Extended Abstracts of the 1994 International Conference on Solid State Devices and Materials, Yokohama, 1994, pp. 169-171

Growth Mode Transition in GaAs/GaP(001) by Molecular Beam Epitaxy

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The growth mode transition in GaAs/GaP heteroepitaxy was studied by means of reflection high-energy electron diffraction pattern analysis. The two phenomena characteristic to the strain relaxation at the edges of the island, the lattice parameter oscillation and the variation of the critical thickness, were observed in GaAs/GaP. We found the transformation of the film structure and the relaxation of the misfit strain even after the stop of growth. The dependence of the critical thickness on the growth rate is explained by assuming two processes at the surface.

Introduction

Considerable interest in semiconductor materials research has been devoted to the growth of heterostructures with lattice mismatched epitaxial layers in order to obtain new characteristics. Several difficulties are reported for the lattice mismatched epitaxy. Predictions of the critical film thickness were reported by van der Merwe[1)], Matthews and Blakeslee[2)], and People and Bean[3)]. They, however, did not refer to the growth mode transition from layer-by-layer growth to three-dimensional island growth. Recently, Snyder et al.[4)] proposed a model which was based on an elastic relaxation at edges of coherent islands in InGaAs heteroepitaxy onto GaAs (001). The transformation from uniformly strained layer to elastically relaxed island which is energetically favored, results in the growth mode transition. They also assert that the critical thickness of the growth mode transition is determined by kinetically controlled formation of the islands[5)].

We have investigated the growth mechanism of GaAs heteroepitaxy onto GaP (001) by molecular beam epitaxy (MBE). We reported that a growth mode transition from layer-by-layer to three-dimensional island occurred around 2ML and an anisotropic shape of the islands is observed by scanning tunneling microscopy[6)]. We also found an anisotropic relaxation of the misfit strain[7)]. These results are consistent with the prediction of the model proposed by Snyder. In this paper we study a kinetic control of the critical thickness of the growth mode transition.

Experiments

The growth was carried out by a conventional MBE method using ULVAC MBC-100 with solid sources. Prior to the heteroepitaxial growth of GaAs, a GaP buffer layer of about 500nm in thickness was grown at 600°C to obtain a well-defined substrate with an atomically flat surface. It was confirmed with reflection high-energy electron diffraction (RHEED) that substrate surface was not degraded by a change of group V beam material from arsenic to phosphorus. The growth mode transition and strain relaxation were investigated by means of analyses of the RHEED pattern.

Results and Discussion

N. Grandjean and J. Massies reported in strained InGaAs/GaAs heteroepitaxial growth that the lattice parameter of the grown film oscillated in opposite phase to RHEED oscillation during layer-by-layer growth regime prior to the growth mode transition[8)]. They stated that the lattice parameter oscillation was caused by the elastic relaxation at free edges of two-dimensional nuclei. If this is also true in GaAs/GaP heteroepitaxy, similar oscillation in the lattice parameter should appear. We examined the lattice parameter oscillation in GaAs/GaP. The experimental condition was selected to increase sensitivity to the topmost layer: the incidence angle of electron beam was less than 1°, and the lattice parameter was calculated from the separation of RHEED diffraction spots around the Laue zone. The growth rate was also lowered to 0.04ML/s in order to form larger two-dimensional nuclei. The intensity of the specular spot and the in-plane lattice parameter are shown in Figs. 1 (a) and (b) respectively, as a function of average layer thickness. The lattice parameter oscillates in opposite phase to the intensity oscillation of the specular spot. These results have the same tendency as reported in InGaAs/GaAs heteroepitaxy except the lattice parameter being not minimum for the deposition corresponding to each monolayer. This may be caused by partial exchange of phosphorus atom with arsenic at the surface of GaP substrate. The partial exchange is caused by an exposure of

