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Research Paper

Temperature Dependent Photoluminescence from InAs/GaAs Quantum Dots Grown by Molecular Beam Epitaxy

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Abstract We have reported structural and optical properties of self-assembled InAs/GaAs quantum dot (QD) grown by molecular beam epitaxy with different arsenic to indium flux ratios (V/III ratios). By increasing the V/III ratio from 9 to 160, average diameter and height of the InAs QDs decreased, but areal density of them increased. The InAs QDs grown under V/III ratio of 30 had a highest-aspect-ratio of 0.134 among them grown with other conditions. Optical property of the InAs QD was investigated by the temperature-dependent photoluminescence (PL) and integrated PL. From the temperature dependence PL measurements of InAs QDs, the activation energies of E_{a1} and E_{a2} for the InAs QDs were obtained 48 ± 3 meV and 229 ± 23 meV, respectively. It was considered that the values of E_{a1} and E_{a2} are corresponded to the energy difference between ground-state and first excited state, and the energy difference between ground-state and wetting layer, respectively.

Keywords: InAs/GaAs, Quantum dot, and molecular beam epitaxy

I. Introduction

InAs/GaAs self-assembled quantum dots (QDs) as a zero-dimensional system have attracted extensive research interest due to the unique physical properties and development of high performance optoelectronics devices such as lasers, detectors, and solar cells [1-3]. Molecular beam epitaxy (MBE) or metal-organic chemical vapor deposition (MOCVD) have been most widely used for growth of self-assembed InAs QDs. Among various technique for formation of InAs QDs, the self-assembled InAs QDs grown by the Stranski–Krastanov (S-K) method have been proposed as a promising way to fabricate high-quality QDs, providing peculiar optical properties such as high quantum yield, tunability, and thermal stability. However, there are still many technical problems to solve to fully utilize their device applications. One of the main problems is the growth condition of InAs/GaAs self-assembled QDs which strongly influence their size, shape, and the wetting layer structure. A number of investigations have been performed to probe the formation mechanisms of InAs QDs by atomic force microscopy (AFM) [4,5], scanning tunneling microscopy (STM) [6,7], or transmission electron microscopy (TEM) [8]. It is well known that the variation of shape and energy gap for self-assembled InAs QDs significantly depends on Arsenic to Indium flux ratios (V/III ratios) [9]. However, there are not many reports for effect of V/III ratios on InAs QDs grown by MBE.

In this paper, the effects of different V/III ratios during MBE growth on the size and areal density of InAs QDs have been studied by using AFM and TEM, and the optical property of the InAs QDs was investigated by measurement of temperature-dependent photoluminescence (PL) and integrated PL.

II. Experiment Details

The samples used in the study were grown by conventional MBE (a Veeco Gen930). The samples were prepared as follows. All samples were grown on substrates in the MBE chamber. Prior to the deposition of InAs QDs, the semi-insulating (100) oriented GaAs substrate was initially deoxidized at 600°C. Afterwards, the substrate temperature was lowered to 580°C and an epitaxial GaAs buffer layer of approximately 100 nm was grown on the substrate. The substrate was cooled to a 480°C for the growth of a single layer of InAs QD. 2.1-ML-thick InAs QDs grown under various V/III ratios from 9 to 160 were formed on the GaAs buffer layer by means of the conventional S-K MBE. The As pressure during the InAs growth was changed from 3.2×10^{-7} to 6.1×10^{-6} Torr, and In flux was fixed at 0.05 ML/s. Next, the 40-nm-thick GaAs capping layer was deposited on the InAs QDs. Finally, the same InAs QDs was repeated on the capping layer to measure morphology of InAs QD.

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The density, height, and width of InAs QDs on top of the samples were investigated by using AFM. TEM images were taken to characterize the shape and structure of InAs QDs in a GaAs matrix. The PL measurements were performed from 15 to 300 K by using an Ar-ion laser with excitation wavelength of 514.5 nm line in a He-cryostat system. The QD emission was detected by using a 105 cm monochromator equipped with a liquid nitrogen cooled InGaAs detector.

