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Mechanism of Multiatomic Step Formation during MOCVD Growth of GaAs on (001) Vicinal Substrate Studied by Atomic Force Microscopy

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The detailed behavior and mechanism of multistep formation process during metalorganic chemical vapor deposition growth of GaAs on vicinal surfaces are systematically investigated using atomic force microscopy. Depending on the growth rate, the step-flow growth mode and the two-dimensional nucleation growth modes are shown to appear, producing "regular steps" and "irregular steps" respectively. In the former mode, the step height is determined by the terrace width corresponding to the migration length of Ga atoms.

1. Introduction

Recently, there has been much interest in formation of multiatomic steps (multisteps) or monoatomic step through the metalorganic chemical vapor deposition(MOCVD) growth on vicinal surfaces. This is because use of multisteps or monoatomic steps has been recognized as a promising approach for the fabrication of the new quantum electron and optical device such as GaAs quantum wires and lateral superlattices[1] without lithographic process. For such a purpose, accurate control of step height is extremely important.

Although monoatomic steps can be maintained on carefully prepared vicinal substrate under normal MOCVD growth conditions, multisteps are also formed especially under low arsenic pressure growth conditions[2], as has been recently shown by atomic force microscopy (AFM)[3]. However, the details of such a multistep formation process are not understood. The purpose of this paper is to systematically investigate the detailed behavior and mechanism of the multisteps formation process during MOCVD growth using AFM.

2. Experimental

A standard horizontal low pressure MOCVD reactor was used. Triethylgallium(TEGa), triethylaluminum(TEAl) and AsH_3 were used as source materials. The AsH_3 partial pressure was kept to 3.33×10^{-4} atm and the TEAl partial pressure, to 8.95×10^{-7}atm. The TEGa partial pressure was changed from 1.98×10^{-6} to 9.92×10^{-6} to change the growth rate from 1.0 to 5.0 Å/sec.

The substrate was heated to about 650°C to remove the surface oxide. The growth temperature was 600°C where TEGa is completely decomposed to produce Ga atoms[4]. The substrates were (001) GaAs surface misoriented toward [110] or [$\bar{1}$10] direction with angle of misorientation of 1.0°-3.0°. In some experiment, prior to growth of GaAs a $(GaAs)_3(AlAs)_3$ superlattice was intentionally grown on the GaAs substrate as buffer layer in order to realize a starting surface which contains only monoatomic steps.

After growth, the sample was quickly cooled down to room temperature in an AsH_3 atmosphere so as to maintain the step structure. It was then transferred from the MOCVD system to on AFM system where the top surface of the samples was investigated in air.

3. Results and Discussion

3.1 Step Bunching Observed by AFM

The AFM images of the GaAs surfaces grown at 600°C at the growth rate 1Å/sec on 1° and 3°-misoriented substrates towards [110] are shown in Fig.1(a) and (b), respectively. Existence of fairly uniform stripes corresponding to steps are clearly seen in both cases with the average step separations of 170-180nm. This separation is much larger than the calculated monoatomic step separations of 16nm and 5nm for (a) and

