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Formation process and lattice parameter of InAs/GaAs quantum dots

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The formation of self-assembled InAs quantum dots (QDs) on GaAs substrates has attracted much interest as a growth technique to fabricate promising nanoscale devices such as QD lasers [1] and QD infrared photodetectors [2, 3] without an additional lithography process because QDs have discrete atom-like energy levels with good optical properties [4]. Self-assembled QDs in lattice-mismatched systems, such as InAs/GaAs and SiGe/Si, can be achieved by using the Stranski-Krastanov (SK) growth mode [5]. In the SK growth mode, the mismatched layer grows on the substrate two-dimensionally at first, then, above a critical thickness, strain-induced and dislocation-free QDs are formed on a residual two-dimensional wetting layer. An appropriate regulation of the respective growth conditions may provide the possibility for controlling precisely the size and the density of the QDs.

The structural properties of grown films are generally investigated by using such methods as transmission electron microscopy [6], atomic force microscopy [7], scanning tunneling microscopy [8], and X-ray diffraction [9, 10]. However, reflection high-energy electron diffraction (RHEED) [11] is a powerful tool to investigate *in situ* growth processes during molecular beam epitaxy (MBE) because the diffraction pattern provides information on the growth mode, the oscillation intensity provides information on the growth rate, and the spot position provides information on the surface lattice parameter [12, 13]. In spite of this, few studies concerning the time evolution of dot formation and the strain relaxation of QDs by using RHEED have been performed. Furthermore, detailed work on the formation process and lattice parameter of InAs/GaAs QDs are very important for enhancing the uniformity of the QDs.

This letter reports on the formation process and the lattice parameter of the QDs in lattice-mismatched InAs layers on (100) GaAs substrates. In particular, we use *in situ* RHEED to investigate the initial growth transition mode and the lattice relaxation during the self-assembled formation process on the InAs thin layers on (100) GaAs substrates. A schematic diagram of total energy as a function of time for the morphology transition is described on the basis of the results of RHEED patterns.

The samples used in this work were grown on semi-insulating (100)-oriented GaAs substrates by using MBE, and the surface coverage due to molecular adsorption was studied using a RHEED system. The substrate temperature was monitored with an infrared pyrometer. The native oxide layer on the substrate surface was thermally removed at 530 °C under an As_4 pressure of approximately 10^{-5} Torr. The InAs and the GaAs growth rates were set to 0.077 and 1.42 monolayer (ML)/s, respectively. The whole growth process was controlled by using *in situ* RHEED. A 100-nm-thick GaAs buffer layer was first grown at 530 °C, followed by 20 periods of an $Al_{0.25}Ga_{0.75}As$/GaAs superlattice buffer layer, and then by a 200-nm undoped GaAs buffer layer. Then, the In/As flux ratio and the growth temperature were varied in order to investigate lattice relaxation during InAs QD formation.

RHEED observation was performed *in situ* during InAs QD formation. The RHEED patterns on the fluorescent screen were recorded with a charge coupled device camera. The specular spot of the RHEED oscillation was focused onto the camera screen. The images were analyzed to investigate the evolution of the spot intensity and the spacing between diffraction rods.

The RHEED pattern is a sensitive monitor of the growth mode transition from a two-dimensional (2D) to a three-dimensional (3D) mode in self-assembled QD formation. A streaky RHEED pattern indicates that

