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Influence of As-stabilized surface on the formation of InAs/GaAs quantum dots

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In this article, we report the growth of InAs/GaAs quantum dots (QDs) grown under different As_4-supply procedures. The growth of the investigated samples carried out by the three procedures of As shutter always opened, As shutter initially opened, and As shutter initially closed. The samples grown by the former two approaches show a uniform QD distribution and the multiple-peak luminescence, which correspond to ground-state, first-excited-state, and second-excited-state luminescence, while that grown by the latter only shows large InAs islands. The results suggest that the As-stabilized condition at the initial stage of QD growth is very critical for the high-quality QD formation. [DOI: 10.1116/1.2912083]

I. INTRODUCTION

Quantum-dot (QD) structures prepared by molecular beam epitaxy (MBE) have attracted much attention in recent years due to their unique optical and electrical characteristics.[1–3] The self-assembled QD structures grown under the Stranski–Krastanov (SK) growth mode are of high uniformity and defect free. Therefore, the QD structures are very promising for the applications of laser diodes and infrared photodetectors.[4] It has been reported elsewhere that the uniformity and optical properties of InAs/GaAs QDs prepared by using an As_4 flux would be greatly improved under the As-deficient condition.[5] However, the influence of As_4 overpressure on the formation of InAs/GaAs QDs has not been clarified yet. In this report, InAs/GaAs QDs grown under different As_4-supply procedures are investigated. Similar surface morphologies and photoluminescence (PL) spectra are observed for QDs with both the As shutter always opened and initially opened procedures, while no uniform QD formation is observed for the sample with the As shutter initially closed procedure. The results suggest that the As-stabilized condition of the GaAs substrate surface is necessary for high-quality QD formation.

II. EXPERIMENT

The InAs/GaAs QD samples investigated in this study were prepared by solid-source MBE under As_4 background pressure. The As_4 beam equivalent pressure was kept at 1×10^{-5} torr during growth. Three samples with different As_4-supply procedures were grown on (100) semi-insulating GaAs substrates. The InAs QDs are formed by SK mode at 510 °C. The InAs growth rate was kept at 0.08 ML/s, which corresponds to a V/III ratio of 50. The epitaxial structures are shown in Table I. The sample grown with As shutter always opened is referred as sample A, while the samples grown with As shutter initially opened and initially closed procedures are referred as samples B and C, respectively. For PL measurements, a 532 nm green laser is used as the pumping laser. The laser light is directed on the samples placed in a cryostat such that PL spectra can be measured from 10 to 300 K by temperature controller. The PLE spectrum is measured by using Jobin Yvon's NanoLog3 system coupled with tungsten-halogen lamp as the light source. The PL and PLE signals were focused to a spectrometer coupled with an InGaAs *p-i-n* detector. Atomic force microscopy (AFM) Digital Instruments Dimension 3100 was used for surface morphology observations.

III. RESULTS AND DISSCUSSION

Figure 1 shows the $1\times1\ \mu m^2$ AFM images of the three samples. As shown in the figure, samples A and B show a uniform QD distribution with a QD density of $\sim2.8\times10^{10}\ cm^{-2}$, a mean dot diameter of 54 nm, and a mean height of 6 nm, while sample C only exhibits large InAs islands. Although the total As_4 supplies for samples B and C are the same, it is shown from the AFM images that the

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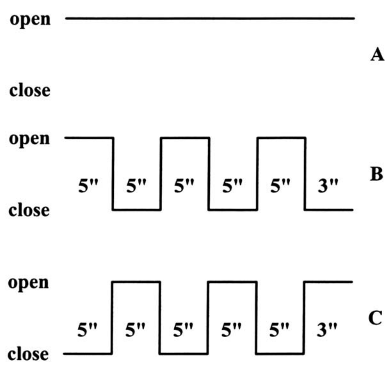
TABLE I. Wafer structures and As shutter close/open procedures of samples A, B, and C.

