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STM Study of Monolayer Steps on GaAs Vicinal Surfaces Grown by MOCVD

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We use scanning tunneling microscopy (STM) to study metalorganic chemical vapor deposition (MOCVD)-grown GaAs step structure on a vicinal substrate. The surface is passivated with As to protect it from air during the transfer from the MOCVD to the STM. At the growth temperature (Tg) of 700 °C the monolayer step edges on the surface misoriented in the [110] direction undulate with 2 times larger amplitude than the surface misoriented in the $[\bar{1}10]$ direction. At Tg=600 °C step bunching occurs and the step height becomes 1 to 5 monolayers. The phase diagram of the step structure as functions of growth temperature and misorientation angle is determined by STM and AFM.

1. INTRODUCTION

The step structures such as monolayer step staircase and multiatomic steps (multisteps) are fundamental phenomena of the crystal growth. One of their applications is the low-dimensional microstructure like the fractional layer superlattice (FLS) grown by molecular beam epitaxy (MBE) and metalorganic chemical vapor deposition (MOCVD).[1,2] Although reflection high-energy electron diffraction (RHEED) can be used to investigate the step structure during MBE growth,[3,4] *in-situ* surface observation techniques like RHEED are difficult to use in the MOCVD growth reactor, because the MOCVD growth is usually performed at H_2 pressures from 10^5 to 10^3 Pa. The few reports on MOCVD-grown step structures utilized X-ray superlattice diffraction[2] and transmission electron microscopy (TEM) plain-view image.[5] However, these techniques cannot reveal details of the surface structure.

Atomic force microcopy (AFM) images of MOCVD-grown GaAs surfaces on a vicinal substrate showed that the multisteps on a substrate misoriented in the $[\bar{1}10]$ direction are straighter than those on a substrate misoriented in the [110] direction.[6] The multistep edges approach $(117)_B$ facets proceeded by step bunching.[7]

In the work reported here, we use scanning tunneling microscopy (STM), which can generate images with resolution on the scale of less than monolayer heights, as well as AFM. We systematically investigate the MOCVD-grown GaAs step structure over wide ranges of the substrate misorientation angle and growth temperature. We observe monolayer steps on the surface at the growth temperature of 700 °C and compare undulation amplitudes of the monolayer steps on the surface misoriented in the [110] direction and the $[\bar{1}10]$ direction quantitatively. We also observe the multisteps on the surface grown at 600 °C. We determine the phase diagram of the step structure as functions of the growth temperature and the misorientation angle by AFM and STM. This kind study had been impossible because there was no way to transfer the sample from the MOCVD system to the STM without exposure to air. We passivated the grown-surface with amorphous As by using an As effusion cell in a vacuum analysis chamber which was connected to MOCVD system.

2. EXPERIMENTAL PROCEDURE

We grew Si-doped GaAs layer ($n \cong 5 \times 10^{17}$ cm^{-3}) on Si-doped GaAs (001) n^+ vicinal substrates for STM samples. The GaAs epitaxial layer was 140 monolayer (ML)- (39.5 nm-) thick. The source materials were triethylgallium (TEGa) and arsine (AsH_3). The working pressure was 1.0×10^4 Pa (76Torr). The AsH_3 partial pressure was 20 Pa. The growth rate was 1/6 ML/s (The V/III ratio=200). For AFM samples we grew undoped 140 ML-thick GaAs layer.

The MOCVD consists mainly of a quartz reactor, an introduction chamber, and a vacuum analysis chamber as shown in Fig. 1. The vacuum analysis chamber was equipped with a RHEED system, and an As effusion cell with a shutter.