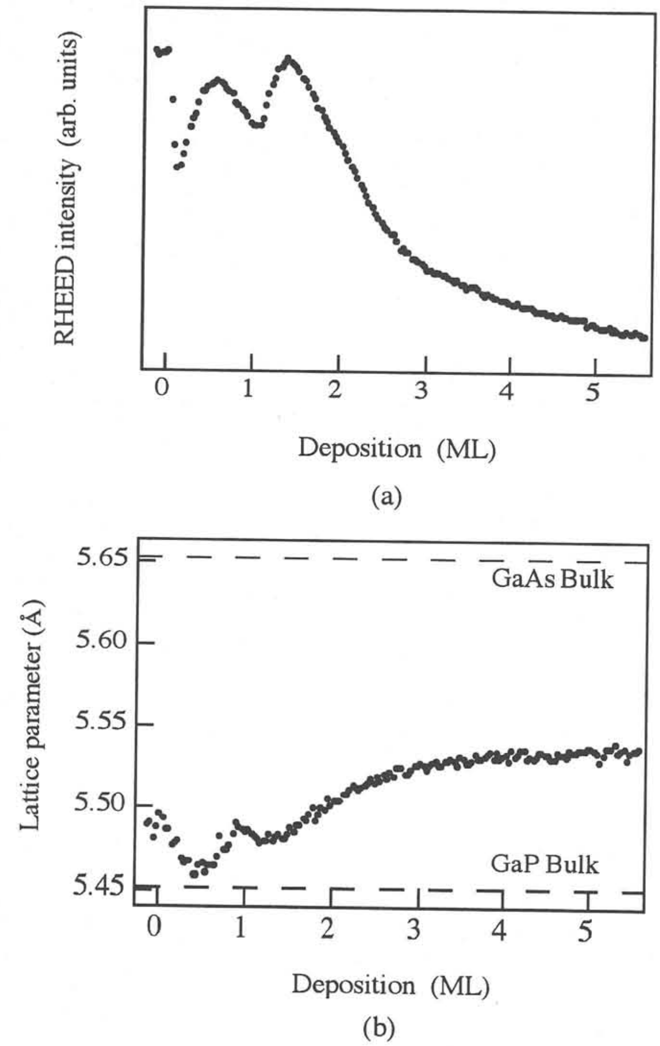

Fig. 1 (a) Intensity of specular spot and (b) in-plane lattice parameter of GaAs growth on GaP(001). The growth rate is 0.04ML/s and the substrate temperature 600°C.

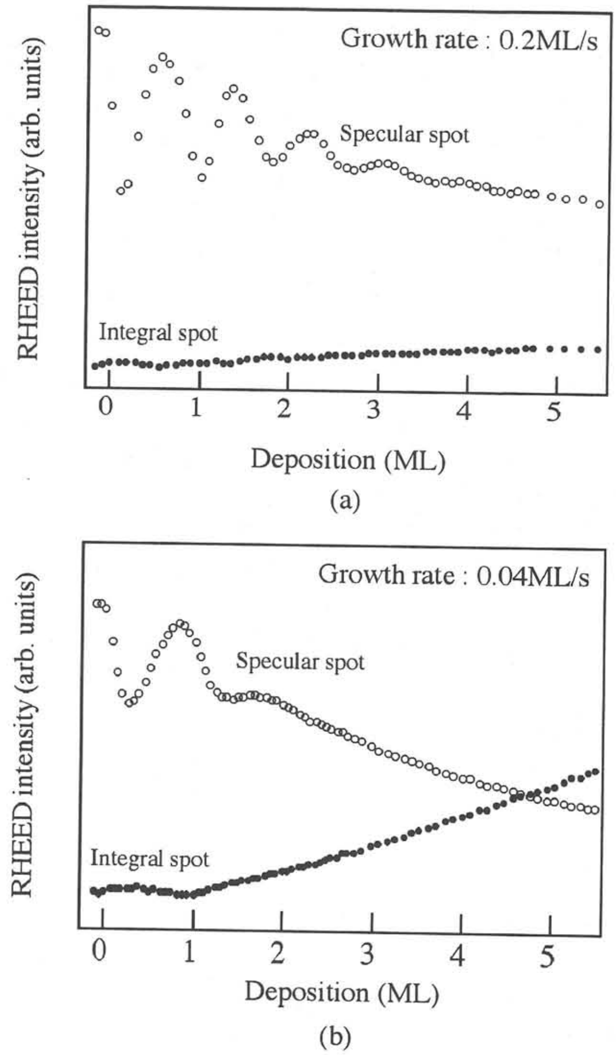

Fig. 2 Intensities of specular and integral spot for the growth rate of (a) 0.04ML/s and (b) 0.2ML/s. The substrate temperature is 600°C in each case.