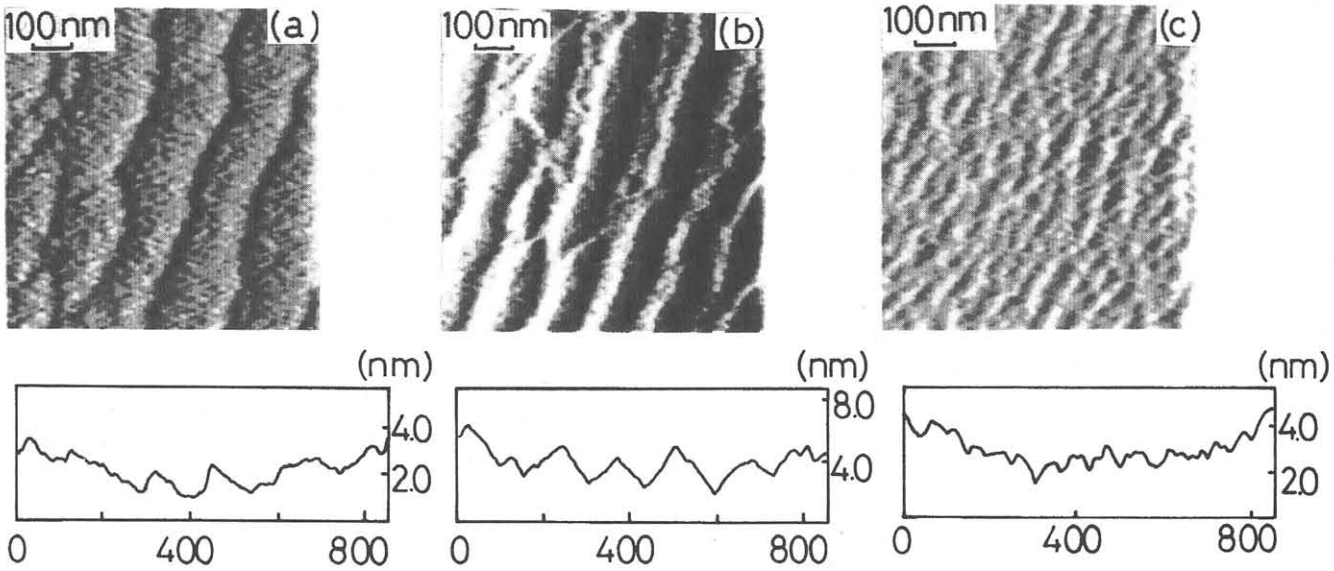

Fig.1:AFM image of GaAs epitaxially grown surfaces. (a) misorientation angle = 1°, 1 Å/sec, (b) misorientation angle = 3°, 1 Å/sec and (c) misorientation angle = 3°, 5 Å/sec.

(b), respectively. Thus, observed steps are multisteps with a height of several tens of atomic layers which were formed obviously by bunching of steps. On the other hand, when the growth rate was 5Å/sec, clear stripe structure was no longer observable as shown in Fig.1(c), and the average separation of these "irregular steps" became much smaller.

An example of measured time evolution of multisteps with regular stripes is shown in Fig.2 in terms of terrace width. It is seen that the terrace width shows saturating behavior and reaches its final value fairly quickly at a growth thickness of 40-60nm. The behavior strongly suggests that this "final terrace width" is a characteristic quantity of a dynamic balance associated with step bunching.

3.2 Behavior of Final Terrace Width

The observed final terrace width is plotted in Fig.3(a) vs. nominal starting terrace width assuming monoatomic step. The final terrace width was determined at the growth thickness of 120nm where complete saturation of the terrace width took place. Fourier spectra of the step profiles determined by AFM shown in the inset. Note that the data for growth rate of 5Å/sec does not represent the terrace width on "regular stripes",but the average separation of the "irregular steps" such as shown in Fig.1(c). The corresponding calculated step heights is shown in Fig.3(b). The terrace width on "regular stripes" is almost independent of the misorientation angle. Comparison Fig.3(a) and (b) suggests that the terrace width is more essential than the height of steps.

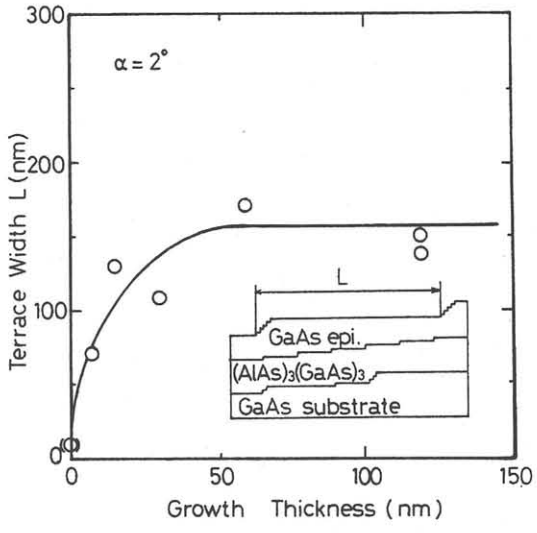

Fig.2:Time evolution of terrace width.

Figure 4 shows the observed dependence of the terrace width on the growth rate. The experiment was done on superlattice buffers which only have monoatomic step. The result strongly suggests that the growth changes abruptly from a mode having well defined terraces on "regular stripes" to a mode with "irregular steps".

