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# Growth mode of $In_xGa_{1-x}As$ $(0 \leqslant x \leqslant 0.5)$ on GaAs(001) under As-deficient conditions

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We have systematically studied the growth modes of $In_xGa_{1-x}As$ on GaAs(001) $(0 \leqslant x \leqslant 0.5)$ under As-deficient conditions. The $In_xGa_{1-x}As$ film coherently grows in a layer-by-layer mode, with a significant amount of In atoms being segregated to the growing surface. The In segregation results in the depletion of In in the $In_xGa_{1-x}As$ film, so that the strain at the coherent interface is reduced. The growth under higher As fluxes suppresses the In segregation and induces the formation of the relaxed $In_xGa_{1-x}As$ islands.

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## I. INTRODUCTION

Molecular-beam epitaxy (MBE) in lattice-mismatched semiconductor systems is usually assumed to proceed in a Stranski-Krastanov (SK) growth mode. In a typical lattice-mismatched system of InAs on GaAs (lattice-mismatch $\approx 7.1\%$), however, the growth mode strongly depends on the surface orientation of the GaAs substrate. InAs islands are formed on the (001) substrate after the ~2 bilayer (BL) (Ref. 1) deposition of InAs under conventional (As-stabilized) MBE conditions. Although it was assumed that the islanding is accompanied by the formation of misfit dislocations, there is now a consensus on the point that initially formed islands are coherently strained to substrates and are free of dislocations.[2,3] In contrast, on the (111)*A*- and (110)-oriented substrates, InAs grows in a layer-by-layer mode, the lattice strain being relaxed by introducing misfit dislocations.[4,5] Such characteristic growth features on these surfaces can be explained by considering a lower formation energy of misfit dislocations than in the case on the (001) surface.[6] However, interestingly, it has been reported that InAs grows in a layer-by-layer mode on GaAs(001) under As-deficient MBE conditions.[7–11]

Snyder, Mansfield, and Orr[7] first reported that the $In_xGa_{1-x}As$ film grows in the layer-by-layer mode at a low temperature of 320 °C. The lattice strain in the layer-by-layer grown film is hardly relaxed even after the growth of 50 BL.[7] It has been shown that islanding of InAs is also suppressed under the In-stabilized growth conditions even at a higher temperature of 520 °C, but the lattice constant is gradually increased as the growth proceeds.[8] Similar results were reported by Ploog and co-workers,[9,10] and Xue *et al.*[11] Thus, the question arises as to whether the strain in the layer-by-layer growing film is relaxed in $In_xGa_{1-x}As$/GaAs(001) systems.

This paper reports a systematic study of the growth processes in $In_xGa_{1-x}As$/GaAs(001) heteroepitaxy $(0 \leqslant x \leqslant 0.5)$ under As-deficient conditions. The $In_xGa_{1-x}As$ film on GaAs(001) grows in the layer-by-layer mode, and is coherently strained to the substrate. We found that a significant amount of In atoms is segregated to the growing surface to form droplets. The depletion of In in the growing film, which is caused by the In segregation, reduces the lattice strain at the interface, so that the coherent growth of $In_xGa_{1-x}As$ is promoted.

## II. EXPERIMENT

The experiments were performed in a dual-chamber MBE system equipped with the reflection high-energy electron diffraction (RHEED), total-reflection-angle x-ray spectroscopy (TRAXS), x-ray photoelectron spectroscopy (XPS), and scanning tunneling microscopy apparatuses. A detailed description of the apparatuses and surface cleaning treatments for the GaAs(001) substrate has been given in our previous papers.[12] Thin $In_xGa_{1-x}As$ films were grown on the GaAs(001)-(2×4) surface at a substrate temperature $(T_{\mathrm{sub}})$ of 350 °C. The deposition rate of $In_xGa_{1-x}As$ was 0.01 bilayer (BL)/s, which was calibrated by RHEED intensity oscillation measurements for the homoepitaxy on the (001)-oriented InAs and GaAs substrates. The beam-equivalent pressure of $As_4$ was controlled to $5.0\times10^{-9} \sim 2.5\times10^{-7}$ Torr. TRAXS measurements, which is a method for detecting characteristic x rays emitted from a solid surface excited by a RHEED beam,[13,14] were performed with the incident electron beam of 15 keV. The take-off angle of the detected x ray was fixed at 0.76°, which is close to the calculated critical angles for total reflection of the In $L\alpha$ line (3.28 keV) by bulk GaAs (0.747°) and bulk InAs (0.769°). The glancing angle of the incident electron beam was 2.0°. The measurements were performed every 90 s during $In_xGa_{1-x}As$ growth in real time. XPS measurements were carried out using monochromatic Al $K\alpha$ radiation (1486.6 eV). Photoelectrons were detected at an emission angle of 35° from the surface.