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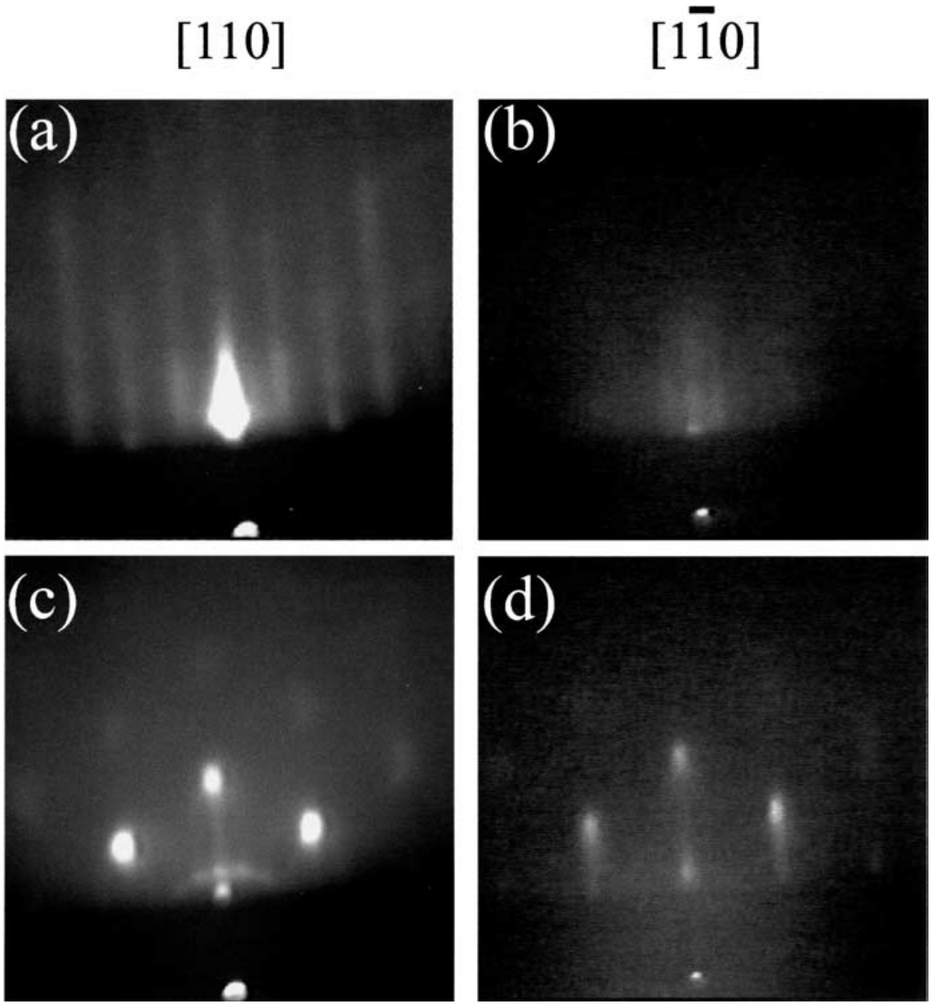

Figure 1 RHEED patterns along the (a) [110] direction and the (b) $[1\bar{1}0]$ azimuthal direction. The InAs layer thicknesses of (a) and (b) are 0, and those of (c) and (d) are 1.54 monolayer. The growth time = 18 s.

the wetting layer is a 2D layer. As soon as 3D islands are formed, the streaky pattern abruptly changes to a spotty one. The thickness of the InAs layer required for the 2D–3D growth mode transition strongly depends on the growth conditions. The RHEED patterns observed before and after QD formation are shown in Fig. 1. Fig. 1a and b are the [110] and the $[1\bar{1}0]$ azimuthal RHEED patterns obtained prior to InAs QD formation. Streaky patterns were clearly observed. The transition from streaky patterns to spots starts at an InAs-layer thickness of about 1.4 ML, indicative of a transition from 2D to 3D growth. Fig. 1c and d are for an InAs thickness of 1.4 ML. When the thickness of the InAs layer is above 1.4 ML, the RHEED pattern returns gradually to a streaky one.