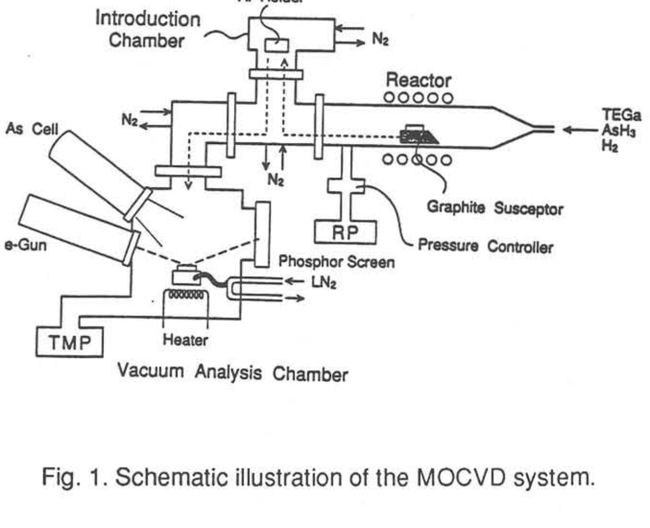


Fig. 1. Schematic illustration of the MOCVD system.

The vacuum analysis chamber was evacuated to less than $1\mathrm{x}10^{-7}$ Pa by using a turbo molecular pump, and the sample in the vacuum analysis chamber could be heated and cooled between -30 and 400 °C.

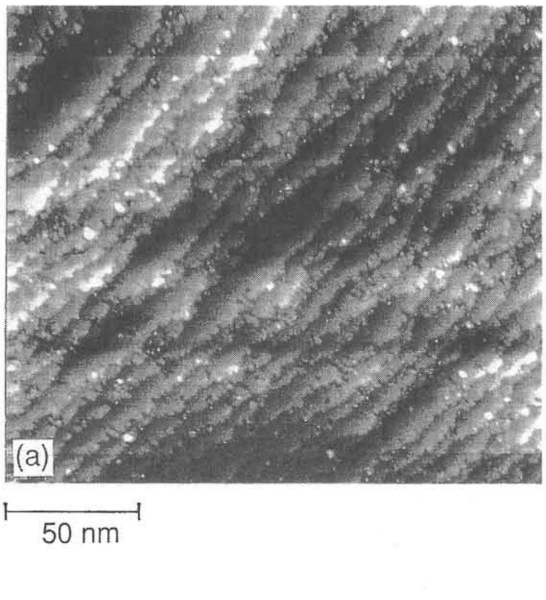
After growth, the sample was then quenched at the rate of -1.7°C/s to room temperature in AsH_3 atmosphere. Then the sample was transferred under N_2 from the reactor through the introduction chamber, to the vacuum analysis chamber. The N_2 gas (purified to a dew point range below -100 °C) enabled us to keep the grown surface contamination-free while transferring the sample from the reactor to the vacuum analysis chamber. In the introduction chamber, the sample was moved from the graphite susceptor to an aluminum holder, because degassing of the graphite susceptor might contaminate seriously the sample surface in the vacuum. Then the pressure in analysis chamber was lowered from atmospheric pressure to $1\mathrm{x}10^{-7}$ Pa for about 1 h. Under this vacuum condition, the RHEED pattern of a vicinal surface grown at 700 °C showed that the diffraction spots split, showing that a monolayer step staircase exits on the vicinal surface, and that the surface has c(4x4) reconstruction.[8] To prevent the sample surface from oxidizing during the transfer from the MOCVD system to the STM system, we deposited amorphous As on it for 2h at -30 °C at an As pressure of $5\mathrm{x}10^{-3}$ Pa.

The STM was installed at a vacuum chamber at a pressure of about $3\mathrm{x}10^{-8}$ Pa. For STM imaging at high temperatures (up to 1200 °C), we heated the sample. We imaged the surface while gradually raising the sample temperature. At around 235 °C, the amorphous As cap layer evaporated, the diffraction spots split and c(4x4) reconstruction appeared in the RHEED pattern of the surface grown at 700 °C.[8]