## III. RESULTS AND DISCUSSION

$In_xGa_{1-x}As$ $(x=0.5)$ grows in the SK mode at 350 °C when the As flux $(P_{\mathrm{As}})$ is higher than $2.0\times10^{-8}$ Torr, which

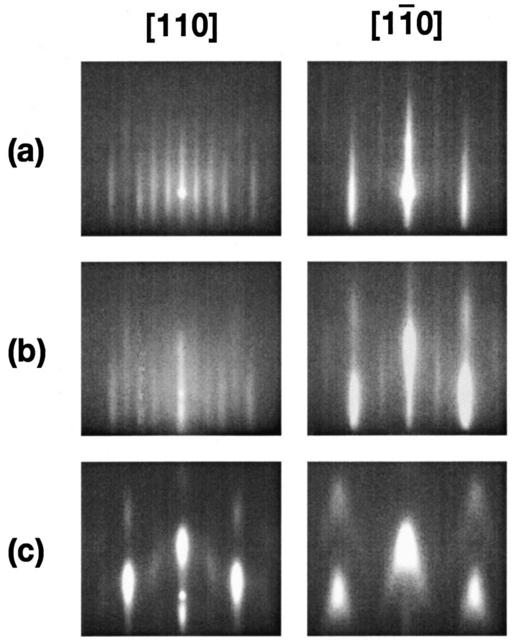

FIG. 1. RHEED patterns observed along the $[1\bar{1}0]$ direction of the $In_xGa_{1-x}As$ films (film thickness = 10 BL) growing on the GaAs(001) substrates under the As flux of $1.0\times10^{-8}$ Torr (a), $1.5\times10^{-8}$ Torr (b), and $2.0\times10^{-8}$ Torr (c).

is evidenced by the changes in RHEED patterns from streaks to spots during the growth [Fig. 1(c)]. On the other hand, for $P_{As}=1.0\times10^{-8}$ Torr [flux ratio of $As_4/(In+Ga)\approx2.5$], the growing surface showed the $(4\times2)$ RHEED pattern throughout the growth, in which no spot pattern originating from $In_xGa_{1-x}As$ islands was observed, as shown in Fig. 1(a). Figure 2 shows the variations in the in-plane lattice constant $d_{110}$ of the growing $In_xGa_{1-x}As$ film, which were measured from the distance between the 0 1 and 0 $\bar{1}$ reflections in the RHEED patterns. The glancing angle of the incident electron beam was ~1.0°. The $d_{110}$ value remained almost unchanged throughout the growth for $P_{As}=1.0\times10^{-8}$ Torr (open circles), while the lattice strain begins to relax after 400 s growth for $P_{As}=2.0\times10^{-8}$ Torr (open squares).

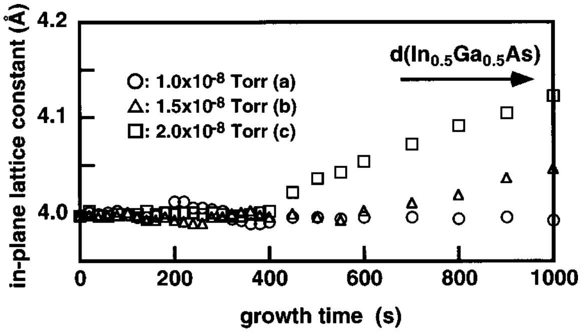

FIG. 2. In-plane lattice constant of $In_xGa_{1-x}As$ on GaAs(001) as a function of the growth time. The As fluxes are $1.0\times10^{-8}$ Torr (a), $1.5\times10^{-8}$ Torr (b), and $2.0\times10^{-8}$ Torr (c).

