Coherent multiatomic step formation on GaAs (001) vicinal surfaces by MOVPE and its application to quantum well wires

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Abstract. Coherent multiatomic steps on vicinal (001) GaAs surfaces during metalorganic vapor phase epitaxial (MOVPE) growth are investigated by atomic force microscopy (AFM). AFM images show coherent multiatomic steps with extremely straight edges over a several micron scale. The average spacing of multiatomic steps depends on growth temperature, growth rate and AsH_3 partial pressure. Similar multiatomic steps also appear on GaAs substrate surfaces after thermal treatment under AsH_3/H_2 atmosphere at temperatures higher than 700°C. Furthermore, we fabricate GaAs quantum well wires (QWWs) on these coherent multiatomic steps. Locally thick GaAs, that is, QWWs are formed at corners of multiatomic steps. Photoluminescence spectra of QWWs show energy shift caused by the QWWs formation. These results suggest that self-organized QWWs can be formed uniformly on coherent multiatomic steps.

1. Introduction

Recently, a novel quantum well wire (QWW) fabrication method using *multiatomic* steps has been demonstrated by metalorganic vapor phase epitaxy (MOVPE) on GaAs (001) vicinal surfaces [1], and molecular beam epitaxy (MBE) on GaAs (110) vicinal surfaces [2]. Similar self-organized QWW structures were also fabricated by using *monoatomic* steps on vicinal (001) GaAs surfaces during MOVPE growth [3] and MBE growth [4]. These *in situ* self-organizing fabrication methods are very promising because high density nanometer-size QWWs can be formed without any damage-introducing processes such as lithography and dry etching, and the size of QWW can be controlled by only adjusting the crystal growth conditions. Therefore, from this point of view, understanding of the behavior and mechanism of the multiatmoic step formation on vicinal surface is very important for application to QWWs using multiatomic steps. Recently, multiatomic step formation phenomena on GaAs (001) vicinal surfaces have been investigated after the MOVPE growth [5-7] and the thermal treatment in both ultra high vacuum (UHV) [8] and AsH_3/H_2 atmosphere [9].

In this paper, we report on the formation of coherent multiatomic steps during MOVPE growth and thermal treatment under AsH_3/H_2 atmosphere. Furthermore, we report on the successful formation and optical characterization of self-organized QWWs using coherent multiatomic steps on GaAs (001) vicinal substrates.

2. Experimental Procedure

The substrates were on GaAs (001) misoriented by 1.0°-5.0° towards [110] and $[\bar{1}10]$ directions and singular (001) GaAs as a reference. A horizontal low-pressure MOVPE system with triethylaluminum (TEAl), triethylgallium (TEGa) and AsH_3 as source materials was used. The total gas flow rate was 3.0 *l*/min, and the working pressure during crystal growth was 76 Torr.

First, a 40-period AlAs/GaAs superlattice buffer layer was grown on the vicinal substrate in order to form the initial surface with *monoatomic* steps. Then, a thick GaAs buffer layer was grown at 600°C-750°C to form the *multiatomic* steps with equal spacing terraces. Multiatomic steps were characterized by atomic force microscopy (AFM) in air.

AlGaAs/GaAs/AlAs single quantum well (SQW) structures were also grown on the GaAs buffer layer with coherent multiatomic steps at 650°C. Multiatomic step edges on the GaAs buffer layer surface became straight after 30-min thermal treatment at 600°C under AsH_3/H_2 atmosphere. A similar SQW structure was simultaneously grown on a singular (001) GaAs substrate as a reference. AsH_3 partial pressures were 4.2×10^{-4} atm for GaAs buffer layer growth and 6.7×10^{-5} atm for GaAs quantum well layer growth. Growth rates for GaAs buffer layer, AlAs lower barrier layer and GaAs quantum well layer were 1.14, 0.067 and 0.046 nm/s, respectively. Photoluminescence (PL) was measured at 20K using an Ar^+ laser.