Fig. 2 shows the dependence of the $[1\bar{1}0]$ azimuthal RHEED patterns on the growth temperature for an InAs deposition time of 20 s. The $[1\bar{1}0]$ RHEED patterns in Fig. 2a and b show V-shaped spots called "chevrons" [9]. So far, the origin of the chevrons is still controversial [14]. They may be attributed to reciprocal lattice rods, which are perpendicular to the side facets of pyramid-like InAs QDs. Characteristic chevrons are observed along the $[1\bar{1}0]$ direction with an angle of 50 ° (±3 °), which reflects at angles of 25 ° (±1.5 °) between the surface and the pyramidal side facets. The chevrons may originate from elongated islands in the $[1\bar{1}0]$ direction with well-defined facets parallel to the $(\bar{1}\bar{1}3)$ and the (113) planes [15]. However, theoretical refraction calculations suggest that chevrons from QDs may be produced because of pure refraction effects [14]. On the other hand, V-shaped spots are not observed along the [110] direction due to the anisotropy of the QD. The intensity of the V-shaped spots increases as the growth temperature increases from 430 to 450 °C. This result indicates that islands elongated in the $[\bar{1}10]$ direction with facets parallel to the $(\bar{1}\bar{1}3)$ and the (113) planes were produced. The V-shaped spots disappear at a growth temperature of 480 °C. This result indicates that islands elongated in the $[\bar{1}10]$ direction with facets were poorly developed and that the 3D island morphology was relatively smooth. These results indicate that a transition of the QD shape had occurred at a growth temperature of 480 °C.

The surface lattice parameter, which is proportional to the inverse of the distance between the (10) rod and the $(\bar{1}0)$ diffraction rods, can be measured directly from the RHEED patterns [12, 13]. Fig. 3 shows the dependence of the InAs in-plane lattice parameter on the As/In ratio for a substrate temperature of 430 °C. The surface lattice parameter abruptly changes before and after the formation of the QDs. Fig. 3a shows the variation of the lattice parameter for As/In = 85. The lattice parameter is almost constant for increasing InAs growth

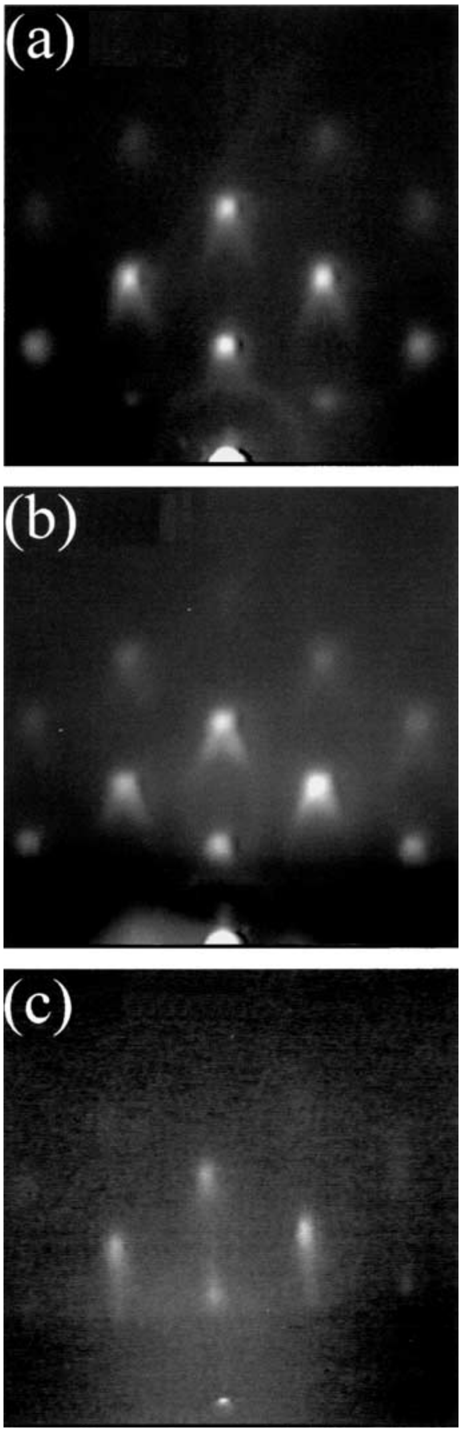

Figure 2 RHEED patterns along the [1$\bar{1}$0] azimuthal direction at different temperatures: (a) 430, (b) 450, and (c) 480 °C, for an InAs deposition thickness of 1.54 monolayer.

time up to a critical thickness, above which it decreases and then dramatically increases. This is indicative of a change in the InAs growth mode from 2D to 3D. After the QDs are formed, the lattice parameter remains almost constant below that of bulk InAs. When the As/In flux ratio is decreased, the lattice constant shows a different behavior, as shown in Fig. 3b. The time required for the QD formation decreases from 14.5 to 13.68 s. After QDs are formed, the lattice parameter rapidly decreases from 6.06 to 5.85 Å, and then smoothly increases. This indicates that the strain is fully relaxed just after the QD formation; then, the QD is strained and slowly relaxed.

