Effects of Post Annealing on Self-organized InAs Islands Grown on (001) GaP by Organometallic Vapor Phase Epitaxy

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Abstract

Using atomic force microscopy (AFM) and transmission electron microscopy (TEM), we have investigated effects of post annealing on self-organized InAs islands grown on (001) GaP by low-pressure organometallic vapor phase epitaxy (LP-OMVPE). The morphology of InAs islands without the cap layer depended strongly on the growth temperature, which was related to migration of InAs adsorbed on GaP. Subjected to the annealing at temperatures higher than the growth temperature, the small islands coalesced each other to form larger islands. The resultant morphology was quite similar to that of the samples grown at the annealing temperature. Cross-sectional TEM observation was carried out on the buried InAs islands. Almost relaxed InAs islands buried by the GaP cap layer with stacking faults were clearly observed. The size and density of the buried InAs islands were similar to those in the samples post-annealed at the growth temperatures of the GaP cap layer, suggesting that the change on the InAs islands was induced during the increase in the substrate temperature for the GaP cap-layer growth. These results indicated that the growth temperature for the GaP cap layer played an important role in fabricating the sandwich structures with frozen small InAs islands.

Introduction

InAs/GaP heterostructures are very attractive since the band-offset in the conduction band is expected to be as large as 1 eV [1]. Furthermore, there is a large mismatch of 11% in lattice constants between InAs and GaP, which modifies their energy band structures. However, the large lattice-mismatch induces difficulties in growth processes. Therefore, there have been few reports on the growth of two-dimensional (2D) InAs on GaP. Chang *et al.* reported that the InAs epilayer grown on (001) GaP by molecular beam epitaxy is almost fully relaxed via the formation of an array of 90°-type edge dislocations at the heterointerface[1, 2]. Such defects at the heterointerface would be fatal in the application to practical devices.

Formation of self-organized islands due to strain has been well studied in some strained systems. Many papers have been published on InAs islands on GaAs substrates [3, 4], InGaAs islands on GaAs substrates [5, 6], InAs islands on InP substrates [7-13], and InGaP islands on GaP substrates [14]. Recently, we reported successful growth of self-organized InAs islands on (001) GaP by LP-OMVPE [15]. InAs islands buried by a cap layer are fundamental in their device application. However, optimum growth temperature of GaP is generally higher than that of formation of small size InAs islands. Therefore, InAs islands suffer from annealing during the increase in the substrate temperature for the growth of the GaP cap layer.

In this paper, we report AFM and cross-sectional TEM studies of InAs islands subjected to post annealing. It is demonstrated that the post annealing results in an increase in size of the InAs islands and a decrease in their density, which are due to the coalescence of the InAs islands. In addition, the cross-sectional TEM observation on buried InAs islands reveals the defects in the GaP cap layer and at the heterointerface.

Experimental

The growth of InAs island was performed on undoped (001) GaP substrates by a LP-OMVPE system with a cold-wall quartz 4-barrel reactor [16]. In the growth system, different group-V sources were supplied into different barrels of the reactor for suppression of intermixing between the sources. Tertiarybutylphosphine (TBP), tertiary-butylarsine (TBAs), trimethylgallium (TMGa) and trimethylindium (TMIn) were used as source materials. The reactor pressure was fixed at 76 Torr. A 80 nm-thick GaP buffer layer was grown at 650℃, followed by lowering the substrate temperature for InAs island growth. The InAs islands were grown at 450-650℃. After the InAs island growth, the substrate temperature was immediately increased in $TBAs/H_2$ ambient for the growth of a GaP cap layer.

Morphological studies of InAs islands without the cap layer were performed by AFM (Nanoscope IIIa) in air with tapping mode. Cross-sectional TEM observation of buried InAs islands was carried out by Hitachi H-800 electron microscope, operating at an accelerating voltage of 200 kV. The specimens for [110] cross-sectional TEM observation were prepared using conventional procedures, i. e., 6 keV argon ion milling on a liquid nitrogen-cooled stage after mechanical polishing.

