Dependence of the InAs size distribution on the stacked layer number for vertically stacked InAs/GaAs quantum dots


Abstract

Atomic force microscope (AFM), transmission electron microscopy (TEM), and photoluminescence measurements were carried out to investigate the dependence of the InAs quantum dot size distribution on the stacked layer number for vertically stacked InAs/GaAs quantum dots (QDs) grown on (001) GaAs substrates. AFM and TEM images showed that the size of the QDs increased with increase in the stacked layer number up to the deposition time of 20 s. However, the size distribution uniformity of the QDs was improved with increase in the stacked layer number when the deposition time and the stacking layer of the InAs QDs gradually decreased. These results can help in an improved understanding of the control of sizes of QDs in InAs/GaAs QD arrays. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

Self-assembled quantum dots (SAQDs) have been fabricated on various material systems [1–3] and utilizing various growth techniques by using the Stranski–Krastanow (S–K) growth mode [4] since SAQDs have both physical properties of discrete artificial atom-like energy levels, thermal stability of states, high gain, $\delta$-function-like density of states, and good optical properties [5], and potential applications such as quantum dot lasers [6], optical quantum memories [7,8] and infrared photodetectors [9,10]. In highly lattice-mismatched systems, this growth process starts a two-dimensional mode, and islands are formed spontaneously remaining the thin wetting layer under dots beyond a certain critical thickness [11]. Since the SAQDs are formed spontaneously, the
precise control of the distribution and the size of the SAQDs are very difficult. Since the size and the site of the quantum dots (QDs) are randomly distributed, the full-width at half-maximum (FWHM) of photoluminescence (PL) spectrum is broadened [12–14]. It is very important to control the size of the QDs for improving their optical efficiency. Recently, it has been found that the multiple stacking process of the QD layers, which form the vertical ordering, is useful to control sizes of the QDs [15,16]. However, the size of the QDs increases with increasing the stacking layer number [17]. Therefore, studies concerning the dependence of the InAs size distribution of vertically stacked InAs/GaAs QDs on the deposition time are very important.

This paper reports data for the dependence of the InAs size distribution of vertically stacked InAs/GaAs QDs grown by using molecular beam epitaxy (MBE) on the deposition time. Atomic force microscopy (AFM) and transmission electron microscopy (TEM) measurements were performed to characterize the surface morphology and the microstructural properties in InAs QD arrays inserted into GaAs layers. PL measurements were carried out in order to investigate the interband transitions in the samples.

2. Experiment

The samples used in this work were grown on semi-insulating (100)-oriented GaAs substrates by using MBE. The growth rate of the InAs and the GaAs was 1.15 A/s. The whole growth process was controlled in situ by reflection high energy electron diffraction (RHEED). The QD samples studied in this study consisted of the following structures. A 100 nm thick GaAs buffer layer was first grown on a GaAs substrate at 530°C. Then, the substrate temperature was lowered to 410°C for the InAs/GaAs QD array growth. The InAs wetting layer was deposited followed by a GaAs spacer layer after a 10s growth interruption. The RHEED pattern became spotty after the deposition of the 1.8 ML InAs layer, indicative of three-dimensional island formation. After the substrate temperature was increased to 430°C, a 10 nm undoped GaAs layer was grown. The sample structure for the multiple stacked InAs/GaAs QDs is the same as the above-described sample except that the spacer thickness is 4 nm. Five periods of the InAs/GaAs SAQD arrays were grown at 410°C via the S–K growth mode. This relatively low temperature growth reduced the sizes of the QDs. S1 represents a single layer consisting of QDs, and S2 five stacked layer QDs without a capping layer. S3, S4 and S5 represent single-stacked, three-stacked and five-stacked QDs layers with capping layers, respectively. The deposition times for vertically stacked InAs/GaAs QDs gradually decreased with a stacking layer. The depositions of the InAs layers are summarized in Table 1.

The AFM observations were performed to measure the size and density of the QDs without capping layers and carried out to check the vertical ordering and the size and density of the QDs with a capping layer, the TEM observations were performed in a JEM 2000 EX transmission electron microscope operating at 200 kV. The samples for the cross-section and plan-view TEM measurements were prepared by cutting and polishing with a diamond paper to a thickness of approximately 30 μm and then argon-ion milling at liquid nitrogen temperature to electron transparency. The PL measurements were carried out using a 0.25 m monochromater equipped with a liquid nitrogen-cooled InGaAs and Si detector. The excitation source was the 514.5 nm line of an Ar⁺ laser.