3. Results and Discussion

3.1. Multiatomic step formation during MOVPE growth and thermal treatment

During GaAs growth on a vicinal substrate, the surface steps bunch each other to form multiatomic steps with atomically flat terraces, and this behavior rapidly saturates at a growth thickness of less than 100 nm. This tendency was already reported by several authors [6,7]. Figure 1 shows the relation between the terrace width and the growth thickness at the growth temperature of 600°C. At the beginning of the growth, the terrace widths increase linearly as the growth thickness, and are independent of the growth rate. This means that the growth time is not main factor for the step bunching. We also tried to reproduce the step bunching process by using a Monte Carlo simulation. The step bunching phenomenon can be observed only when the activation energy for the migration adatom to the up side step is higher than that to the down side step. From the fitting of the Monte Carlo simulation to the experimental data, we estimated that the activation energy to the up step and down step sites compared with that to the terrace site are 0.47 and 0.07 eV, respectively. Detail about the simulation is reported elsewhere [10]. At the growth temperature of 700°C, the terrace widths have no longer linear development process for the growth thickness, and depend on the growth rate. These results suggest that, at higher growth temperature, the atoms once incorporated in the step sites are easily detached and contribute to the step bunching process as well as the atoms from the vapor phase.

Next, we investigated the effect of thermal treatment under AsH_3/H_2 atmosphere on GaAs vicinal substrates. The substrate misorientation angle is 2.0°. Figure 2 shows AFM images of (a) a chemically etched GaAs surface, and GaAs surface after thermal treatment (b) under low AsH_3 partial pressure (1.3 x 10^{-4} atm) and (c) under high AsH_3 partial pressure (1.3 x 10^{-3} atm) conditions, respectively. After thermal treatment, the clear multiatomic steps appeared. For more detailed investigation about the step bunching mechanism during thermal treatment, AlGaAs/GaAs single quantum well(SQW) structure with thermal treatment process to GaAs quantum well surface were grown by MOVPE. GaAs quantum well layer surface was annealed at 800°C for 5 min under AsH_3/H_2 atmosphere. The photoluminescence spectra of the SQWs were almost unchanged, compared with the reference sample without thermal treatment process which has no bunching steps on GaAs quantum well layer surface [11]. These results suggest that the atoms detached from the step edge migrate on the terrace and are re-adsorbed to the step sites to form the multiatomic steps, and the amount of the evaporation of the adatoms on the terraces is negligible.

3.2. Fabrication and characterization of quantum well wires using coherent multiatomic steps

A schematic illustration of GaAs QWW structure is shown in Fig. 3. Average thicknesses of AlAs lower barrier layer, GaAs quantum well layer (L_w), and $Al_{0.35}Ga_{0.65}As$ upper barrier layer were 17, 5 and 150 nm, respectively.

First, in order to form coherent multiatomic steps with extremely straight edges prior to QWWs formation, we investigated multiatomic steps on GaAs buffer layer after thermal treatment. Figure 4(a) shows an AFM image of GaAs buffer layer surface after 30-min thermal treatment at 600°C under AsH_3/H_2 atmosphere and the distribution of the step spacing. AsH_3 partial pressure was 6.7 x 10^{-5} atm. Coherent multiatomic steps with extremely straight edges were observed over a several micron area. Average height and spacing of multiatomic steps were 5.5nm and 63nm, respectively, and the fluctuation of the step spacing is ±16%. Next, AlAs lower barrier layer surface grown on GaAs buffer layer were observed by AFM. Figure 4(b) shows AFM image of the AlAs lower barrier layer surface on the 5.0°-misoriented substrate grown at 650°C and the distribution of the step spacing within the observed area. In this sample, the 3-nm-thick GaAs cap layers were grown on the lower barrier layer for AFM observations in air. Average height and spacing of the multiatomic steps were 5.7nm and 66nm, respectively, and the uniformity of the step spacing was ±14%. The multiatomic step edges on AlAs surface were straight, and the step spacing was almost the same as that on the underlying GaAs surface. In our previous study, it was found that the step edges tended to undulate on the AlGaAs surface, and that the step spacing of multiatomic steps was smaller than that on the underlying GaAs buffer layer surface.[1] Therefore, this result indicates that the QWW structures can be uniformly fabricated over a wide area by using AlAs as the lower barrier layer rather than AlGaAs.

