Origin of step formation on the GaAs(311) surface

Z. M. Wang, V. R. Yazdanpanah, C. L. Workman, W. Q. Ma, J. L. Shultz, and G. J. Salamo
Physics Department, University of Arkansas, Fayetteville, Arkansas 72701

(Received 27 August 2002; published 27 November 2002)

GaAs(311) surfaces grown by molecular beam epitaxy are investigated by *in situ* ultrahigh-vacuum scanning tunnelling microscopy. The observation of an atomically flat Ga(2×1)-reconstructed GaAs(311) surface and its transformation to a 8×1-reconstructed GaAs(311) surface leads to an improved understanding of the processes involved in the step formation. The high density of steps observed on the 8×1-reconstructed GaAs(311) surface along the $[\bar{2}33]$ direction originates from the change of surface atomic density required to accommodate the surface transition from the Ga(2×1) surface to the 8×1 surface. This understanding is further supported by the observation of independent step formation.

DOI: 10.1103/PhysRevB.66.193313 PACS number(s): 81.15.Hi, 61.14.Hg, 81.07.−b, 68.37.Ef

Self-assembled nanostructures, made by depositing a few monolayers of one semiconductor material onto another with a significant lattice mismatch, have been the focus of much research. The potential of these nanostructures[1,2] is particularly noted by recent achievements with quantum dot lasers and detectors, although their performance have been rather limited by their lack of uniformity of both size and shape. In particular, the current lack of uniformity typically associated with self-assembled growth has resulted in a rather large broadening in the density of states, presenting a formable obstacle for the application of nanostructures. The difficulty in obtaining uniformity has, in turn, sparked recent interest in the growth on high index surfaces, with an interest in patterning growth. In addition, not only are investigations of such surfaces meaningful for the application of nanostructures, they simultaneously provide a significant opportunity to gain a greater understanding of the GaAs surface[3–7] in order to uncover its unique electrical and optical properties for advanced optoelectronic devices.[8–14]

An example of the importance of surfaces with high Miller indices, such as the (311) GaAs surface, to the formation of nanostructures is their significant role in determining their shape[15–18] and size uniformity.[19,20] Along these lines, the (311) surface (reportedly the first stable high index GaAs surface[3–5]) was recently used as a template to form highly uniform quantum dot arrays.[21,22] These exciting observations led to the possibility of using other high index surfaces, and consequently to the more fundamental question of when and why high index surfaces are stable. In order to understand the origin of high index surfaces, it is necessary to study them directly. Such an understanding may led to the ability to control and select the morphology of surfaces.

In this investigation a surface phase, with atomically flat Ga(2×1) reconstructed wide terraces, is observed under Ga-rich conditions, and its transformation into many narrow 8×1-reconstructed terraces separated by randomly nucleated steps along the $[\bar{2}33]$ direction is studied. These steps, interesting in their own right, can have a strong impact on the physical properties of GaAs(311)-based structures, such as the optical polarization and electric transport anisotropy. For example, by assembling these steps, a one-dimensional quantum structure was observed and further combined with pattern growth to provide uniform quantum dots.[23,24] Our investigation demonstrates that these steps are formed to accommodate the change of the surface atom density required by the Ga(2×1) to 8×1 phase transition.

Experiments were carried out in a combined Riber molecular beam epitaxy/Omicron scanning tunneling microscope (STM) ultrahigh-vacuum system. For the experiment described here, epiready *n*-type GaAs substrates with both sides polished were used for comparative studies of the (311)A and (311)B surfaces.[25] In this paper, however, we focus on the (311)B surface, which is located about halfway in between the As-terminated (111)B and Ga-terminated (100) surface orientations, although a similar step behavior was observed on the (311)A surface. A 1-μm-thick GaAs buffer layer is grown by molecular beam epitaxy at 580 °C with a growth rate of 1.0 ML_{100}/S (1 ML_{100} = 2.83 Å) and an As_4/Ga beam equivalent ratio of 20. Reflection high-energy electron diffraction (RHEED) was used *in situ* to monitor the growth process. A (1×1)-like RHEED pattern was observed during growth, which was denoted as Ga(1×1) before.[5] This pattern was kept stable by closing the Ga and As shutters at the same time, and quickly removing the sample to the transfer module and cooling it down there out of the residual As_4 background influence. The sample was then transferred to the STM chamber without breaking ultrahighvacuum conditions. The base pressure of the transfer module and the STM chamber are around 2×10^{-10}Torr. Filled-state STM images were collected at sample voltages of 2–3 V and tunnelling currents of about 0.1 nA.

