ELSEVIER

Applied Surface Science 87/88 (1995) 364–367

applied surface science

MBE-STM study of the Ga-rich 4 × 2 phase of the GaAs(001) surface

Qikun Xue *, T. Hashizume, J.M. Zhou [1], T. Sakata [2], T. Sakurai

Institute for Materials Research, Tohoku University, Sendai 980-77, Japan

Received 11 July 1994; accepted for publication 12 September 1994

Abstract

We studied the atomic structure of the GaAs(001) 4 × 2 and 4 × 6 phases by scanning tunneling microscopy (STM) and determined that both the 4 × 2/c(8 × 2) and 4 × 6 phases are Ga-terminated. The 4 × 2/c(8 × 2) structure can be described better by Biegelsen et al.'s bilayer Ga dimer model than by Skala's As model.

1. Introduction

While the As-terminated 2 × 4/c(2 × 8) reconstruction has been most extensively investigated so far, few works have been reported on the Ga-rich surface reconstructions, such as 2 × 6, 4 × 2/c(8 × 2) and 4 × 6 [1–15]. There are no studies being published dealing with the as-deposited Ga-terminated GaAs(001) surface, due to difficulty in preparing these Ga-rich surfaces under conventional MBE (molecular beam epitaxy) growth conditions. The past reports all dealt with the surface obtained by a post-growth UHV heating process.

There are four models for the 4 × 2/c(8 × 2) phase at present: (i) three Ga dimer model by Frankel et al. [10], (ii) two Ga dimer bilayer model (called Ga model hereafter) by Biegelsen et al. [12] and Northrup and Froyen [13], (iii) mixed Ga–As model by Falta et al. [14] and (iv) Skala et al. their As model [15]. In the case of the 4 × 6 phase, no direct observation of the surface phase by STM was reported so far [1–11].

2. Experimental

We have carried out the first in situ STM (scanning tunneling microscopy) investigation of the Ga-rich 4 × 2 and 4 × 6 phases, making a good use of the migration enhanced epitaxy (MEE) technique [16], which is ideal and indispensable for the present research. In the MEE growth, Group III and V atoms are alternatively supplied to the substrate for enhanced migration on the surface (Fig. 1) [17], thus, high quality surfaces with large domains can be realized at relatively low temperatures (~ 500°C) [18]. It is also possible to obtain various surface As/Ga ratios with this technique.

The experiments were carried out in the MBE equipment combined with an UHV field ion scan-

* Corresponding author. Fax: +81 22 2152020.

[1] Permanent address: Institute of Physics, Academia Sinica, Beijing, People's Republic of China.

[2] Permanent address: Osaka Prefecture University, Osaka, Japan.

SSDI 0169-4332(94)00509-5

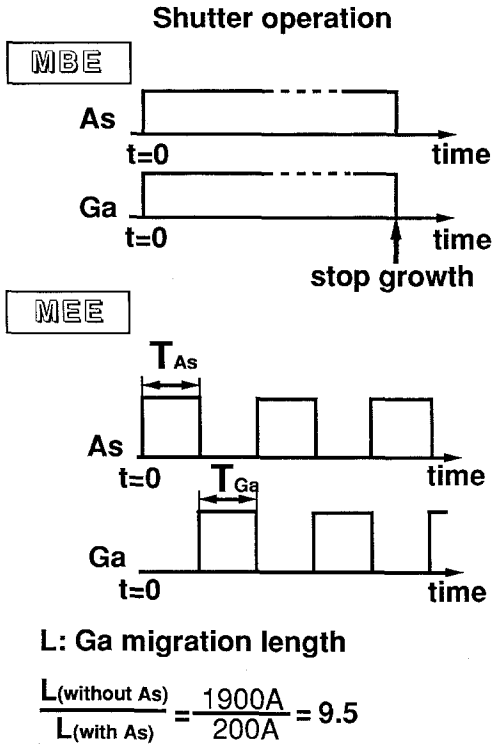

Fig. 1. Schematics showing the differences between conventional MBE and new MEE techniques.

ning tunneling microscope (FI-STM) system which has been described elsewhere [19,20]. The high quenching rate of the present apparatus allows us to study various surface phases obtained during growth by in situ STM observation. It has been successfully used in the surface morphology improvement by MEE and other studies [21–23].