Next, we measured PL spectrum from QWWs and compared with that from quantum well (QW) on singular (001) GaAs substrate. The PL spectra at 20K are shown in Fig. 5. The average quantum well widths (L_w) were 5.5 nm (1.617eV). The PL peak position of the QWWs on the 5.0°-misoriented substrate was 1.593eV. It was found that the PL peak energy of the QWWs was smaller than that of the QW formed on the singular (001) GaAs substrate. Although the peak energy should shift toward the high energy region due to two-dimensional quantum confinement, these energy shifts indicate that locally thick quantum-wire-like structures are formed at the corners of the multiatomic steps.

In these PL spectra at 20K, the full width at half-maximum (FWHM) of the QWWs was 25meV. The result shows that the size uniformity of the QWWs with AlAs lower barrier layer was much improved to the previous report.[1] From the AFM observations, this size uniformity of the QWWs is probably due to the spacing uniformity of the multiatomic steps on AlAs lower barrier layer surfaces. Moreover, in the PL spectra of the sample with AlAs as lower barrier layer, we observed an additional weak spectrum at about 1.748eV. This peak probably corresponds to the quantum wells formed on the (001) terraces connected with QWWs. The result indicates that the most of the photo-excited electrons and holes diffuse to lower energy regions, that is, the QWW regions.

Next, the polarization dependence of PL intensity (I) from the QWWs using AlAs as lower barrier layer was also measured at 20K, and the results are shown in Fig. 6. Since the misorientation direction is $[\bar{1}10]$ and the direction along the QWWs is [110], we defined the degree of polarization as $(I[110]-I[\bar{1}10])/(I[110]+I[\bar{1}10])$. The degree of polarization for the QWWs was found to be 0.12. For normal QW on the singular (001) GaAs substrate, this value was about 0.04, and no large effect due to internal stress is expected in this material system. Therefore, the observed polarization anisotropy supports the successful formation of QWWs at the corners of the multiatomic steps.

Furthermore, we observed fine-area PL spectra for QWWs using Au/Ge patterned mask. Open area is 2μm wide and 800μm long; thus we can obtain PL spectra from about thirty QWWs. It was found that the FWHM of PL spectra from about thirty QWWs is 17meV, which is 8meV smaller than that from the whole area. This result suggests that the size fluctuation of QWWs still remain for a wide area.

4. Conclusions

In this paper, we investigated the behavior and mechanism of the coherent multiatomic step formation during MOVPE growth and thermal treatment under AsH_3/H_2 atmosphere, and also fabricated self-organized QWWs using these coherent multiatomic steps on GaAs (001) vicinal surfaces. Coherent GaAs multiatomic steps with extremely straight edges were formed over a several micron area. It was also found that the underlying coherent GaAs multiatomic steps were *well traced* by AlAs as lower barrier layer of SQW structure. Furthermore, we fabricated self-organized AlGaAs/GaAs/AlAs QWWs on these coherent multiatomic steps, and measured PL spectra at 20K. The peak energies of PL spectra from the QWWs were smaller than those from the QW formed on singular (001) GaAs substrates, and the polarization anisotropy of the QWWs was also observed. These energy shifts and polarization anisotropy of PL spectra indicate that locally thick QWW structures were successfully formed at the corners of the multiatomic steps. Since the FWHM of PL spectrum from about thirty QWWs was 17meV, which is 8meV smaller than that from a whole area, the size fluctuation of QWWs still remain in a several micron area.