The resulting surface topography is shown in Fig. 1(a). More than 100-nm-wide atomic-flat terraces are separated by 1-ML_{311}-height (1 ML_{311} = 1.7 Å) steps. This surface morphology is comparable in quality with the GaAs(100) surface, and consistent with the observed excellent RHEED oscillations on GaAs(311)B. The high-resolution STM image of Fig. 1(b) reveals a reconstruction of the GaAs(311)B surface, 2×1. The basic unit cell as outlined in Fig. 1(b) is 8×13.3 Å^2. The period along $[01\bar{1}]$ is 8 Å, double the face-centered ideal substrate unit, although RHEED shows a diffusive 1× pattern. 2×1 is the smallest unit cell for GaAs(311) located between (100) and (111), concerning the atomic dimerization of GaAs(100) surfaces and the energy

 ©2002 The American Physical Society

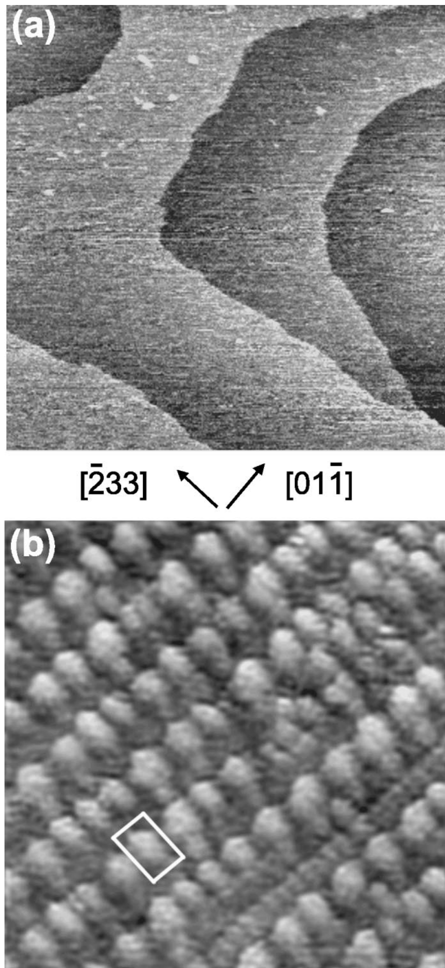

FIG. 1. (a) 600×600 nm^2 STM image showing the wide flat Ga(2×1)-reconstructed terraces. (b) Atomic resolution STM image of a Ga(2×1) reconstruction. The reconstruction Ga(2×1) unit cell is indicated.

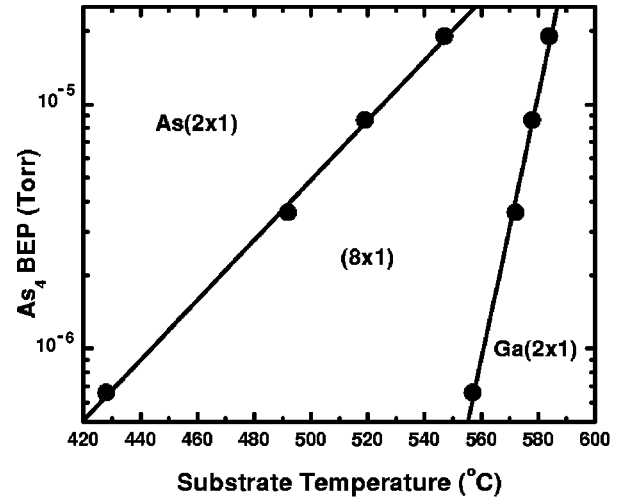

FIG. 2. GaAs(311)B surface phase diagram. The surface reconstructions are plotted vs both the substrate temperature and the As_4 beam equivalent pressure.