III. Results and Discussion

Figure 1 shows AFM images of 2.1 ML InAs QDs grown under different V/III ratios from 9 to 160. Here, the scanned area is 1x1 μm^2. The areal density, and average diameter and height of the QDs are plotted as a function of V/III ratios in table 1. Among the QDs grown under V/III ratio from 9 to 160, the QDs grown under V/III ratio of 30 showed the highest-aspect-ratio (~0.134). The QD areal density increased nearly by one order of magnitude from 5×10^9 cm^{-2} (V/III ratio = 9) to 6.0×10^{10} cm^{-2} (V/III ratio = 30). Also, the average diameter decrease correspondingly from 35.5 to 33.1 nm. By further increasing the V/III ratio from 70 to 160, the areal densities of QDs increased slowly from 9.2×10^{10} cm^{-2} to 1.1×10^{11} cm^{-2}, and the corresponding average diameter decreased from 32.7 to 21.6 nm. Also, we noticed that the uniformity of QDs gets worse with increasing V/III ratio from 70 to 160. From these facts, increasing the V/III ratio from 9 to 160 induced a suppressed surface migration of indium atoms, so that the suppressed surface diffusion length resulted in an increase in the QD areal density [10]. Furthermore, By the effect of suppressed migration length of In atoms, the QD average diameter and height decreased with increasing V/III ratio, which was resulted from suppressing forming larger QDs by the process of island-island interaction [11]. Therefore, we can conclude that as V/III ratio increased from 9 to 160, the QDs areal density increased while the QDs average diameter and height decreased.

Table 1. Average height, width, and density of InAs QDs grown with V/III ratios from 9 to 160

Sample	V/III ratio	Avg. Height (nm)	Avg. Width (nm)	Aspect Ratio	Density (cm^{-2})
a	9	3.89	35.5	0.110	5.0×10^9
b	13	4.17	34.2	0.122	5.5×10^{10}
c	30	4.45	33.1	0.134	6.0×10^{10}
d	70	4.01	32.7	0.122	9.2×10^{10}
e	100	2.82	24.6	0.114	1.0×10^{11}
f	160	1.89	21.6	0.087	1.1×10^{11}

Figure 2 shows cross sectional TEM image of the InAs QDs grown under various V/III ratio from 9 to 160. InAs QDs grown under V/III ratio of 30 showed the best crystal quality and a highest-aspect-ratio, while InAs QDs grown under V/III ratio of 9 showed the degraded crystal quality. As V/III ratio increased from 70 to 160, the crystal quality of InAs QD was more degraded.

Figure 3(a) shows PL spectra of InAs QDs grown under V/III ratio from 9 to 160 at 10 K. As shown in AFM and TEM images of figure 1 and 2, FWHM of PL intensity for the InAs QDs grown under V/III ratio of 30 is sharper than any other samples. Because InAs QDs grown under V/III ratio of 30 showed the best crystal quality in the QDs, the temperature-dependence PL measurement for this sample was conducted in temperature ranging from 15 to 300 K. Figure 3(b) shows PL spectra measured at 10 K from the InAs QD grown under V/III ratio of 30. The ground-state and first excited state energy of the InAs QDs were observed at 1.104 and 1.154 eV, respectively, and the signal of InAs wetting layer appeared at 1.335 eV. The temperature-dependence of the ground-state PL peak