Recent studies have shown that the initial strain relaxation in growing InAs on GaAs(001) is anisotropic.[15] Thus, RHEED measurements were also performed along the another principal azimuth of $[1\bar{1}0]$. While, for $P_{As}\geqslant2.0\times10^{-8}$ Torr, the strain in the $[1\bar{1}0]$ is relaxed more rapidly than in the [110] direction after ~400 s growth, both $d_{110}$ and $d_{1\bar{1}0}$ values for $P_{As}=1.0\times10^{-8}$ Torr are almost all the same as those of the GaAs(001) substrate even after 3000 s growth. From these results, it appears that $In_xGa_{1-x}As$ pseudomorphically grows in a layer-by-layer mode on the GaAs(001) under As-deficient conditions.

The growth rate of $In_{0.5}Ga_{0.5}As$ was measured from the RHEED intensity oscillation profile obtained during the growth. The estimated growth rate is 0.0047 BL/s, which is significantly smaller than the expected value of 0.01 BL/s (see Fig. 5 below). Since there is a deficiency of As molecules at the growth front under low As fluxes, the probabilities of In and Ga atoms being locked by As molecules are reduced.[16] Thus, the present result indicates that only ~50% of group-III atoms are consumed in the formation of the $In_xGa_{1-x}As$ lattice under As-deficient conditions. Since the present growth experiment on the (001) substrate were performed at a low temperature of 350 °C, the desorption of In and Ga is unlikely to occur during the growth. This prompted us to investigate chemical composition of the growing film.

Information about chemical composition of the growing surfaces is obtained by TRAXS measurements. Figure 3(a) shows variations of In $L\alpha$ intensity measured during $In_{0.6}Ga_{0.5}As$ growth on GaAs(001) under the As flux of $1.0\times10^{-8}$ Torr. Also shown in Fig. 3(a) are the result on the (111)$A$-oriented substrates of GaAs under the conventional MBE condition ($T_{sub}=450$ °C and $P_{As}=\sim2.5\times10^{-7}$ Torr). The In $L\alpha$ intensity for (001) growth increases more rapidly than that for the (111)$A$ orientation. We have already reported that $In_{0.5}Ga_{0.5}As$ grows in the layer-by-layer mode on GaAs(111)$A$, with a very small amount of In atoms being segregated on the growing surface.[17] Thus, the present TRAXS result suggests that more In atoms are segregated to the growth front, when $In_xGa_{1-x}As$ grows on GaAs(001) under As-deficient conditions.

In order to obtain details about the surface segregation of In, we performed XPS measurements. For the measurements, the samples were quenched just after the growth interruption and subsequently transferred to the XPS chamber, so as to avoid unintentional changes in surface features. Figure 3(b) shows the In-3$d$ photoelectron intensities as a function of the growth time. The In 3$d$ intensities for the (001) orientation manifest themselves in lower rises than those for the (111)$A$ orientation, in clear contrast with the TRAXS results shown in Fig. 3(a). Since, in general, the escape depth of photoelectrons (< 30 Å) is significantly smaller than that of characteristic x rays (> 1 $\mu$m), the In 3$d$ photoelectrons generated at deeper layers are hardly detected. Thus, if we assume that the In atoms, which segregate to the growing surface, form large islands, the results in Figs. 3(a) and 3(b) can be explained in a consistent manner.