minimizing of 2×2-reconstructed GaAs(111) surfaces. The tilted bright bump within the 2×1 unit cell shows the asymmetry of GaAs(311) surface along [$\bar{2}$33]. The reconstruction is denoted as Ga(2×1) here to distinguish it from another GaAs(311)B (2×1) phase observed at high As_4 pressures and low substrate temperatures,[26,27] denoted as As(2×1) here.

Figure 2 displays the GaAs(311)B static surface phase diagram determined from RHEED observations as the function of the substrate temperature and the As_4 beam equivalent pressure. As mentioned above, the 1×1-like RHEED pattern observed during growth was kept stable after quenching the sample. This indicates that the Ga(2×1) reconstruction observed on the quenched surface is maintained during the growth and can be stabilized after growth. As shown in Fig. 1(b), the Ga(2×1)-reconstructed surface is laterally disordered on an atomic scale, which leads to the observed diffusive 1×1-like RHEED pattern. However, an 8×1-reconstructed surface was created after growth by closing the Ga shutter and terminating growth. After a 5-min annealing period at the growth temperature of 580 °C under As_4 pressure, the 8×1 RHEED pattern becomes well ordered.[5] The resulting 8×1 surface morphology was stabilized by turning off the power to the substrate heater and gradually reducing the As_4 background by fine controlling the As valve. The sample was then transferred under ultrahigh vacuum to the STM chamber. A typical STM image is shown in Fig. 3(a). The observed GaAs(311) surface is characterized by narrow 8×1-reconstructed terraces with a high density of straight, 1-ML_{311}-height steps along the [$\bar{2}$33] direction. The bright and dark rows running along the [$\bar{2}$33] direction with a 3.2-nm period result from the corrugation of the 8×1 reconstruction, which is well resolved in Fig. 2(b). Its unit cell of 32×13.3 Å^2 is outlined. The corrugation height is 3.4 Å, 2 ML_{311}, resolved with three clear gray levels. This corrugated surface with ridges along [$\bar{2}$33] is believed to reduce the surface atom mobility along the [01$\bar{1}$] direction.

The physics for the origin of the steps along [$\bar{2}$33] direction on the 8×1-reconstructed GaAs(311) surface is not currently understood. However, the transformation of the Ga(2×1)-reconstructed surface to an (8×1) reconstructed surface makes it possible to elucidate this question. The terrace widths separated by these steps are measured over 12 images similar to Fig. 3(a), but covering different areas. In order to quantify the observation, the resulting images are scanned row by row approximately every 80 nm in the [01$\bar{1}$] direction to avoid correlation effects. The terrace widths are calibrated using the $8\times$ reconstruction, which is 3.2 nm wide. The terrace width distribution for up and down steps was evaluated separately and show only insignificant differences. As a result only the overall terrace width statistical distribution over 639 terraces is shown in Fig. 4. On average, there is one step along [$\bar{2}$33] per 13.2 nm. By comparing Figs. 1(a) and 3(a), it is clear that most of the steps along [$\bar{2}$33] on the 8×1-reconstructed surface are formed during the phase transition from Ga(2×1) to 8×1.