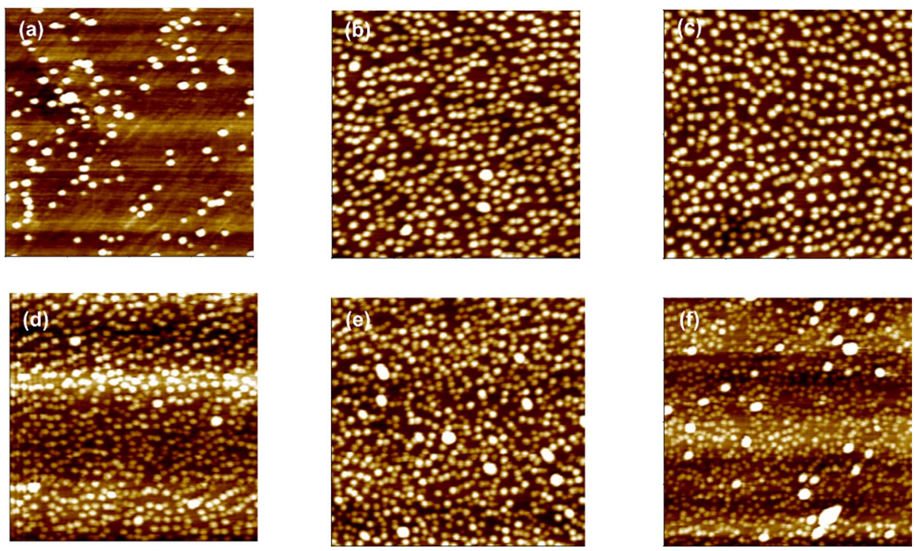

Figure 1. (a) ~ (f) shows AFM images (1 × 1 μm^2) of InAs QDs grown under different V/III ratios of 9, 13, 30, 70, 100, and 160, respectively. Here, InAs was deposited 2.1 ML under a fixed indium flux of 0.05 ML/s.

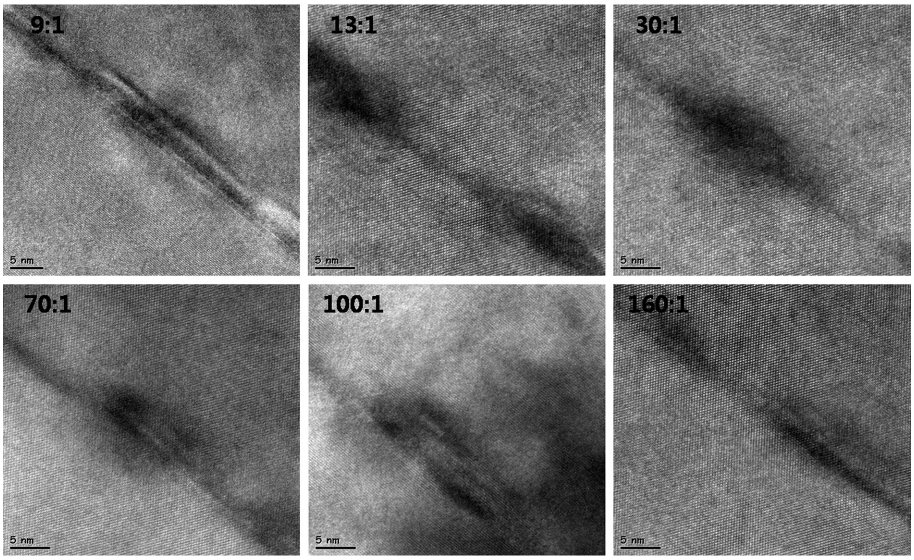

Figure 2. Cross-sectional FE-TEM images of InAs QDs grown under different V/III ratios from 9 to 160.