GaP substrate to arsenic beam prior to GaAs heteroepitaxy to maintain the surface stoichiometry. However the coincidence of the maximum of lattice parameter to the minimum of specular spot intensity shows that the oscillation of lattice parameter is caused by the variation in the surface step density. Therefore, the present result is consistent with the model proposed by Snyder.

N. Grandjean and J. Massies also pointed out that the critical thickness of the growth mode transition in InGaAs/GaAs heteroepitaxy depended on the growth rate due to the kinetic evolution[8]. We also examined this dependence of the critical thickness on the growth rate in GaAs/GaP heteroepitaxy. Figure 2 (a) and (b) show the intensity oscillations of the specular spot and the intensity of the integral spot for 0.2ML/s and 0.04ML/s, respectively. The onset of the GaAs growth is in layer-by-layer mode characterized by the intensity oscillation of the specular spot. The oscillation damps and the intensity of integral spot start to increase around 2ML for the lower growth rate. On the other hand, five cycles of the intensity oscillation are observed and the intensity of integral order spot increases more gradually for the higher growth rate. These results indicate that the critical thickness increases with the increase of the growth rate as reported in InGaAs/GaAs.

The increase of the critical thickness of the growth mode transition was explained by kinetically controlled evolution of the island. However, the details of the island evolution process have not been understood. We examined the process of the island evolution as follows: the growth of GaAs was stopped at the onset of the growth mode transition, and the transformation from layer to island was studied by an analysis of intensity profile of the RHEED pattern. Figure 3 shows the intensity profile of RHEED pattern along the reciprocal lattice rod after 6ML growth. The substrate temperature was kept at 600°C and the surface of the film was exposed to arsenic beam during and after the growth to maintain the surface stoichiometry. The change of the intensity profile with time reveals that the structure of GaAs film is unstable on GaP substrate. The shoulder at the specular spot position just after the growth reflects the existence of the layer structure. The decrease of the intensity at the specular spot po-

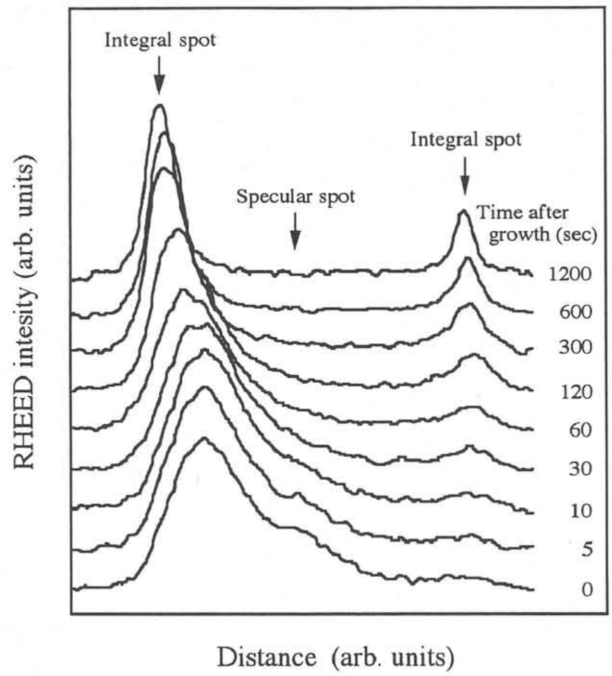

Fig. 3 RHEED intensity profiles along the reciprocal lattice rod after the 6ML growth of GaAs on GaP with growth rate of 0.3 ML/s. The substrate temperature is kept 600°C during and after the GaAs growth.