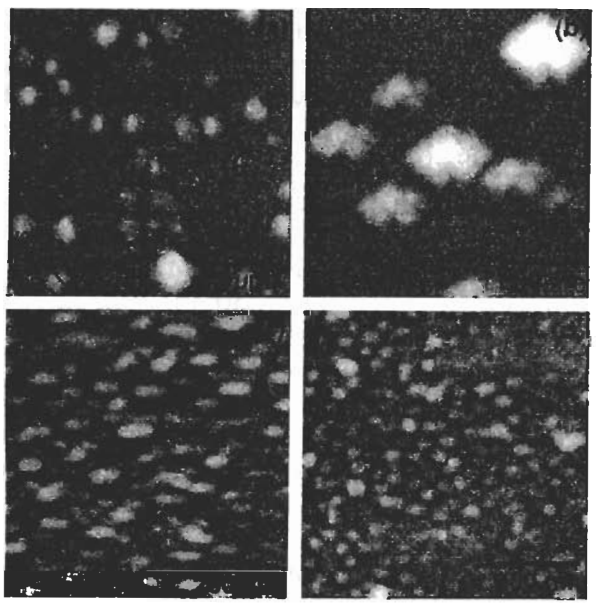

Fig. 1 AFM images of the samples grown at (a) 650℃, (b) 550℃ (note the change of scale), (c) 500℃ and (d) 450℃, respectively. The crystallographic orientation is shown in (a).

Results and discussion

AFM images of the samples grown at 450-650℃without a GaP cap layer are shown in Fig. 1. The morphology of the InAs islands depends strongly on the growth temperature. At 650℃ [Fig. 1(a)], the InAs islands are about 50 nm in height and 400 nm in width, and the areal density is approximately $3.2x10^8$ cm^{-2}. The edges of the islands are parallel to the <100>. As the growth temperature is decreased to 550℃, the size of the islands decreases to about 50 nm in height and 150 nm in width, as shown in Fig. 1(b). The density of the islands increases to $1.1x10^9$ cm^{-2}. Furthermore, it is clearly observed that each island consists of a few grains, suggesting that the islands are generated by the coalescence among small islands.

Figure 1(c) shows the AFM image of InAs islands grown at 500℃. The surface morphology changes drastically. The islands are elongated along the $[1\bar{1}0]$, resulting in an oval shape. The width of the islands is about 80 nm along the $[1\bar{1}0]$ and about 50 nm along the [110]. The height is about 12 nm. The density of the islands increases significantly to $2.2x10^{10}cm^{-2}$. At 450℃, as shown in Fig. 1(d), the shape of the islands remains similar to that at 500℃. The size of the islands slightly reduces; about 55 nm along the$[1\bar{1}0]$, about 40 nm along the $[1\bar{1}0]$ and about 5 nm in height. The islands are $4.8x10^{10}$ cm^{-2} in areal density. The drastic change in surface morphology against the growth temperature is related to migration of InAs adsorbed on GaP. At temperatures higher than 550℃, the adsorbed InAs can reach stable sites, leading to the formation of the islands with facets. The migration of InAs is, on the other hand, not so sufficient to reach the stable sites at temperatures lower than 500℃. The elongated shape of the islands observed in the samples grown at the low temperatures indicates an anisotropy in migration length.

Based on the AFM observation of the as-grown InAs islands, the growth temperature to obtain the smallest islands with the highest areal density is concluded to be 450℃. Since the GaP growth is not possible at 450℃, the growth temperature for the GaP cap layer must be raised after the InAs islands formation. The lowest growth temperature for the GaP growth is 500℃ and the optimum growth temperature is 650℃ in our growth system. As mentioned below, we fabricated GaP/InAs islands/GaP sandwich structures by capping the 450℃ - grown InAs islands with GaP at 500 and 550℃, respectively. In this procedure, the InAs islands suffer from annealing during the increase in the substrate temperature for the growth of the GaP cap layers. Therefore, it is interesting to investigate effects of the post annealing on the InAs islands.

