Microscopic structure of GaSb(001) $c(2\times6)$ surfaces prepared by Sb decapping of MBE-grown samples

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In this study we report on the microscopic structure of GaSb(001) $c(2\times6)$ surfaces prepared by Sb decapping. Molecular beam epitaxy grown GaSb(001) layers capped with a protective Sb layer were transferred through the atmosphere into an UHV-analysis system and investigated by scanning tunneling microscopy (STM) and low energy electron diffraction (LEED). After thermal desorption of the capping layer clear $c(2\times6)$ LEED patterns were observed. STM images show flat surface areas with a rowlike, somewhat disordered structure. High-resolution images resolve individual Sb dimers on the surface. The surface is covered by an incomplete layer of dimerized Sb, adsorbed on a complete second layer of Sb, which is also dimerized in that regions not covered by the fractional Sb top layer. [S0163-1829(97)00503-1]

I. INTRODUCTION

Despite the technological relevance of III-V(001) surfaces, their structural and electronic properties are not well understood yet. Most of the basic research effort has concentrated on GaAs(001) surfaces so far. There are a number of experimental and theoretical studies that have been performed to determine the interplay between stochiometry, structure, and electronic properties.[1–6] Other III-V(001) surfaces are rather believed than known to reveal similar properties as found for the GaAs(001) model case.

One reason for the comparably few investigations of III-V(001) surfaces has to do with reproducible preparation of well ordered surfaces, since preparation by sputter/annealing leads to nonideal surfaces. Ideal surfaces can be investigated on *in situ* molecular beam epitaxy (MBE) grown epilayers, which, however, requires a combination of MBE and surface analysis equipment in the same ultrahigh vacuum (UHV) apparatus. Another solution of the preparation problem is the capping of MBE-grown layers by a protective adlayer for transfer through atmospheric surrounding to a separate UHV-analysis chamber, where finally a clean surface is restored by thermal desorption of the cap. This so-called decapping technique has successfully been used to study GaAs(100) surfaces.[7] Also for GaSb(001) it has been demonstrated that a Sb-capping layer is able to protect the surface against contamination during ambient storage: Auger electron spectroscopy and low energy electron diffraction (LEED) experiments have shown that clean, $c(2\times6)$ reconstructed GaSb(001) surfaces can be prepared by the Sb-decapping technique.[8] More recently, the $c(2\times6)$ reconstruction prepared by decapping was studied by means of reflectance anisotropy spectroscopy to access optical anisotropies related to surface dimers. However, in contrast to studies on GaAs(001) surfaces only weak surface anisotropy contributions were found.[9]

In this work we present a study of the microscopic surface structure of GaSb(001) $c(2\times6)$ surfaces prepared by Sb decapping. Atomically resolved scanning tunneling microscopy (STM) images show a structure built up by a fractional layer of dimerized Sb atoms adsorbed on a complete second Sb layer. This structure is similar to the As-terminated $c(4\times4)$ surface of GaAs(001), but less well ordered and both the outermost Sb layers are partly dimerized yielding Sb dimers along both the [110] and [$\bar{1}$10] direction.

II. EXPERIMENT

GaSb layers (p-doped, 3×10^{16} cm^{-3}) were prepared by MBE growth on GaAs(001) substrates after initial deposition of an AlSb nucleation layer. After growth an additional Sb-passivation layer was deposited on top of the GaSb.[9] The MBE grown Sb caps were investigated *ex situ* by Raman spectroscopy. After intermediate storage under nitrogen atmosphere, the samples were inserted into an UHV chamber equipped with STM and LEED facilities. Clean surfaces were prepared by thermal desorption of the Sb cap. LEED patterns of the decapped GaSb(001) surfaces were recorded using a four-grid reverse-view LEED optic equipped with a digital video camera. The STM images were taken in a constant current mode. The bias values refer to the sample voltage with respect to the tip held at virtual ground. All measurements were performed at a base pressure below 1×10^{-10} mbar.