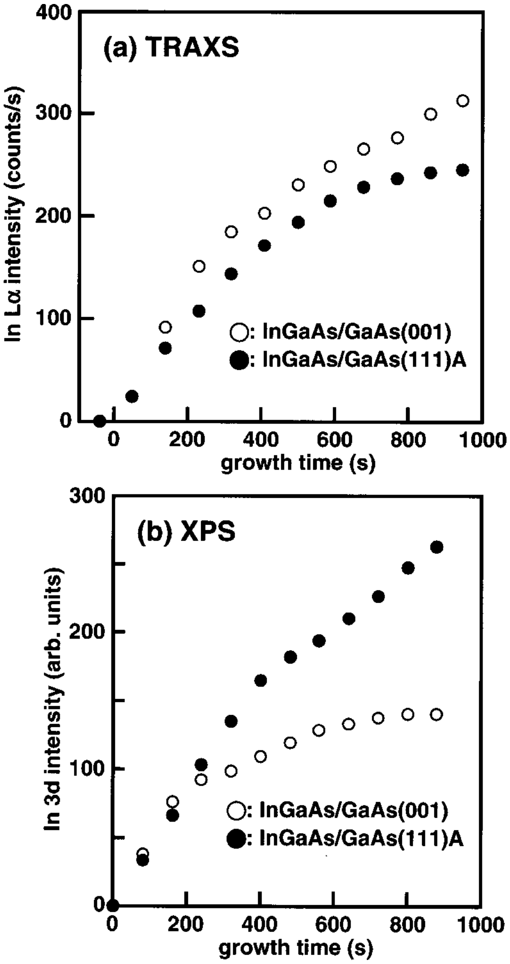

FIG. 3. Variations in the intensities of the In $L\alpha$ line (a) and In $3d$ line (b) as a function of the growth time of $In_xGa_{1-x}As$ on GaAs(001).

Figure 4 shows a typical scanning electron microscopy (SEM) image obtained after the growth of $In_{0.5}Ga_{0.5}As$ for 1000 s (nominal film thickness is 10 BL). Large islands were observed with a density of $\sim 1 \times 10^5$ cm$^{-2}$. The size of these islands was estimated from atomic-force microscopy images (not shown) and was found to be $1.6 \pm 0.1$ $\mu$m in diameter and $0.6 \pm 0.1$ $\mu$m in height. Since the island density is sufficiently low, our RHEED observations could detect the flat regions having $(4 \times 2)$ reconstructions between islands, as shown in Fig. 1(a).

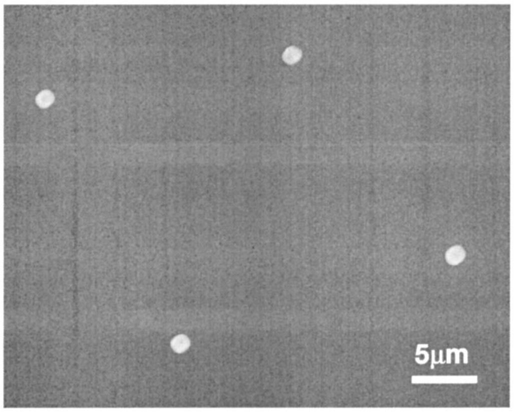

FIG. 4. Typical SEM image of the $In_{0.5}Ga_{0.5}As$ film on the GaAs(001) substrate.

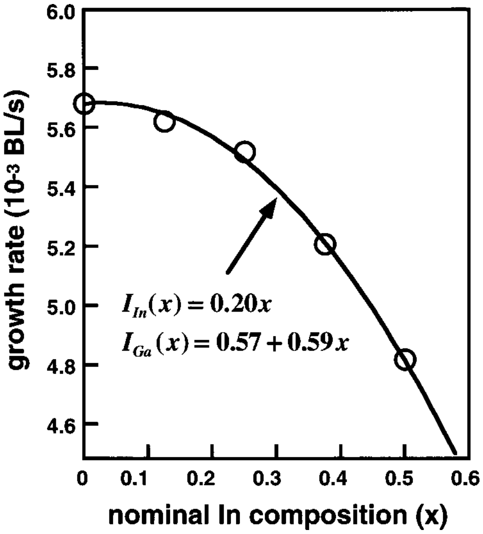

FIG. 5. The growth rate of $In_xGa_{1-x}As$ on GaAs(001) under As-deficient conditions as a function of the $x$ value. The substrate temperature is fixed at 350 °C.

The In-$3d$ photoelectrons generated at inner part of the large islands shown in Fig. 4 are hardly detected because of their small escape length, as mentioned above: photoelectrons that undergo inelastic scattering leave the solid surface with lower energies and contribute to a background signal. On the other hand, x rays from the inner atoms in the islands can reach the detector without considerable attenuation. Here, we note that the In $L\alpha$ x rays could be generated at inner part of the large islands, although the penetration length of incident electrons (15 keV) is not much greater than the escape length of the In-$3d$ photoelectrons. Since the incident electrons have the energy significantly larger than that of the In $L\alpha$ line (3.28 keV), it is likely that the majority of electrons are sufficiently energetic to excite the In $L\alpha$ x rays after losing their energy.