Si-doped well-oriented GaAs(001) substrates (4×10 mm^2) were cleaned by standard cleaning and etching procedures. An approximately 0.3 μm thick buffer layer was first grown by the conventional MBE technique at 600°C under optimal growth conditions. At the end of the growth, all shutters and substrate heating were stopped and the As and Ga cells were set to the appropriate temperature in order to produce desirable fluxes for the low temperature MEE growth with various As/Ga flux ratios. The sample was annealed at 500°C under the As flux until the well-ordered 2×4 RHEED pattern appeared, then immediately started the MEE growth controlled by the computer. Typically 12 cycles of the MEE were deposited onto the buffer layer, then the sample was quenched to room temperature in less than 2 s right after the last Ga cycle was completed in order to minimize the residual As deposition on the surface. All STM images were acquired at room temperature with a constant tunneling current of 40 pA and negative bias (V_b) to the sample. After STM imaging, all samples were tested by RHEED to confirm the surface reconstructions.

3. Results and discussion

Fig. 2 shows a typical filled-states image of the $4 \times 2/c(8 \times 2)$ surface prepared by MEE using the 1:8 Ga/As_4 beam equivalent pressure (BEP) ratio and with 2 s As and 2 s Ga pulsed exposure. Randomly distributed two-dimensional large islands reveal the layer-by-layer growth made possible by MEE. Even with a relatively low growth temperature, there are only three terraces over an area of 900 Å $\times$ 900 Å (each level corresponds to a bilayer step height (2.8 Å) of the GaAs(001) surface). The degree of the ordering is almost perfect. Most apparent are the bright lines running along the [110] direction with a regular spacing of 16 Å in the [$\bar{1}$10] direction, corresponding to the four-fold periodicity of the surface as observed by other groups [12,15].

Fig. 2b is a zoom-in image of a small section of Fig. 2a, showing the details of the structures. *Each bright line consists of two rows with each being made of regular bumps with a 4 Å separation.* This image is basically the same as that obtained by Skala et al. (Fig. 1 of Ref. [15]). The two-fold periodicity along the [110] direction originates from the well spaced dimers, and the four-fold periodicity in the other direction as discussed above is clearly observed. Some individual dimers and the 4×2 unit cell are highlighted in the lower left corner of the image. (From this image, we can also observe the out-of-phase arrangement of the Ga dimers in adjacent 4×2 subunit cells giving rise to the $c(8 \times 2)$ structure.)

While the obtained STM images are similar, we propose a completely different model from Skala et al.'s. We suggest that the bright protrusions are the *second layer As atoms*, not the first layer As, and

that the faintly imaged dimers are the *first layer Ga dimers*. The most plausible atomic model for this phase is the Ga model proposed by Biegelsen et al. (Fig. 3a). For comparison, the As model by Skala et al. is shown in Fig. 3b. The apparent discrepancy between the images and the Ga model is that the individual protrusions forming two (bright and faint) rows in the [$\bar{1}$10] direction are shifted from each other, while Ga and As atoms line up in the model. We believe that each As atom forming bright rows in the second layer has two electrons in its dangling orbital and contributes significant amounts of charge at the surface, whose overlapping results in a hump at the middle of the neighboring As atoms.