3. STM RESULTS AND PHASE DIAGRAM

The STM images of GaAs surface grown at 700 °C on a vicinal substrate misoriented in the [110] and [$\bar{1}$10] directions are shown in Figs. 2(a) and 2(b). The misorientation angle here was 2.0°. At 235 °C, the surface atomic structure was stable; no more As atoms evaporated and the steps did not change during the several-hour observation time. The sample bias was -5.0 V, and the STM image represents the filled state, that is, the electron localized on the As atoms. All the step heights were measured to be 0.28 nm, corresponding to half of the lattice constant, so we can conclude that they were monolayer steps. The average terrace width was 8 nm. This value agrees with the calculated monolayer step separation of vicinal surface misoriented by 2.0°. The STM images indicate that the A-steps, that is, those on the vicinal surface misoriented in the [110] direction, have an undulation appearance. The B-steps, on the other hand, that is, those on the vicinal surface misoriented in the [$\bar{1}$10] direction appear to be relatively straight.

We quantitatively analyzed the step undulation apparent in the STM images of Fig. 2(a) and 2(b). The step undulation amplitude is the distance between peaks in the undulations seen in plan views of the steps. This amplitude is 11±1 nm for the A-steps and 6±1 nm for the B-steps. The step undulation amplitude for the A-steps is twice larger than those for the



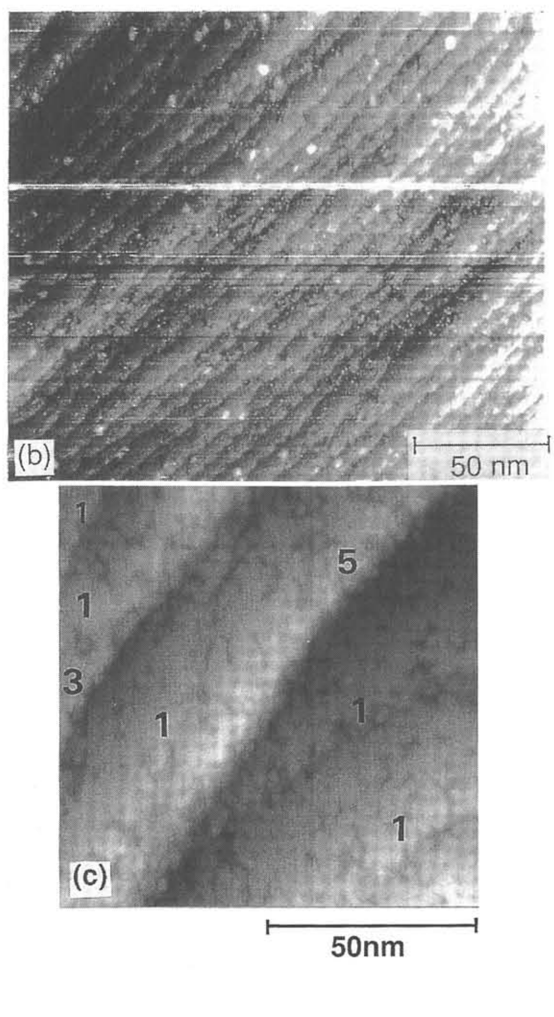


Fig. 2. STM images of GaAs (001) vicinal surfaces. The misorientation angles are 2.0°: (a) misorientation direction *d*=[110] and growth temperature Tg=700 °C; (b) *d*=[$\bar{1}$10] and Tg=700 °C; (c) *d*=[$\bar{1}$10] and Tg=600 °C. The number inside the figure is the step height. The unit is MLs.

B-steps, and this is the contrary to that of MBE-grown (2x4) reconstruction surface.[9] The step straightening has been explained to be due to preferential sticking at the kink rather than at other sites of the step.[10] The smaller undulation amplitude for the B-steps indicates that the migrating species stick more easily at the kinks in the B-step than at kinks in the A-step.

The STM image of a GaAs surface grown at 600 °C is shown in Fig. 2 (c). The substrate was misoriented by 2.0° in the [$\bar{1}$10] direction. The step height was measured in constant current mode. The step height became 1 to 5 MLs. Comparison of Fig. 2(b) and 2(c) shows that the monolayer steps bunch as the growth temperature decreases from 700 °C to 600 °C.