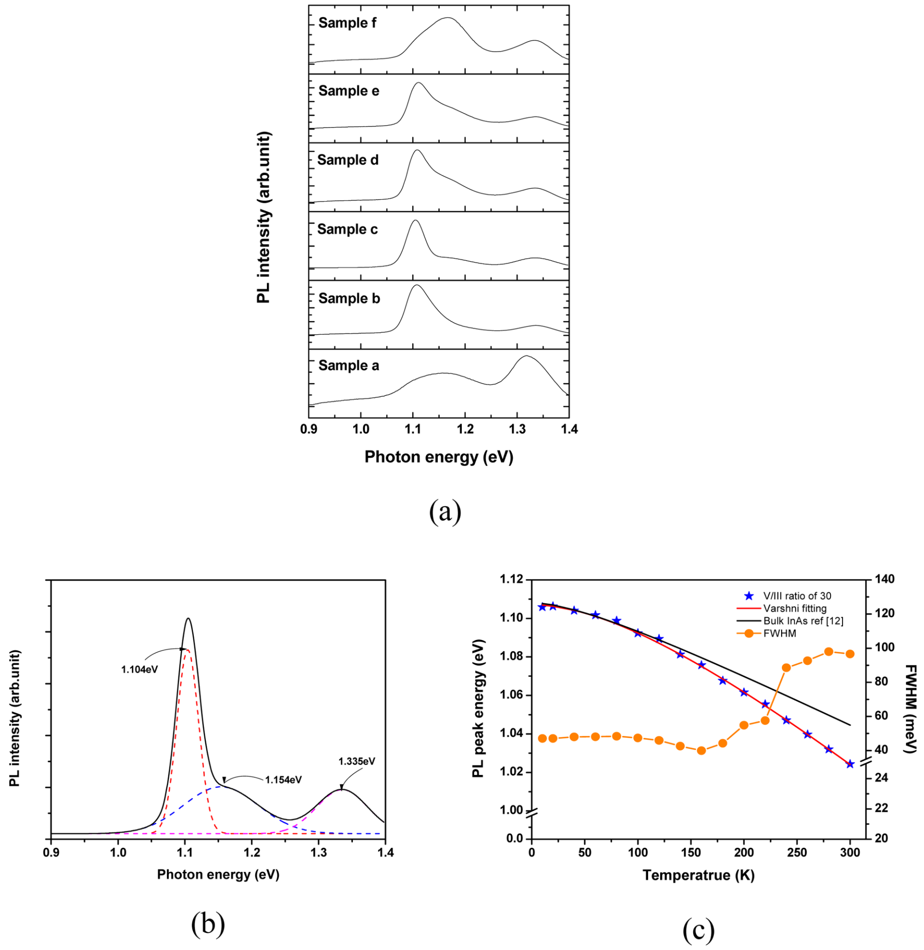

Figure 3. (a) PL spectra of InAs QDs grown under V/III ratio from 9 to 160 at 10 K (b) PL spectra of InAs QDs grown under V/III ratio of 30 at 10 K and (c) temperature-dependence ground-state PL energy and FWHM of InAs QDs grown under V/III ratio of 30.

position and FWHM of InAs QDs grown under V/III ratio of 30 are shown in Fig. 3(c). Here, the black dashed lines indicate the bulk InAs band gap shrinking obtained by Varshni's formula using the coefficient of bulk InAs. The Varshni coefficients (2.7×10^{-4} eV/K and 83 K) for bulk InAs were taken from other literatures [12]. The blue stars are the measured data from the ground-state PL peak position of the InAs QDs, and the red solid line is the theoretical fit to the experimental data by Varshni's formula. As shown in Fig. 3(c), the ground-state energies

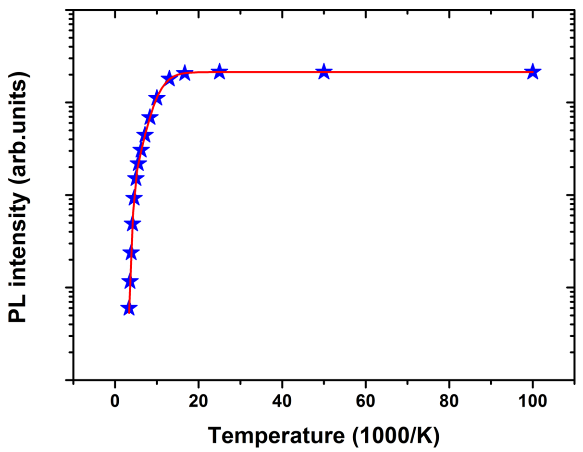

Figure 4. Integrated PL of InAs QDs grown under V/III ratio of 30. Blue-star marks are integrated PL intensities as a function of temperature from 10 to 300 K. The red line is fitting lines obtained by the equation for thermal activation energy.