The idea of a phase transition driving step formation was implied by Van Vechten some 25 years ago.[28] The basis of

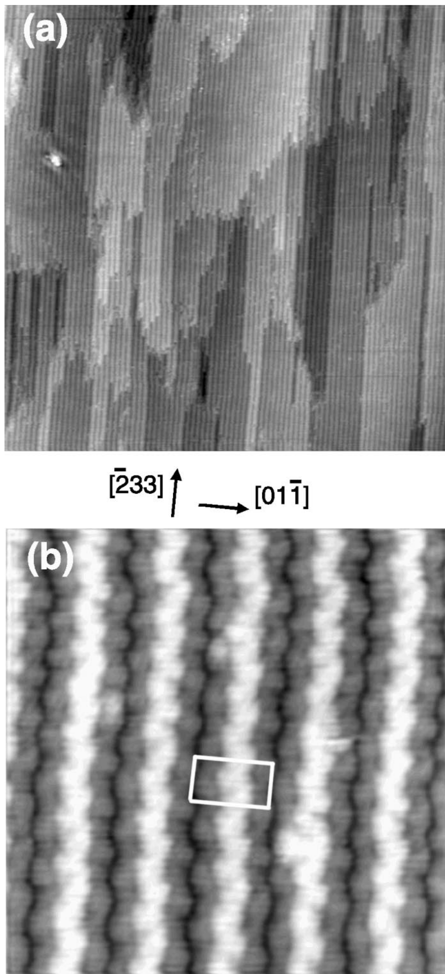

FIG. 3. (a) 300×300-nm^2 STM image showing the high density of steps along [$\bar{2}$33] on an 8×1-reconstructed surface. (b) Atomic resolution STM image of an 8×1 reconstruction. The reconstruction 8×1 unit cell is indicated.

this idea is that a change in surface reconstruction can act as a source of material for an increase in the step density. The important observation is that different surface reconstructions require a different density of surface atoms. Consequently, by considering the surface reconstruction shown in Figs. 1(b) and 3(b), we observe that a huge mass transport during the phase transition between Ga(2×1) and 8×1 is necessary. The change of surface atom density must be accomplished for the 8×1-reconstructed surface to form if it is starting from the Ga(2×1)-reconstructed surface. One possible way is to use the surface steps of the Ga(2×1)-reconstructed surface as the source of the mass change. In this scenario the mass transport involves long distance surface diffusion[29] while the existing steps on the Ga(2×1)-reconstructed surface provide the needed edge to absorb or released atoms by moving backward or forward. Another scenario, one more consistent with our observations, is illustrated in Fig. 5. Here, the released atoms in going from Ga(2×1) to 8×1 are accommodated by forming new steps thereby increasing, rather dramatically, the step density. That is, the reconstruction would randomly nucleate, and consequently released atoms migrate across the previous

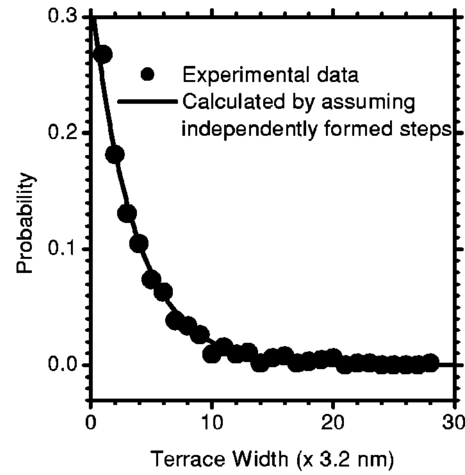

FIG. 4. Terrace width distribution of an 8×1-reconstructed surface. Circles are experimental results, and solid lines are calculated by assuming an independent step formation.

Ga(2×1)-reconstructed surface. If the existing steps cannot accommodate the released atoms, due to the long distance surface migration, additional steps must nucleate. In this case, the step formation is closely related to the nucleation of the 8×1 reconstruction, even though it is difficult to say which step is correlated to which nucleation event. Since the nucleation process is random, the step formation is expected to be independent from each other. For independently formed steps, the resulting terrace width distribution P_n is given by,[30]

$$P_n = P_1(1-P_1)^{n-1}. \quad (1)$$

where P_1 is the probability to find a step at a 3.2 nm unit and n is the terrace width divided by 3.2 nm. The value of P_1 is determined from the measured number of steps divided by the total length of terrace widths, which is 0.243. The solid line in Fig. 4 shows a good fit to the experimental data. To our knowledge, our result is the first report of an exponential terrace width distribution. While interesting in its own right, the significance here is that it supports the independent step formation model. The terrace width distribution has been widely studied to reveal the interaction between steps based on thermodynamic equilibrium models.[31–34] All other reported terrace distributions fit a peaked function because of step interaction or entopic repulsion.