Wafer structure
2.4 ML InAs QD
60 nm GaAs Layer
30 nm AlAs Layer
60 nm GaAs Layer
2.4 ML InAs QD
60 nm GaAs Layer
30 nm AlAs Layer
GaAs Buffer
(100) SI GaAs Substrate

As shutter close/open procedure



initial surface conditions are critical for the InAs QD formation. For sample B, the As are sufficient for the initial InAs epitaxial growth with the As shutter initially opened procedure. Therefore, similar surface morphology with As shutter always opened procedure, sample A, is obtained for sample B. For the case of sample C with In shutter initially closed procudure, the In coverage is merely 0.43 ML within the 5 s As shutter closed period. Although the period of time is too short for clear reflection high energy electron diffraction pattern observation, it is reasonable to assume that the background As_4 pressure is not sufficient to prevent metal-rich surface formation. In this case, possible In droplet formation may be obtained at the initial growth stage of sample C.[6] The following In adatoms would tend to accumulate nearby the In droplets, which leads to large InAs island formation for sample C.

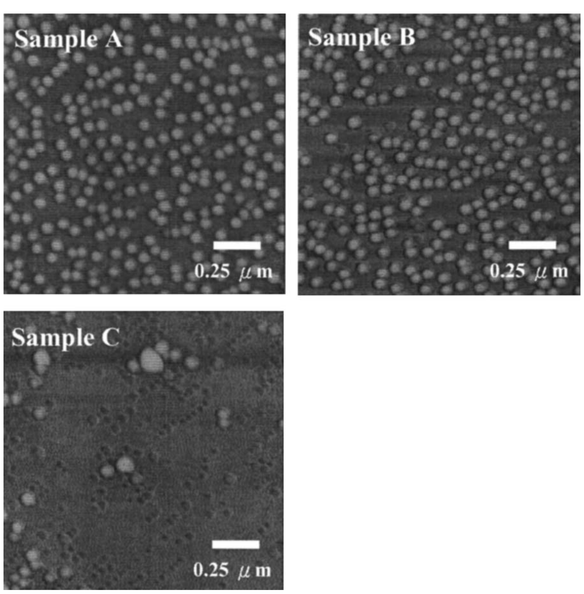


FIG. 1. 1×1 μm^2 AFM images for the three samples A, B, and C.

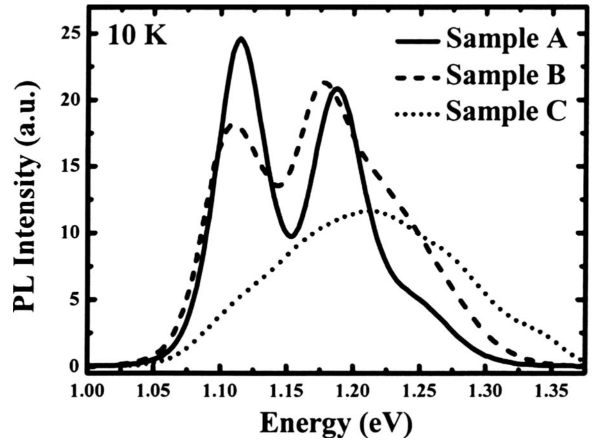


FIG. 2. 10 K PL spectra of samples A, B, and C.

Figure 2 shows the 10 K PL spectra of samples A, B, and C. Samples A and B exhibit the multiple-peak luminescence, which correspond to ground-state, first-excited state, and second-excited-state luminescence. The similar PL performance from samples A and B shows that both the samples have similar crystal quality. This also reveals that as long as the initial InAs film is grown under As-stabilized condition, the close/open procedure of the As shutter does not play an important role of the QD formation. For sample C without the QD formation, the higher-energy peaks in the PL spectrum should be attributed to the InGaAs clusters resulting from In/Ga interdiffusion at the initial stage. The PL results are consistent with the observation of AFM images for the three samples.