Since the TRAXS and XPS results showed that a significant amount of In atoms segregate to the growth front and form islands, it is reasonable to consider that the islands seen in Fig. 4 are mainly composed of In atoms. In order to confirm this, we performed growth experiments at a fixed As flux of $1.0 \times 10^{-8}$ Torr, changing the nominal In composition in the $In_xGa_{1-x}As$ film. Figure 5 shows the growth rate of $In_xGa_{1-x}As$ on GaAs(001) as a function of the $x$ value, which is estimated on the basis of the RHEED intensity oscillation measurements. For $x = 0$, i.e., GaAs(001) homoepitaxy, the growth rate is ~57% of the expected value. The growth rate decreases with increasing $x$ value, indicating that the incorporation probability of In in the growing film is significantly smaller than that for Ga.

The data in Fig. 5 can be fitted using a simple model. We have calculated growth rate assuming that the incorporation probabilities of Ga and In are $I_{\mathrm{Ga}}(x)=0.57+0.59\times x$ and $I_{\mathrm{In}}(x)=0.20\times x$, respectively. The estimated growth rate is shown by the solid curve in Fig. 5. It is apparent from these two equations that the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ film contains less In atoms than expected. For example, the incorporation probabilities of Ga and In for $x=0.5$, are $I_{\mathrm{Ga}}(0.5)=0.86$ and $I_{\mathrm{In}}(0.5)=0.10$, respectively, which corresponds to the film composition of $\mathrm{In}_{0.1}\mathrm{Ga}_{0.9}\mathrm{As}$. This reasonably explains why the in-plane lattice constant of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ hardly changes throughout the growth under the As flux of $1.0\times 10^{-8}$ Torr [Fig. 2(a)], because the strain energy at the coherent $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$/GaAs interface decreases with $x$ value. Also, the result in Fig. 5 teaches us that each island consists of 0.7 ML of Ga and 4.5 ML of In. Using these values and the bulk densities of In and Ga, the volume of each island in Fig. 3 is estimated to be $8.2\times 10^{-13}$ cm$^3$. This value is in good agreement with that estimated from SEM image ($\sim 8\times 10^{-13}$ cm$^3$).

As mentioned at the beginning of this paper, the layer-by-layer growth of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ on GaAs(001) has been reported by several groups.[7–11] Snyder, Mansfield, and Orr have shown that the $\mathrm{In}_{0.5}\mathrm{Ga}_{0.5}\mathrm{As}$ film grows in the layer-by-layer mode at a low temperature of 320 °C:[7] the growing $\mathrm{In}_{0.5}\mathrm{Ga}_{0.5}\mathrm{As}$ film has the lattice constant almost identical to that of the GaAs substrate throughout the growth, similarly to the case in this study. Thus, the result in Ref. 7 can also be explained by assuming that a significant amount of In atoms segregate to the growth front, although the actual growth rate has not been given in Ref. 7. It has also been reported that even at a higher temperature of 500–520 °C, InAs grows two dimensionally with the $(4\times 2)$ surface reconstructions when the $\mathrm{As}_4$ to In flux ratio is controlled to 1.8.[8] However, at high temperatures and low As fluxes, In atoms are easily desorbed from the growing surface, which suppresses the formation of thick InAs films, as we have already reported.[16] Thus, it appears likely that the $(4\times 2)$ reconstruction in Ref. 8 is ascribed to the adsorption structure of In on GaAs(001).

On the other hand, Ploog and co-workers have reported that the lattice strain in the layer-by-layer growing InAs film is gradually relaxed under the $\mathrm{As}_4$ to In flux ratio of 0.7–0.9 at a relatively low temperature of 430 °C.[9,10] They have also shown that the actual film thickness is very close to the nominal value,[9] suggesting that no significant In droplet formation occurs. These results are in clear contrast with our results.

At first sight, one may attribute the difference in growth features to the different deposition rates (0.4 BL/s in Refs. 9 and 10, and 0.01 BL/s in this study). This prompted us to perform growth experiments changing the deposition rate from 0.005 to 0.2 BL/s at fixed flux ratios. However, the observed growth rates were about ~50% of the expected values in any experiment at 350 °C.