The phase diagram of the step structure for the [$\bar{1}$10] misorientation direction was determined by STM and AFM as shown in Fig. 3. The misrorientation angle of 11.4° corresponds to $(117)_B$ surface. We considered the growth conditions for which multisteps with more than 3-ML height were observed as step bunching conditions. We can see from this phase diagram that the step bunching occurred at growth temperature ranging from about 560 °C to about 680 °C and at misorientation angles ranging from less than 9°. The step bunching region

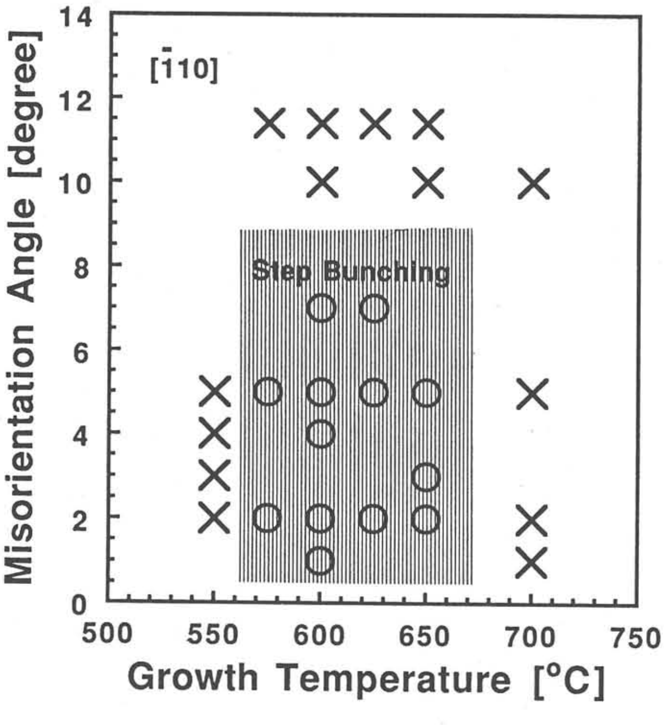


Fig. 3. Phase diagram of the step structure determined by STM and AFM. The growth conditions for which multisteps with more than 3-ML height were observed are considered as step bunching conditions. The circles represent step bunching observed and the crosses represent the absence of step bunching.

includes the conditions used to make the STM image shown in Fig. 2 (c). The STM image shown in Fig. 2 (b), however, which was made under conditions outside the step bunching region, shows that above 680 °C the surface is a monolayer step staircase. On the other hand, AFM image suggests that the surface outside the step bunching condition below 560 °C has two-dimensional islands.

The transition of the multistep to the monolayer step cannot be explained in terms of the surface diffusion. This is because the surface diffusion length should increase as the growth temperature. Rather, the transition should be considered to relate to something of the surface stability, e.g., the surface atomic structure. We reported previously that the multistep edge approaches the $(117)_B$. Then we consider that the $(117)_B$ atomic structure which mainly forms the multistep edge becomes no longer stable above 680 °C and the transition occurs around the temperature.

4. CONCLUSIONS

We studied using STM MOCVD-grown GaAs step structure on a vicinal substrate. The surface was passivated with As to protect it from air during the transfer from the MOCVD system to the STM system. The STM images revealed that at the growth temperature of 700 °C the monolayer step edges on the surface misoriented in the [110] direction undulate with about 2 times larger amplitude than the surface misoriented in the [$\bar{1}$10] direction. On the surface at the growth temperature of 600 °C, step bunching occurs and the step height becomes 1 to 5 MLs. The reason of the transition from the monolayer step to the multistep is considered to originate from the stability of $(117)_B$ facet which mainly forms the multistep edge below around 680 °C. The phase diagram of the step structure as functions of the growth temperature and the misorientation angle was determined by STM and AFM.

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