Figure 2 shows an AFM image of the sample grown at 450℃ and then annealed in $TBAs/H_2$ ambient for 2min at 550℃. The surface morphology drastically changes by such short

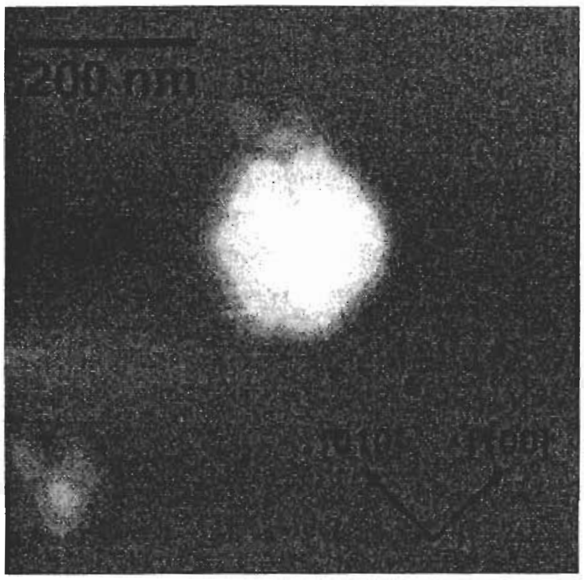

Fig. 2 AFM image of the sample grown at 450℃ and then annealed at 550℃.

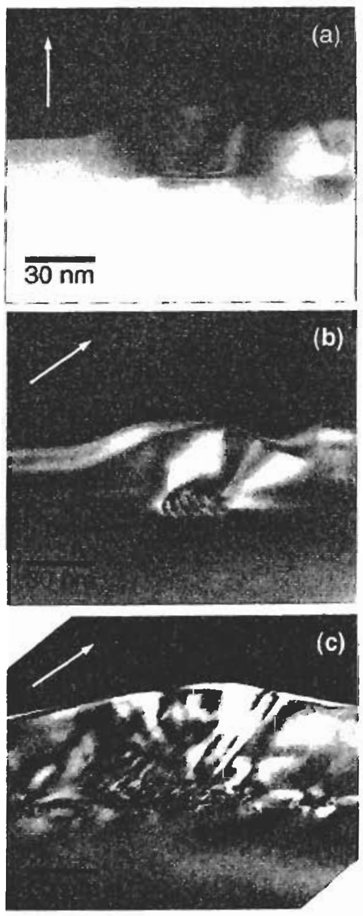

Fig. 3 [110] cross-sectional dark field TEM images of buried InAs islands. GaP cap layers were grown at (a) 500℃ (g=[002]), (b) 500℃ (g=$[1\bar{1}1]$) and (c) 550℃ (g=[111]) (note the change of scale), respectively. The Arrows indicate the direction of the diffraction vector.

annealing at 550℃ and it is similar to that of the sample grown at 550℃ [Fig. 1(b)]. The InAs islands are enlarged to about 50 nm in height and 200 nm in width. Each island exhibits clear fingerprints of the coalescence of small islands. Total amount of InAs remains almost constant before and after the post annealing, suggesting no evaporation of InAs during the post annealing.