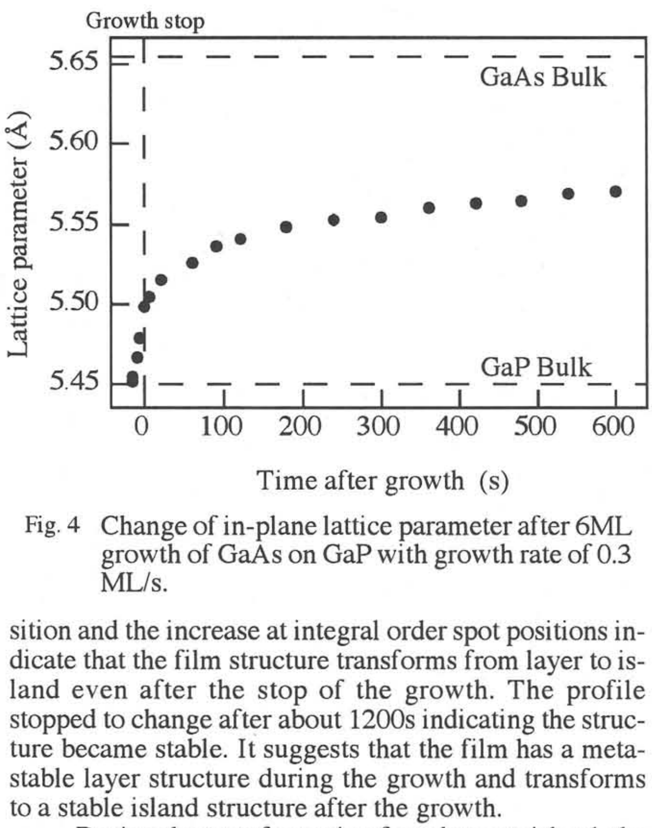

Fig. 4 Change of in-plane lattice parameter after 6ML growth of GaAs on GaP with growth rate of 0.3 ML/s.

sition and the increase at integral order spot positions indicate that the film structure transforms from layer to island even after the stop of the growth. The profile stopped to change after about 1200s indicating the structure became stable. It suggests that the film has a metastable layer structure during the growth and transforms to a stable island structure after the growth.

During the transformation from layer to island, the in-plane lattice parameter of the grown film was measured as a function of time after the growth. The result is shown in Fig. 4. The lattice parameter is calculated from the separation of RHEED diffraction streaks at integral order positions for the purpose of measuring the lattice parameter of the islands. The lattice parameter increases toward that of bulk-GaAs during and after the stop of growth. This result indicates that a strain of the film relaxes with the transformation of the film structure. The formation of island relaxes the misfit strain due to the elastic relaxation at the free edges of islands. Present results indicate that the film has a metastable strained layer structure during the growth and transforms into relaxed island structure which have lower energy.

Kinetic control of the critical thickness is explained by assuming two processes at the surface. The growth process consists of (i) the formation of a metastable layer structure by migration of gallium atom supplied to the surface and its incorporation to neighboring kink sites, and (ii) the transformation of the film structure from layer to island by migration of metastable layer. The rate of the former process is mainly dominated by the arrival rate of gallium atoms, while the rate of the latter process by the substrate temperature. The increase of the critical thickness of the growth mode transition is explained that a surpassing of the formation rate of layer over that of island causes the increase of the critical thickness because the increase of the growth rate enhances the formation rate of the metastable structure.

Conclusion

The growth mode transition in GaAs/GaP heteroepitaxy was studied by means of analyses of the RHEED pattern. The two phenomena characteristic to the strain relaxation at the edges of the island, the lattice parameter oscillation and the variation of the critical thickness of the growth mode transition, were observed in GaAs/GaP. We found the transformation of the film structure and the relaxation of the misfit strain even after the stop of growth. Increase of the critical thickness with its growth rate is explained by assuming two processes at the surface: (i) the formation of a metastable layer structure, and (ii) the island formation by migration of the metastable layer. The increase of the critical thickness with the growth rate is due to a surpassing of the formation rate of layer over that of island because the increase of the growth rate enhances the formation rate of the metastable layer.

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