Acknowledgment

The authors are grateful to Prof. H. Hasegawa for fruitful discussions.

References

[1] S. Hara, J. Ishizaki, J. Motohisa, T. Fukui and H. Hasegawa: J. Cryst. Growth **145**(1994)692.

[2] K. Inoue, K. Kimura, K. Maehashi, S. Hasegawa, H. Nakashima, M. Iwane, O. Matsuda and K.Murase: J. Cryst. Growth **127**(1993)1041.

[3] T. Fukui and H. Saito: Appl. Phys. Lett. **50**(1987)824.

[4] P. M. Petroff, A. C. Gossard and W. Wiegmann: Appl. Phys. Lett. **45**(1984)620.

[5] T. Fukui and H. Saito: Jpn. J. Appl. Phys. **29**(1990)L483.

[6] M. Kasu and T. Fukui: Jpn. J. Appl. Phys. **31**(1992)L864.

[7] J. Ishizaki, S. Goto, M. Kishida, T. Fukui and H. Hasegawa: Jpn. J. Appl.Phys. **33**(1994)721.

[8] S. L. Skala, S. T. Chou, K. -Y. Chen, J. R. Tucker and J. W. Lyding: Appl. Phys. Lett. **65**(1994)722.

[9] K. Hata, A. Kawazu, T. Okano, T. Ueda and M. Akiyama: Appl. Phys. Lett. **63**(1993)1625.

[10] J. Ishizaki, K. Ohkuri and T. Fukui: to be submitted.

[11] K. Ohkuri, J. Ishizaki, S. Hara and T. Fukui: J. Cryst. Growth, to be published.

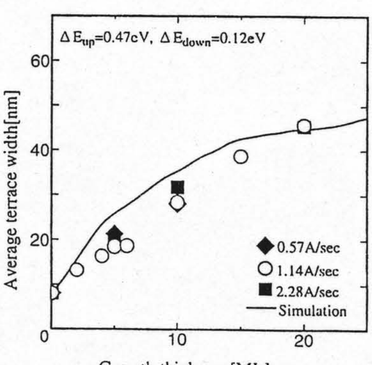

Fig. 1. The relation between the terrace width and the growth thickness at the temperature of 600°C. For fitting the simulation value to the experimental data, the fitting parameters of the activation energy to the up site and down step sites compared with that to the terrace site of 0.47 and 0.07 eV were used.

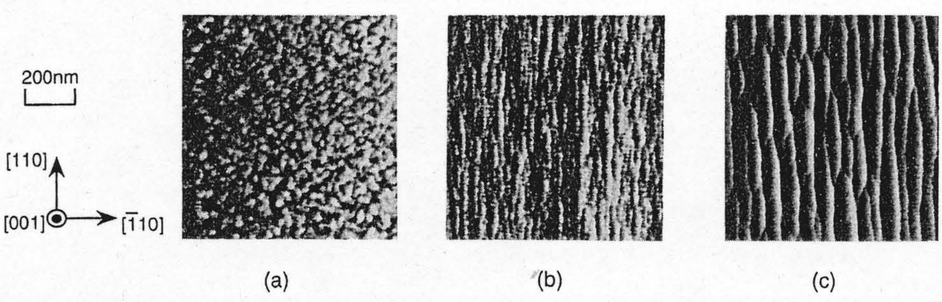

Fig. 2. AFM images of (a) a chemically etched GaAs surface, and GaAs surface after thermal treatment under (b)low and (c) high AsH_3 partial pressure conditions.

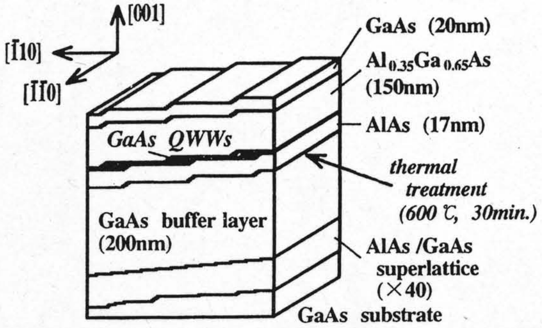

Fig. 3. Schematic illustration of QWW structures on the GaAs vicinal substrate.