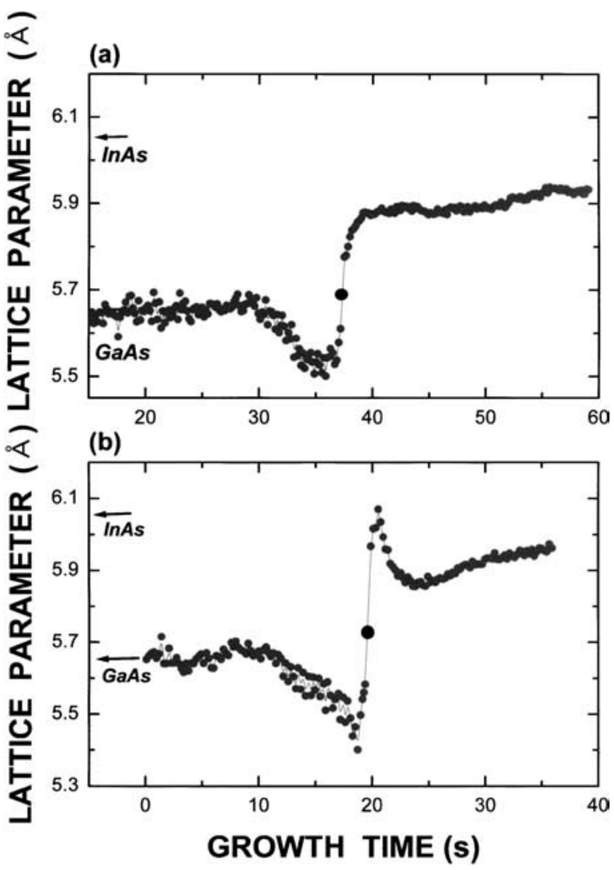

Figure 3 Surface lattice parameter estimated from the distance between the (10) and the ($\bar{1}$0) rods at the [110] incident azimuth of InAs grown at 430 °C: (a) As/In = 55 and (b) As/In = 20.

Fig. 4 shows the temperature dependence of the InAs in-plane lattice parameter for As/In = 85. Fig. 4a and b show the lattice parameters corresponding to substrate temperatures of 430 and 460 °C, respectively. As soon as the wetting layer is formed, the lattice parameter slightly increases and then smoothly decreases, as shown in Fig. 4a. After the QDs are formed, the lattice parameter abruptly increases and becomes nearly constant. The result of Fig. 4b is similar to that of Fig. 3b, except that the lattice parameter slowly decreases from 6.06 to 5.9 Å after the QDs are formed, and then smoothly increases. These results indicate that the strain was fully relaxed just after the QD formation; then, the QD was strained and slowly relaxed.

Fig. 5 shows a schematic diagram of the total energy as a function of time for the 2D-3D morphology transition [16]; (a) 2D layer-by-layer growth, (b) 2D metastable layer-by-layer growth, (c) 2D-3D transition, and (d) ripening period. "t_{cw}" is the thermodynamically defined critical wetting layer thickness. E_{A} is the barrier for formation of 3D islands, E_{E} is the excess energy stored in the supercritically thick wetting layer, and X is the point where a pure strain-induced transition would become possible.