3.3 Discussion

The results obtained in this study can be explained in the following way. Depending on the material supply, there are two modes of growth, i.e., the step-flow mode with step

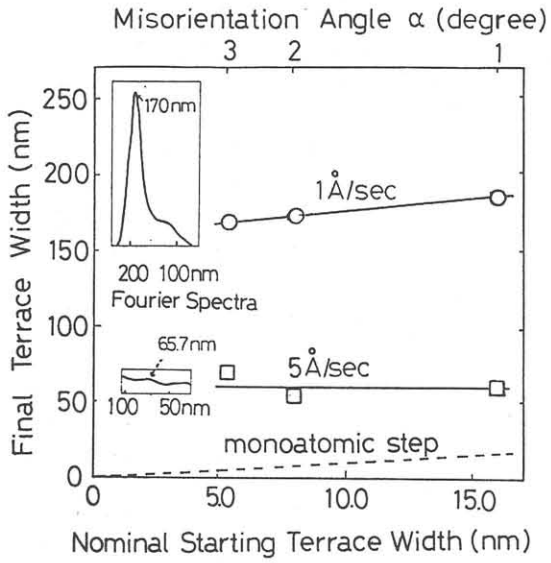

Fig.3(a):Relation between final terrace width and nominal terrace width.

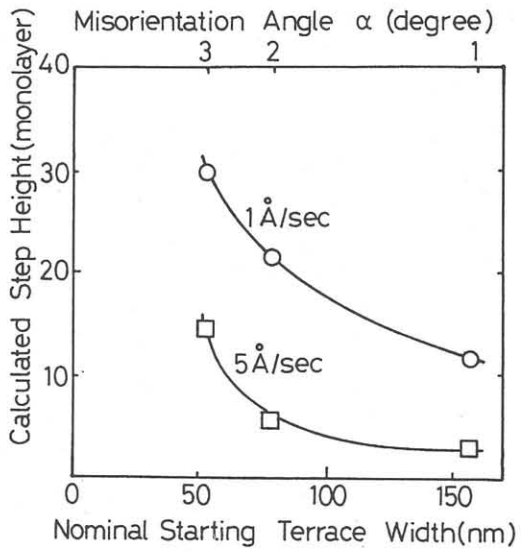

Fig.3(b):Relation between calculated step heights and nominal terrace width.

bunching and two-dimensional nucleation growth mode producing "irregular steps".

In the step flow mode growth, step bunching is caused by relatively free migration of Ga atoms on the terrace combined with their preferential sticking at steps. Thus, the final value of the terrace width is primarily determined by the migration length of Ga atoms, being almost independent of starting terrace width. The observed extremely weak dependence of the final terrace width on the initial terrace width strongly indicates that the difference between the barrier potentials at the terrace site and the step site is small so that the surface migration length is limited primarily by the migration life time, and not by preferential sticking at steps. If, on the other hand, the deference between the potentials is large, the final terrace width should have depend on the misorientation angle or the initial terrace width.

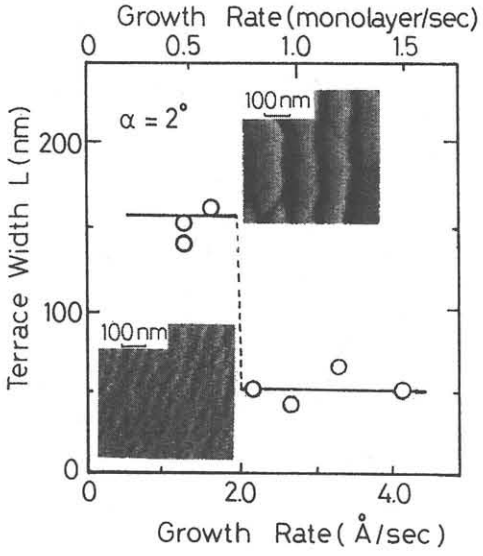

Fig.4:Dependence of the terrace width on the growth rate.

4. Conclusion

The detailed behavior and mechanism of multistep formation process during MOCVD growth on vicinal GaAs substrates were investigated using AFM. The AFM images showed presence of regular stripes and irregular steps depending on the growth rate. The terrace width of the regular stripes showed saturation to values within 170-180nm being independent of the misorientation angle. The results were explained in terms of step-flow mode and the two-dimensional nucleation growth mode where the final terrace width in the former mode is determined by migration length of Ga.

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