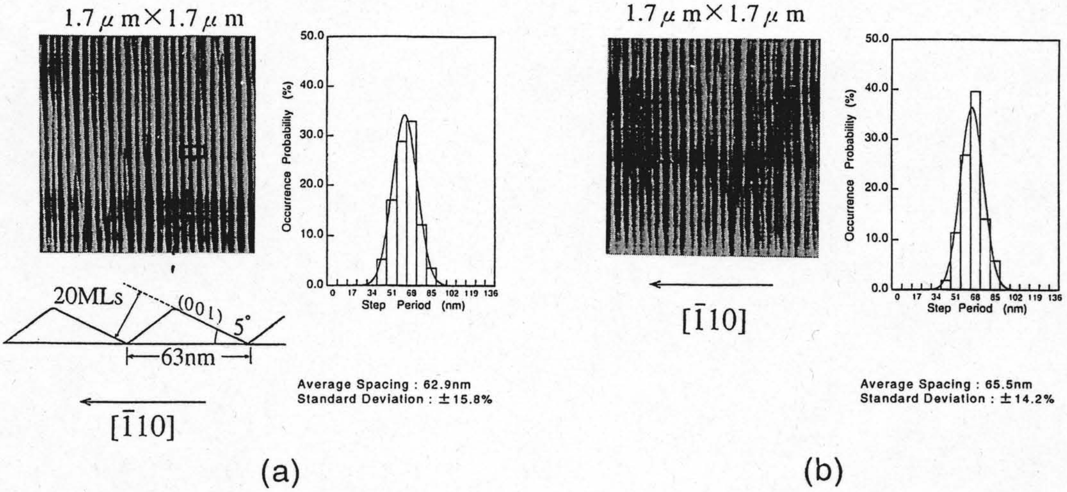

Fig. 4. AFM images and a cross-sectional illustration of GaAs buffer layer surface on vicinal substrate misoriented by 5.0° towards the [Ī10] direction. Observed area is 1700nm x1700nm. (a) GaAs buffer layer surface, (b) AlAs lower barrier layer surface grown on coherent GaAs multiatomic steps at 650°C.

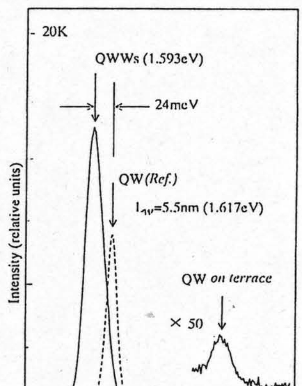

Fig. 5. PL spectra of QWWs on the 5.0°-misoriented substrate, and QW on the singular (001) substrate at 20K. Upper spectra are from quantum structures with AlAs as lower barrier layer, and lower ones are from those with AlGaAs as lower barrier layer. Thicknesses of well layer on the singular (001) substrates, L_W, are also shown.

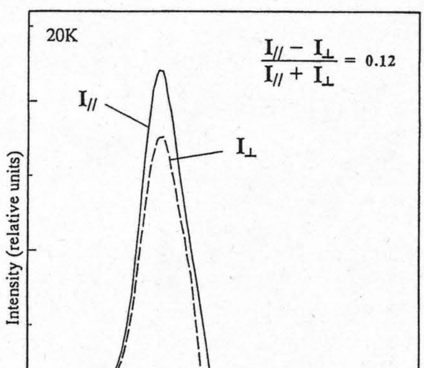

Fig. 6. Polarization dependence of PL spectra from the QWWs at 20K. Solid line is PL intensity parallel to the QWWs ([110] direction) and dashed line is that perpendicular to the QWWs ([Ī10] direction). Degree of polarization is about 0.12.