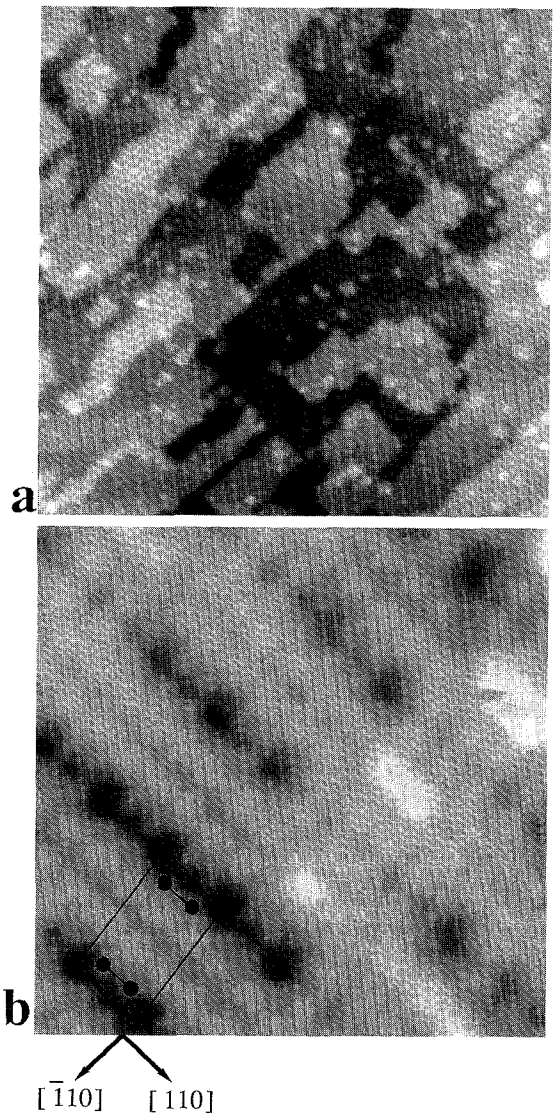

Fig. 2. (a) Large area (900 Å×900 Å) STM image of the 4×2/c(8×2) surface and (b) zoom-in image showing the details. The deposition rate is 0.25 μm/h, the growth temperature is 500°C. During the MEE cycle, a Ga/As flux ratio of 1/8 was used. The image was obtained with a sample bias voltage (V_s) of −1.8 V.

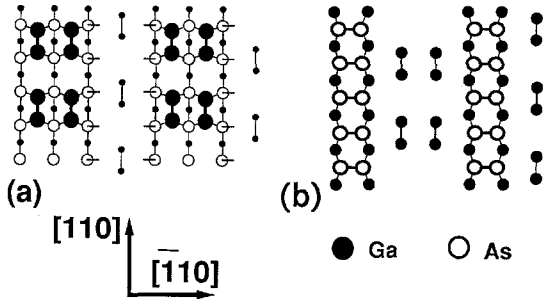

Fig. 3. (a) The bilayer model and (b) the As-dimer model for the 4×2/c(8×2) phase.

The justification for the Ga model is as follows:

(1) The surface preparation process using MEE favors a Ga-rich surface, since the final exposure is by the Ga beam.

(2) The observed height difference between the bright row and the faint row is only 0.6 Å. If the brighter protrusions correspond to As in the first layer, a much higher image contrast (at least 1.4 Å) is expected between the As atoms (of the first layer) and the Ga atoms (of the second layer), since the Ga atom's dangling orbital is empty. The STM-related characteristics of Ga and As atoms have been well documented by Feenstra et al. [24]. If indeed, the surface is covered by As dimers, it is expected that only the first layer As be imaged bright, similarly to the 2×4/c(2×8) surface.

(3) According to the As model, the first layer As atoms form dimers, and thus, their bond length is approximately less than 3 Å [23]. The STM images (also including Skala et al.) show at least 5 Å between two protrusions along the [$\bar{1}$10] direction. This large discrepancy cannot be explained in the present STM study even though the STM is mapping the charge contour and not the atom positions. Furthermore, a nodal structure is expected for the (As)

dimer in the filled states image. Instead, we observe two isolated humps clearly shown in Fig. 2b [25]. (In Ref. [26] which was quoted by Skala et al. to support the As model, the authors clearly indicated a value of about 2.9 Å for this.)

