long distance and tunnel to nearby quantum dots, consequently, the thermalized carriers can repopulate in quantum dots. As the carrier lifetime for optical recombination process in InAlAs quantum dots (250 ps)[15] is more than the time for tunneling process (around 1 ps),[13] in addition, the average distance between dots was estimated as ~15 nm by the dot average size and density, the tunneling process must be taken into account. Under this condition, the tunneling occurrence will be more frequent for predominant quantum dot sizes due to their Gaussian distribution, resulting in a narrower linewidth with increase temperature. When the temperature is high enough, the electron-phonon scattering becomes a dominant contribution. The PL linewidth starts to increase with increase temperature. Thus the two-stage behavior appearing in the temperature dependence of FWHM, which cannot be observed in a single quantum well, confirmed that the high energy peak was unlikely attribute to the wetting layer (quantum well).

Taking into account quantum-size effect on the peak energy,[13] we believe that the excitons localized in a family of smaller dots are contribute to high energy peak, while the excitons localized in a family of larger dots are contribute to low energy peak. In our work, the intensity of high energy peak was stronger than the low energy peak, and indicated that the density of smaller dots must be larger than the density of larger dots. This result accord with the statistical distribution of dot lateral size from the AFM image. The FWHM of low energy peak was narrower than that of high energy peak, this case may be that the fluctuation of energy levels due to the size-distribution is more pronounced in smaller size. The multimodal distribution of dot sizes may still explain why the FWHM of low energy peak increased rapidly at low temperature (50K), the rare dot nearby larger dots results in the carriers tunneling difficult to larger dots, the electron-phonon scattering becomes a dominant contribution rapidly.

CONCLUSION

Self-organized $In_{0.55}Al_{0.45}As$/AlGaAs quantum dots have been successfully grown on GaAs (001) substrates by MBE, and shown that there is a bifurcation distribution of dot sizes by AFM. We present the optical properties of lateral size bimodal distribution of quantum dots. Taking into account quantum-size effect on the peak energy, we propose that the high (low) energy peak results from a smaller (larger) dot family.

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