Next, we have investigated the temperature dependence of the growth rate of $\mathrm{In}_{0.5}\mathrm{Ga}_{0.5}\mathrm{As}$ ($P_{\mathrm{As}}=1\times 10^{-8}$ Torr). As shown in Fig. 6(a), the growth rate increases with decreasing $T_{\mathrm{sub}}$. This means that the incorporation probabilities of In and Ga atoms are sensitive to the population of As molecules at the growth front, because the sticking probability of As molecules decreases with increasing $T_{\mathrm{sub}}$. Indeed, as shown in Fig. 6(b), the growth rate increases as the As flux is increased, and the growth rate is ~0.01 BL/s for $P_{\mathrm{As}}=2\times 10^{-8}$ Torr. Under this condition, the lattice strain is almost relaxed after 1000 s (open squares in Fig. 2). However, as mentioned earlier in this paper, the growth mode changes from layer-by-layer to SK as the As flux exceeds $2.0\times 10^{-8}$ Torr. These results clearly indicate that the increase in the As flux suppresses the surface segregation of In atoms, but induces the formation of the relaxed $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ islands.

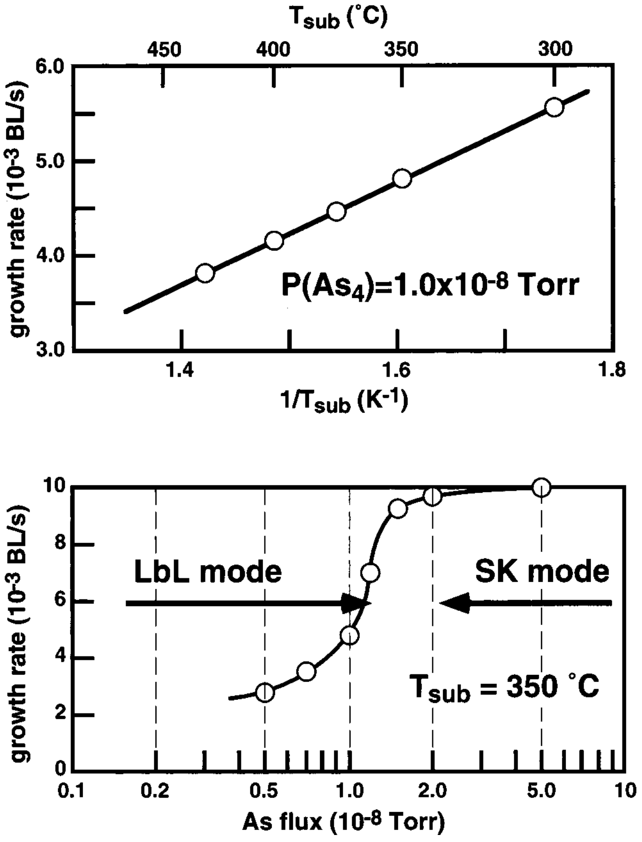

FIG. 6. The growth rate of $\mathrm{In}_{0.5}\mathrm{Ga}_{0.5}\mathrm{As}$ on GaAs(001) as a function of the substrate temperature (a) and the As flux (b).

When the As flux is $\sim 1.5\times 10^{-8}$ Torr, the actual growth rate is close to the nominal value, and the strain in the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ film is gradually relaxed, as shown by open triangles in Fig. 2. The growing surface showed streaky RHEED patterns [Fig. 1(b)] along the [110] direction, while elongated spots are visible along the $[1\bar{1}0]$ direction. Thus it appears that the growth under the As flux of $1.5\times 10^{-8}$ Torr is effective in suppressing the formation of both In droplets and $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ islands, although the ideal layer-by-layer growth could not be realized in this system.

## IV. CONCLUSIONS

We have studied the growth modes of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ on GaAs(001) under As-deficient conditions. The growth rate of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ decreases and increases with increasing $x$ value and As flux, respectively. For $x=0.5$ and the As flux of 1

$\times 10^{-8}$ Torr, only ~10% of In atoms are incorporated in the layer-by-layer-growing film and the rest of In atoms segregate to the growing surface to form droplets. The lattice mismatch at the interface is significantly reduced owing to the In segregation, which allows the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ film to coherently grow on the substrate. The In segregation is suppressed for the growth under higher As fluxes, which is accompanied by the formation of relaxed $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ islands.

## ACKNOWLEDGMENTS

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