III. RESULTS AND DISCUSSION

Surface preparation was performed by stepwise annealing of the samples up to at least 450 °C. For each annealing step changes in the LEED patterns were monitored after cooling to room temperature. At approximately 350 °C the desorption of most of the Sb-cap layer is indicated by a increase in pressure up to 5×10^{-8} mbar followed by a fast pressure decrease. For incomplete Sb desorption, facet spots in the LEED pattern are observed as being assigned to Sb crystallites of fixed orientation with respect to the GaSb(001) sub-

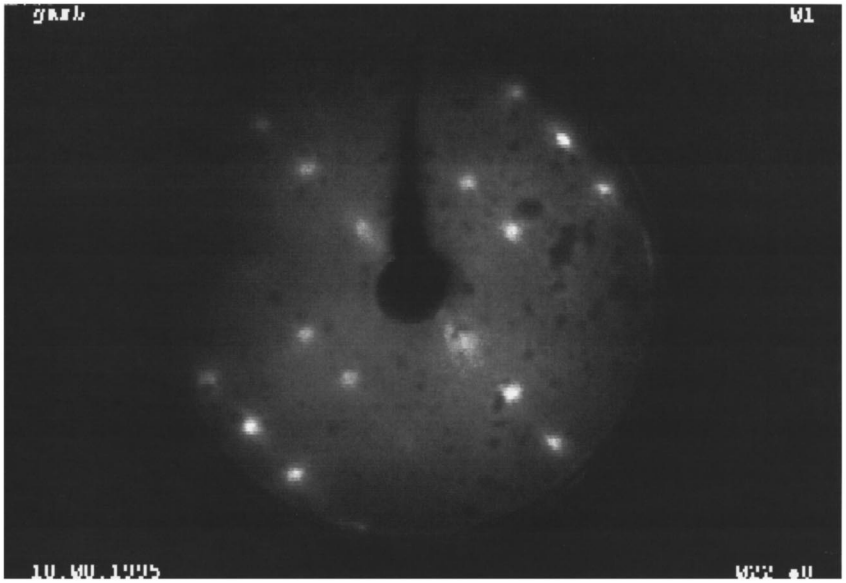

FIG. 1. LEED pattern taken at 22 eV electron energy, observed after annealing the sample at 450 °C. The $c(2\times6)$ structure is clearly revealed.

strate. This finding is supported by *ex situ* Raman experiments on the capped samples. They reveal the E_g and A_{1g} phonon modes of Sb indicating a crystalline capping layer. After complete desorption of the capping layer by further annealing a clear $c(2\times6)$ LEED pattern is observed (Fig. 1). The integral as well as the fractional order spots are sharp and intense, clearly revealing the centered symmetry of the reconstruction. Figure 2 shows a large scale STM image (500×500 Å^2) of a sample with a LEED pattern of optimum quality. At a sample bias of -4 V filled surface states are imaged. In accordance with previous results,[10] the STM image shows somewhat disordered rows mainly arranged in a distance of three lattice constants. At some locations, also fourfold distances are observed. Annealing to higher temperatures causes degradation of the surface, already by optical inspection the surface appears rough and another type of facet spots occurs in the LEED patterns. These can either be due to the formation of GaSb facets or Ga islands upon prolonged annealing.[8,9]

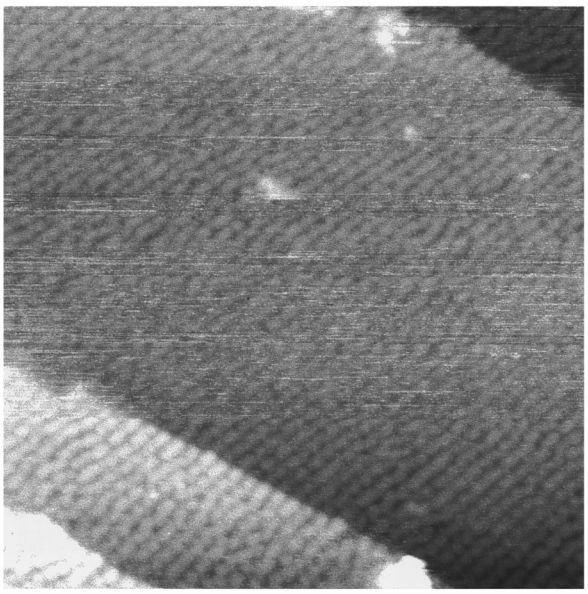

FIG. 2. STM image (500×500 Å^2, sample bias -4 V) of the $c(2\times6)$ reconstructed GaSb(001) surface. Somewhat disordered rows along the $[1\bar{1}0]$ direction mainly arranged in a distance of three lattice constants appear.