During initial growth, the deposition follows a 2D layer-by-layer growth which is thermodynamically stable. Then, the InAs wetting layer receives compressive stress during a metastable period. Thus, the lattice parameter of the InAs wetting layer slightly decreases due to the elastic distortion of the InAs lattice during

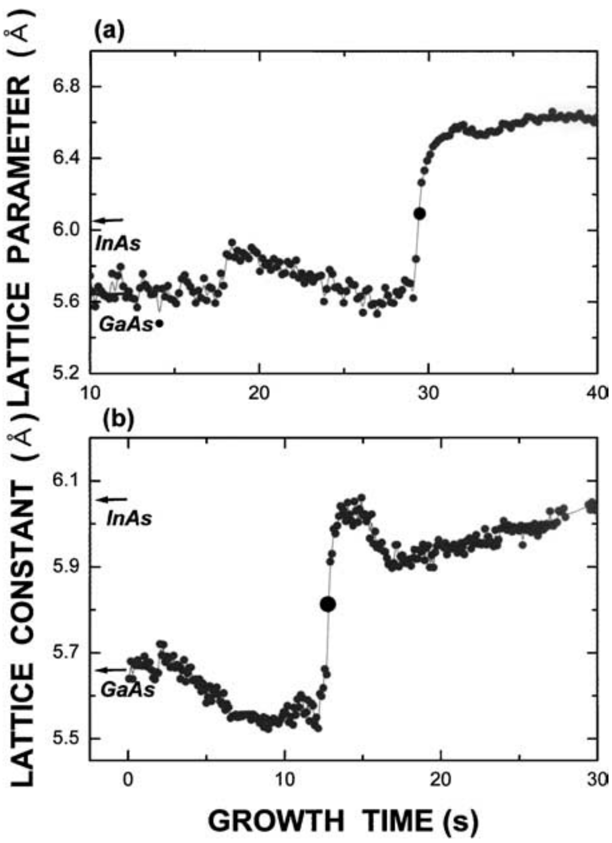

Figure 4 Surface lattice parameter estimated from the distance between the (10) and ($\bar{1}$0) rods at the [110] incident azimuth of InAs grown at As/In = 85 for growth temperatures of (a) 430 and (b) 460 °C.

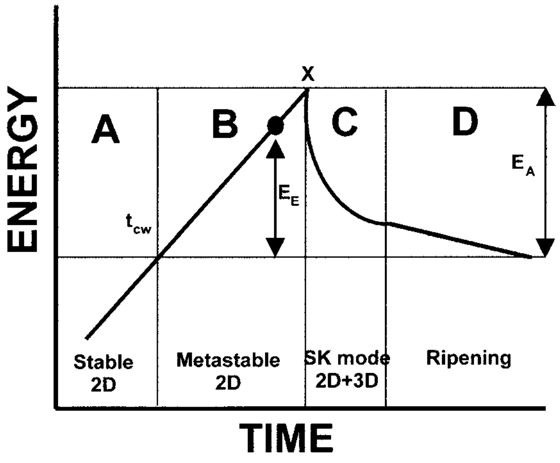

Figure 5 Schematics of total energy as a function of time for the morphology transition from a two-dimensional to a three-dimensional mode: (a) two-dimensional layer-by-layer growth, (b) two-dimensional metastable layer-by-layer growth, (c) transition from two-dimensional to three-dimensional mode, and (d) ripening period. "t_{cw}" is the critical wetting layer thickness, which is defined thermodynamically. E_A is the barrier for formation of three-dimensional islands, E_E is the excess energy stored in the supercritically thick wetting layer, and x is the point where a pure strain-induced transition would become possible.

layer-by-layer growth. At point X, the 2D-3D transition occurs. At that point, the lattice parameter increases abruptly, and the stored strain energy is partially relaxed. After the strain-induced 2D-3D transition occurs, the lattice parameter approaches the value of bulk InAs due to the ripening effect of the QD.

In summary, the formation process of InAs QDs grown on a GaAs substrate was observed by using RHEED. The growth process of the InAs layer during the initial stage is clearly divided into 2D growth and QD formation. The in-plane surface lattice parameter of the InAs layer was measured to investigate the strain relaxation behavior of the InAs QDs. The temperature dependence of the InAs in-plane lattice parameter was discussed. Based on the RHEED results, total energy as a function of time for the morphology transition from the 2D to the 3D mode is presented. These results provide important information on the growth process and the strain-relaxation behavior of InAs/GaAs QDs.

Acknowledgments

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