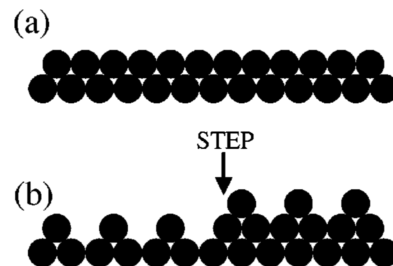

FIG. 5. Schematic drawing of the step formation due to the transition from the surface phase (a) to (b) to accommodate the change of the surface atom density.

To form the steps by the above mechanism, it appears that only one prerequisite has to be fulfilled: *the two stable surface reconstructions involved in the phase transition have a different density of surface atoms*. That is indeed the case for most of the surface reconstructions.[35–37] As a result, the step formation induced by the phase transition is expected to be a very common phenomenon. However, it is important to note that to experimentally observe the formation of steps by the phase transition, another requirement must be fulfilled. In particular, the surface diffusion mobility must be limited. Otherwise, the existing step edges would propagate and result in a smooth large terrace ripening. In the case of the GaAs(311) surface, the 8×1 reconstruction nucleates in two dimensions still but propagates mainly along the $[\bar{2}33]$ direction as indicated by the resulting surface morphology. Hence, the very limited surface atom mobility along $[01\bar{1}]$ is very important to guarantee the observation of the formation of the steps along $[\bar{2}33]$.

In summary, this letter reports the observation of a Ga(2×1)-reconstructed GaAs(311) surface. This is one of only a few stable, high index GaAs surfaces, for which the structure is clearly resolved. The surface is very flat on an atomic scale. The evolution of the surface morphology induced by the surface phase transition from a Ga(2×1)-reconstructed GaAs(311) surface to a 8×1-reconstructed GaAs(311) surface, was studied using STM and RHEED. The atomically flat Ga(2×1)-reconstructed wide terrace is transformed into many narrow 8×1-reconstructed terraces separated by randomly nucleated steps along the $[\bar{2}33]$ direction. These steps are formed to accommodate the change of the surface atom density required by the phase transition. Finally, the terrace width distribution is observed to follow an exponential probability. This observation supports the independent step formation model.