To further investigate the optical characteristics of the QD samples, Fig. 3(a) shows the normalized PL spectra at 10 K of sample A measured for different laser excitation powers. At an excitation power of 34 mW, there are three peaks denoted as E_0 (1.12 eV), E_1 (1.19 eV), and E_2 (1.25 eV), corresponding to the ground-state, first-excite-state, and second-excited-state luminescence, respectively. The phenomena are attributed to the state-filling effect.[7] Due to the longer electron lifetime of excited states of the QD structures; the photoexcited electrons would fully occupy the states with lower energies. Therefore, luminescence from the higher-energy state would be observed from the PL spectra. By reducing the laser excitation power to 15 mW, the E_2 peak disappears and the intensity ratio of peak E_1 to peak E_0 also decreases, as shown in Fig. 3(a). The results are attributed to the generation rate of photoexcited electrons at the first excited state to be lower than their recombination rate. Therefore, the first excited states are empty for the sample under such a measurement condition. The multiple-peak luminescence of sample A observed under such a low excitation power exhibited its high crystal quality. To verify the attribute of the transition mechanisms discussed above, the 10 K photoluminescence excitation (PLE) spectrum of sample A with detection energy at E_0 (1.12 eV) is shown in Fig. 3(b). As shown in the figure, the peaks located at 1.19 and 1.25 eV are observed from the PLE spectrum, which are consistent with the

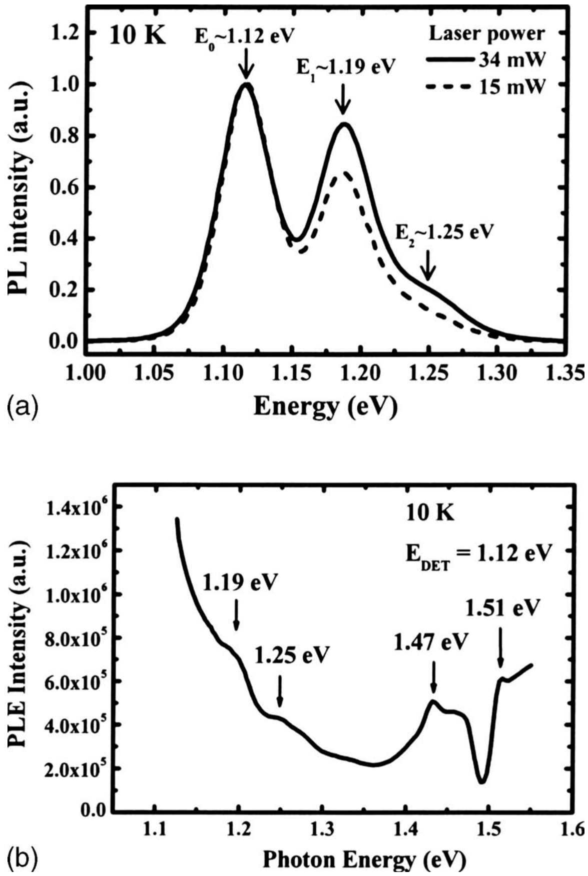


FIG. 3. (a) Normalized 10 K PL spectrum measured under different laser excitation powers and (b) the 10 K PLE spectrum of sample A.

PL peaks denoted as E_1 and E_2. The identical peak positions obtained from two different measurement methods have confirmed the attribute of transition mechanisms discussed above. Also shown in the PLE spectrum are two additional peaks located at 1.47 and 1.51 eV, which result from the InAs wetting layer and GaAs bandedge luminescence, respectively.[8]

IV. CONCLUSIONS

We have demonstrated the growth of InAs/GaAs QDs under different As_4-supply procedures. Similar surface morphologies and PL spectra are observed for the quantum dots grown with both the As shutter always opened and initially opened procedures, while no uniform QD formation are observed with the As shutter initially closed procedure. The phenomenon is attributed to the In droplet formation at the GaAs surface under the As-deficient condition. In this case, the In adatoms would tend to accumulate nearby the In droplets such that large InAs islands would be observed. The results suggest that the As-stabilized condition at the initial stage of QD growth is very critical for the high-quality QD formation. Identical peak positions observed from the multipeak PL spectrum and PLE spectrum have revealed the higher-order confinement states of the QD structures.

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