In conclusion, the experiments reported here strongly support Biegelsen et al. their Ga model for the (4×2) phase.

References

[1] J.H. Neave and B.A. Joyce, J. Cryst. Growth 44 (1978) 387.
[2] A.Y. Cho, J. Appl. Phys. 42 (1971) 2074.
[3] P.K. Larsen and D.J. Chadi, Phys. Rev. B 37 (1988) 8282.
[4] A.J. Van Bommel, J.E. Crombeen and T.G. Van Oirschot, Surf. Sci. 72 (1978) 95.
[5] W. Ranke and K. Jacobi, Prog. Surf. Sci. 10 (1981) 1.
[6] P. Drathen, W. Ranke and K. Jacobi, Surf. Sci. 77 (1978) L162.
[7] J. Massies, P. Etienne, F. Dezaly and N.T. Linh, Surf. Sci. 99 (1980) 121.
[8] J.R. Creighton, Surf. Sci. 234 (1990) 287.
[9] C. Deparis and J. Massies, J. Cryst. Growth 108 (1991) 157.
[10] D.J. Frankel, C. Yu, J.P. Harbison and Frarrell, J. Vac. Sci. Technol. B 5 (1987) 1113.
[11] I. Kamiya, D.E. Aspnes, L.T. Florez and J.P. Harbison, Phys. Rev. B 46 (1992) 15894.
[12] D.K. Biegelsen, R.D. Bringans, J.E. Northrup and L.E. Swartz, Phys. Rev. B 41 (1990) 5701.
[13] J.E. Northrup and S. Froyen, Phys. Rev. Lett. 71 (1993) 2276.
[14] Falta, R.M. Tromp, M. Copel, G.D. Pettit and P.D. Kirchner, Phys. Rev. Lett. 69 (1992) 3068.
[15] S.L. Skala, J.S. Hubacek, J.R. Tucker, J.W. Lyding, S.T. Chou and K.Y. Cheng, Phys. Rev. B 48 (1993) 9138.
[16] Y. Horikoshi, M. Kawashima and H. Yamaguchi, J. Jpn. Appl. Phys. 25 (1986) L868.
[17] S. Nagata and T. Tanaka, J. Appl. Phys. 48 (1977) 940; J.H. Neave, P.J. Dobson and B.A. Joyce, Appl. Phys. Lett. 47 (1985) 100.
[18] Q.K. Xue, T. Hashizume, J.M. Zhou, H. Chaya and T. Sakurai, unpublished.
[19] T. Sakurai, T. Hashizume, I. Kamiya, Y. Hashigawa, N. Sano, H.W. Pickering and A. Sakai, Prog. Surf. Sci. 33 (1990) 3.
[20] T. Hashizume, I. Sumita, Y. Murata, S. Hyodo and T. Sakurai, J. Vac. Sci. Technol. A 9 (1991) 742.
[21] J.M. Zhou, Q.K. Xue, H. Chaya, T. Hashizume and T. Sakurai, Appl. Phys. Lett. 64 (1994) 583.
[22] Q.K. Xue, J.M. Zhou, H. Chaya, T. Hashizume and T. Sakurai, J. Appl. Phys. 75 (1994) 5021.
[23] T. Hashizume, Q.K. Xue, J.M. Zhou, K. Ichimeya and T. Sakurai, Phys. Rev. Lett. 73 (1994) 2208.
[24] R.M. Feenstra, J.A. Stroscio, J. Tersoff and A.P. Fein, Phys. Rev. Lett. 58 (1987) 1192.
[25] M.O. Schweitzer, F.M. Leibsle, T.S. Jones, C.F. McConville and N.V. Richardson, Surf. Sci. 280 (1993) 63.
[26] V. Bressler Hill et al., J. Vac. Sci. Technol. B 10 (1992) 1881.