of InAs QDs showed the temperature dependence. Due to the change of lattice parameter and electron-lattice interaction, the free excitonic energy follows the well-known Varshni's empirical formula:

$$E_g(T) = E_g(0) - \frac{\alpha T^2}{T + \beta} \quad (1)$$

where $E_g(T)$ is the PL peak energy gap at temperature T, $E_g(0)$ the energy gap at 0 K, and α and β are Varshini's fitting parameters [13]. In our calculation, the values of α and β for the InAs QDs were obtained 4.9×10^{-4} eV/K and 234 K, respectively. These fitting values are in good agreement with the reported result for InAs/GaAs QDs [12]. Due to the carrier localization effects, the ground-state PL peak energies of InAs QDs are more or less a constant below 40 K [13]. The PL peak energy of the QDs decreased as increasing temperature from 40 to 120 K and was similar to the behavior of InAs bulk material. At temperature above 120 K, however, the PL peak energy of InAs QDs was deviated from the Varshini curve of InAs bulk. PL peak energy of the InAs QDs red-shifted more faster than the band gap shrinkage of bulk InAs. On the other hand, a full-width at half-maximum (FWHM) of the InAs QDs at temperature ranging from 15 to 180 K was maintained about 40 meV. As increase the temperature above 180 K, the FWHM drastically increased with temperature due to the effect of electron-phonon scattering and thermal broadening [14]. It was considered that the fast red-shift of ground-state PL peak position and the increase of FHWM at the temperature above 180 K may be resulted from an increase in the migration rate from the small QDs with higher energy level to larger QDs with lower energy level [15].

Figure 4 shows integrated PL ground-state intensities for InAs QDs grown at V/III ratio of 30 as a function of temperature from 15 to 300 K. It appeared the onset of the thermal quenching at about 77K. The experimental data can be fitted using the well-known equation,

$$I = \frac{I_0}{1 + C_1 \exp\left(\frac{Ea1}{k_B T}\right) + C_1 \exp\left(\frac{Ea2}{k_B T}\right)} \quad (2)$$

where I(T) is the integrated PL intensity of the QDs at temperature T, k_B is the Boltzmann constant, T is the measuring temperature, C_i is a fitting constant proportional to the density of the non-radiative recombination center, and E_{ai} is activation energy. The values of $E_{a1}$and E_{a2} were extracted to 48 ± 3 meV and 229 ± 23 meV, respectively. The value of E_{a1} was well corresponded to the energy difference (50 meV) between PL peak of ground state and first excited state at 10 K, and the value of E_{a2} was considered to be the energy difference (225 meV) between PL peak of ground state and wetting layer at 10 K.

IV. Conclusions

The effect of V/III ratio on the size and areal density of InAs QDs during MBE growth by S-K mode was studied. As the V/III ratio increased from 9 to 160, the average width and height of the InAs QDs decreased, but the areal density of them increased. From AFM and cross-sectional TEM image, the InAs QD grown under V/III ratio of 30 had a high-aspect-ratio of 0.134. The ground-state PL peak position of this InAs QD system showed the fast red-shift and its FHWM significantly increased from 180 K, which can be explained by an increase in the migration rate from the small QDs with higher energy level to larger QDs with lower energy level. From the integrated PL intensity for the InAs QDs versus temperature, the activation energies of E_{a1} and E_{a2} were obtained 48 ± 3 meV and 229 ± 23 meV, respectively. The value of E_{a1} was in good agreement with the energy difference between ground-state and first excited state, and E_{a2} was considered to be the energy difference between ground-state and wetting layer.

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