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<th>InAs layer number</th>
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The last InAs layers of S1 and S2 samples are uncovered. The last InAs layers of S3, S4, and S5 samples are covered with GaAs.
3. Results and discussion

AFM images for the single and the five-stacked InAs/GaAs QDs without capping layers are shown in Figs. 1(a) and (b), respectively. Fig. 1(a) shows a high density \( \sim 9 \times 10^{10} \text{cm}^{-2} \) of QDs. The average peak height of the QDs is 3.3 nm, and the average diameter of the QDs about 20 nm. Fig. 1(b) shows that the density of the S2 QDs is approximately \( 5 \times 10^{10} \text{cm}^{-2} \). The average peak height of the QDs is about 6 nm and their average
diameter of the QDs is approximately 40 nm. These results indicated that the density of the QDs decreased but the size of the QDs increased with increasing the stacking layer number.

Cross-sectional micrographs of the S2 sample are shown in Fig. 2. Fig. 2(a) and (b) are a (004) bright-field and dark-field TEM image, respectively. Coherently strained islands are clearly observed for the S2 sample, which is indicated by the characteristic dark and the bright contrasts related to an inhomogeneous lattice deformation in the three-dimensional structure and to the absence of dislocations [18]. The strain field surrounding the islands extends to 5–10 nm into the GaAs buffer and cap layers. Fig. 2 shows that the size of the upper side of the QDs is larger than that of the lower side of the QDs, which is also in reasonable agreement with the results of the AFM images, as shown in Fig. 1. These results indicated that the size of the QDs increases with increase in the stacked layer number up to the deposition time of 20 s.

The uniform SAQDs with a high density and a small size are necessary for device application, such as QD lasers. One of the possibilities to achieve the QDs with high density is to stack InAs QD layers closely separated by a thin GaAs spacer. However, when the stacked layer number increases, the size of upper side of the QDs increases [17]. This result is in reasonable agreement with the AFM results, as shown in Fig. 1. In
In order to improve the size uniformity of the stacked layer QDs, the variation of the size and the optical properties of the QDs dependent on the deposition time of the InAs wetting layer have been investigated. The deposition times for the vertically stacked InAs/GaAs QDs decreased with a stacking layer number. Figs. 3(a–c) show cross-sectional micrographs for S3, S4, and S5 samples, respectively. They show vertically aligned quantum columns. Since the InAs QDs are largely lattice-mismatched to the GaAs layers, the strain relaxation can occur. However, there are no dislocations around the InAs QDs embedded in the GaAs barriers, as shown in Fig. 3. It is clearly seen that the size of upper size QDs is almost the same as that of the lower side QDs.

Figs. 4(a–c) show plan-view micrographs for S3, S4, and S5, respectively. The apparent diameter of the islands was determined by measuring the length of the white and black boundary on the plan-view micrograph of Fig. 4. Due to the extent of the strain field around the islands, the apparent diameter is larger than the actual island diameter. The distribution of the apparent diameter of the islands for S3, S4, and S5 is centered between 20 and 30 nm. The areal density is approximately $5 \times 10^{10}$ cm$^{-2}$, which is in reasonable agreement with AFM results, as shown in Fig. 1. These results indicated that the size distribution of the QDs was improved with increase in the stacked layer number when the deposition time of the InAs QDs gradually decreased with stacking layers.

Fig. 5 shows PL spectra at 26 K for S3–S5 samples. Strong luminescence peaks corresponding to interband transitions from the ground electronic subband to the ground heavy-hole band are observed in all the samples. Only one broad peak due to the size fluctuation of QDs is observed for S3 and S5. However, two broad peaks are observed for S4, which is attributed to the bimodal distribution in the size of the QDs. The appearance of two broad peaks is related to the existence of the large and the small dots, as shown in Fig. 4(b). The shift in the PL peak position to the low-energy side or to the high-energy side by changing the number of the stacking layers is attributed to the size distribution of QDs. The size of the QD increases or decreases by increasing the stacking layer number. While the InAs QDs with smaller sizes emit at higher energies, the InAs QDs with larger sizes emit at lower energies. Therefore, the peak position for the five-stacked QD layers is located at the lowest energy side. The PL peak position increases, and the FWHM of the PL peak at low temperature becomes smaller as the stacked number increases. The FWHM of the PL peak for the QDs with five-stacked layers is 84 meV, and this value is the smallest, indicative of the improvement of the size distribution of the QDs. Thus, when the deposition time of the InAs wetting layer is controlled, the narrow size distribution can be achieved, and the integrated intensity of the PL peak might be increased.

4. Conclusions

Dependence of the microstructural and the optical properties on the growth times for vertically stacked InAs/GaAs QDs was investigated. The size of the QDs increases by increasing the stacking layer number when the growth time of the InAs layer is constant. However, the results of TEM and PL measurements showed that the narrow size distribution of vertically stacked InAs QD arrays inserted into GaAs barriers could be achieved when the growth time of InAs QDs gradually decreased by increasing the stacking
layer number. These results can help in an improved understanding of the control of sizes of QDs in InAs/GaAs QD arrays.

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References