[1] P.M. Petroff, A. Lorke, and A. Imamoglu, Phys. Today **54**(5), 46 (2001).
[2] R. Notzel, Semicond. Sci. Technol. **11**, 1365 (1996).
[3] M. Wassermeier, J. Sudijono, M.D. Johnson, K.T. Leung, B.G. Orr, L. Daweritz, and K. Ploog, Phys. Rev. B **51**, 14 721 (1995).
[4] J. Marquez, L. Geelhaar, and K. Jacobi, Phys. Rev. B **62**, 9969 (2000).
[5] Z.M. Wang, L. Daweritz, and K.H. Ploog, Appl. Phys. Lett. **78**, 712 (2001).
[6] J. Marquez, P. Kratzer, L. Geelhaar, K. Jacobi, and M. Scheffler, Phys. Rev. Lett. **86**, 115 (2001).
[7] L. Geelhaar, J. Marquez, P. Kratzer, and K. Jacobi, Phys. Rev. Lett. **86**, 3815 (2001).
[8] Y. Hanein, N. Nenadovic, D. Shahar, H. Shtrikman, J. Yoon, C.C. Li, and D.C. Tsui, Nature (London) **400**, 735 (1999).
[9] S.J. Papadaki, E.P.D. Poortere, M. Shayegan, and R. Winkler, Phys. Rev. Lett. **84**, 5592 (2000).
[10] P.A. Ramos and E. Towe, Appl. Phys. Lett. **69**, 3321 (1996).
[11] M. Takahashi, M. Hirai, K. Fujita, N. Egami, and K. Iga, J. Appl. Phys. **82**, 4551 (1997).
[12] M. Higashiwaki, S. Shimomura, S. Hiyamizu, and S. Ikawa, Appl. Phys. Lett. **74**, 780 (1999).
[13] K. Tateno, Y. Ohio, C. Amano, A. Wakatsuki, and T. Kurokawa, Appl. Phys. Lett. **70**, 3395 (1997).
[14] P.O. Vaccaro, H. Ohnishi, and K. Fujita, Appl. Phys. Lett. **74**, 3854 (1999).
[15] R.P. Mirin, J.P. Ibbetson, K. Nishi, A.C. Gossard, and J.E. Bowers, Appl. Phys. Lett. **67**, 3795 (1995).
[16] H. Lee, R. Lowe-Webb, W. Yang, and P.C. Sercel, Appl. Phys. Lett. **72**, 812 (1998).
[17] Y. Hasegawa, H. Kiyama, Q.K. Xue, and T. Sakurai, Appl. Phys. Lett. **72**, 2265 (1998).
[18] J. Marquez, L. Geelhaar, and K. Jacobi, Appl. Phys. Lett. **78**, 2309 (2001).
[19] L.G. Wang, P. Kratzer, M. Scheffler, and N. Moll, Phys. Rev. Lett. **82**, 4042 (1999).
[20] I. Daruka, J. Tersoff, and A.L. Barabasi, Phys. Rev. Lett. **82**, 2753 (1999).
[21] R. Notzel, J. Temmyo, and T. Tamamura, Nature (London) **369**, 131 (1994).
[22] S. Lan, K. Akahane, H.Z. Song, Y. Okada, and M. Kawabe, J. Vac. Sci. Technol. B **17**, 1105 (1999).
[23] R. Notzel and K.H. Ploog, Jpn. J. Appl. Phys. **39**, 4588 (2000).
[24] R. Notzel, Z.C. Niu, M. Ramsteiner, H.P. Schonherr, A. Trampert, L. Daweritz, and K.H. Ploog, Nature (London) **392**, 56 (1998).
[25] Z.M. Wang, V.R. Yazdanpanah, J.L. Shultz, and G.J. Salamo, Appl. Phys. Lett., **81**, 2965 (2002).
[26] J. Marquez, L. Geelhaar, and K. Jacobi, Phys. Rev. B **65**, 165320 (2002).
[27] M. Kawase, Y. Ishikawa, and T. Fukui, Appl. Surf. Sci. **130-132**, 457 (1998).
[28] J.A.V. Vechten, J. Cryst. Growth **38**, 139 (1977).
[29] K. Kanisawa and H. Yamaguchi, Phys. Rev. B **56**, 12080 (1997).
[30] B.S. Swartzentruber, Y.-W. Mo, R. Kariotis, M.G. Lagally, and M.B. Webb, Phys. Rev. Lett. **65**, 1913 (1990).
[31] F. Lelarge, Z.Z. Wang, A. Cavanna, F. Laruelle, and B. Etienne, Europhys. Lett. **39**, 97 (1997).
[32] P. Hanesh and E. Bertel, Phys. Rev. Lett. **79**, 1523 (1997).
[33] E.D. Williams, Surf. Sci. **299/300**, 502 (1994).
[34] N. Bartelt, T. Einstein, and E. Williams, Surf. Sci. Lett. **240**, 591 (1990).
[35] D.K. Biegelsen, R.D. Bringans, J.E. Northrup, and L.E. Swartz, Phys. Rev. B **41**, 5701 (1990).
[36] Q. Xue, T. Hashizume, J.M. Zhou, T. Sakata, T. Ohno, and T. Sakurai, Phys. Rev. Lett. **74**, 3177 (1995).
[37] T. Hashizume, Q.K. Xue, J. Zhou, A. Ichimiya, and T. Sakurai, Phys. Rev. Lett. **73**, 2208 (1994).