In order to obtain further information on GaP/InAs islands/GaP sandwich structures, we have performed cross-sectional TEM observation of buried InAs islands. Figure 3 shows dark field cross-sectional TEM images of the InAs islands which were grown at 450℃ and buried by a GaP cap layer at 500 and 550℃, respectively. Figure 3(a) and (b) show the cross-sectional TEM images of the same buried InAs island with different g ((a) g=[002] and (b) g=$[1\bar{1}1]$). A thin wetting layer and fringes are clearly observed for both samples. The fringes are moiré fringes, originating from two overlapped crystals with different lattice constants, because the direction of the fringes is almost longitudinal to that of g. Therefore, the area of these fringes corresponds to the buried InAs island. As seen in Fig. 3(b), InAs islands buried at 500℃ are 10 nm in height, 30 nm in width and 2.4×10^4 cm^{-1} in line density. At 550℃, on the other hand, the size of InAs islands increases drastically to 30 nm in height and 150 nm in width, while the line density decreases to 1×10^5 cm^{-1}, which is shown in Fig. 3(c). The size and density of the InAs islands buried at 550℃ are coincident with those in the sample grown at 450℃ and annealed at 550℃ [Fig. 2], suggesting that the InAs islands coalesce each other to be large during the increase in the substrate temperature from 450℃ to 550℃ for the growth of the GaP cap layer. Furthermore, compared with InAs islands grown at 450℃ [Fig. 1(d)], the InAs islands are drastically enlarged by being buried at 550℃ rather than 500℃. This result indicates that effects of temperature for the GaP cap-layer growth on the InAs islands significantly change between 500℃ and 550℃. Similar behaviors are observed in Fig. 1.

The moiré fringes taken under g=$[1\bar{1}1]$ two-beam conditions exhibit 3.79 nm (500℃) and 3.86 nm (550℃) in spacing [Fig. 3(b) and (c)]. The parallel moiré fringe spacing D is theoretically given by the expression $D = | g_{InAs} - g_{GaP} |^{-1}$. Here g_{InAs} and g_{GaP} are reciprocal lattice vectors of InAs and GaP, respectively. Using the spacing determined experimentally, the lattice constants of buried InAs islands are calculated to be 5.945 Å(500℃) and 5.935 Å (550℃). These values mean 98 % of degree of strain relaxation, suggesting that the InAs islands are almost fully relaxed by the generation of dislocations at the heterointerface between GaP and InAs islands.

Some fringes not due to moiré fringes are also observed in the GaP cap layer for both samples. The measured angle between these fringes and (001) surface agrees with that of {111} plane and (001) surface. Thus, the fringes correspond to stacking faults on the {111} plane. We have not succeeded in the observation of the photoluminescence from buried InAs islands even at 4.2K. Based on the TEM observation, no luminescence from buried InAs islands is well understood by the nonradiative recombination of photogenerated carriers at the stacking faults and dislocations. In order to obtain in-depth knowledge on the stacking faults and dislocations, high-resolution TEM (HRTEM) is now in progress and the results will be reported elsewhere. The defects might be induced by large InAs islands. Further study should be required to fabricate GaP/InAs islands/GaP sandwich structures with frozen small InAs islands.

Conclusions

Effects of post annealing on self-organized InAs islands grown on (001) GaP by LP-OMVPE have been investigated by using AFM and cross-sectional TEM observation. The morphology of InAs islands without the cap layer depended strongly on the growth temperature, which is related to migration of InAs adsorbed on GaP. The smallest InAs islands with the highest areal density were successfully obtained at 450℃. Subjected to the annealing at temperatures higher than the growth temperature of 450℃, the small islands coalesced each other to form larger islands. The resultant morphology was quite similar to that of the samples grown at the annealing temperatures. Cross-sectional TEM observation was carried out on GaP/InAs islands/GaP sandwich structures fabricated by capping the 450℃-grown InAs islands with GaP at 500 and 550℃, respectively. Almost relaxed InAs islands buried by the GaP cap layer with stacking faults were clearly observed. The size and density of the buried InAs islands were quite different from those of the 450℃ -grown islands, and were similar to those in the samples post-annealed at the growth temperatures of the GaP cap layer, suggesting that the change on the InAs islands is induced during the increase in the substrate temperature for the GaP cap-layer growth. These results indicated that the growth temperature for the GaP cap layer plays an important role in fabricating the practical sandwich structures with frozen small InAs islands.

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