The microscopic structure of the $c(2\times6)$ GaSb(001) surface has previously been investigated by Franklin *et al.* on MBE-grown samples by soft x-ray photoemission spectroscopy (SXPS), MEED, and STM.[10] SXPS revealed two Sb $4d$- and one Ga $3d$-surface components besides the bulk emission lines. From the surface core level shifts and the intensities of the Sb and Ga emission lines a basic (1×3) surface structure was proposed: the topmost layer should contain Sb and Ga atoms, the Sb forming dimers oriented along the $[1\bar{1}0]$ direction allocated in rows along the $[110]$ direction, separated by rows of single Ga atoms. This topmost layer should be adsorbed on a complete second layer of Sb. Their STM images supported the presence of the proposed Sb-dimer rows in the outermost layer. The dimer rows, however, were not perfectly ordered, and the $c(2\times6)$ symmetry of the diffraction pattern could not be identified in the STM images. Thus, even if the large scale STM patterns observed in this study are in accordance with those of Franklin *et al.*, their preliminary structure model cannot explain the real structure giving rise to clear $c(2\times6)$ LEED patterns as observed on the decapped GaSb(001) surfaces. This discrepancy should be resolved by high-resolution STM images.

Figure 3 shows a high resolution STM image taken in the surrounding of a step edge. On a scale of 105×105 Å, the rows are resolved, they consist of individual elongated features oriented perpendicular to the rows. These features should represent individual Sb dimers of the outermost layer. The partial disorder of the surface is a consequence of the fact that after a sequence of several Sb dimers (most times three dimers) along the $[1\bar{1}0]$ direction the rows are inter-

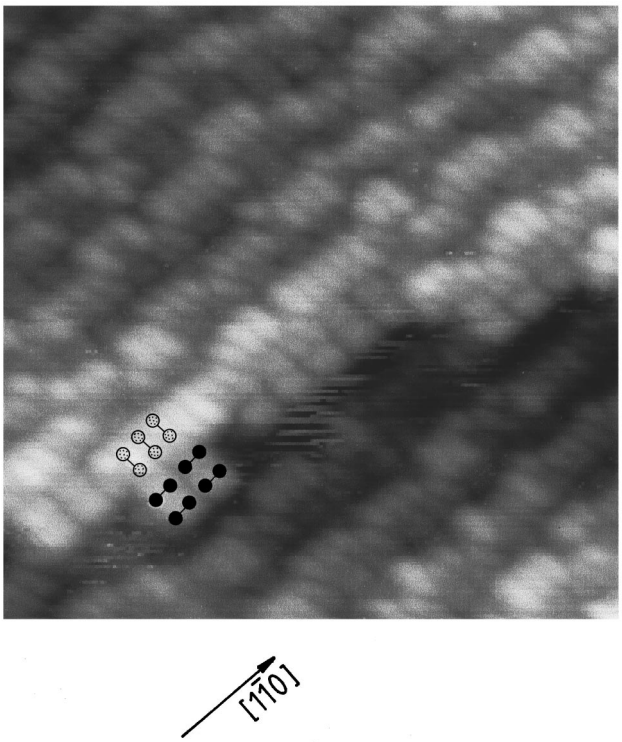

FIG. 3. High-resolution STM image (105×105 Å^2, sample bias -2.18 V). The rows consist of individual features. The elongated ones represent individual Sb dimers, the round features correspond to single Sb atoms. At the step edge pairs of elongated protrusions along the [110] direction are clearly observed, which indicate the existence of Sb dimers in the second layer, perpendicular to those of the outermost layer. On the terraces the second-layer Sb dimers also occur in between the rows of first-layer Sb dimers.

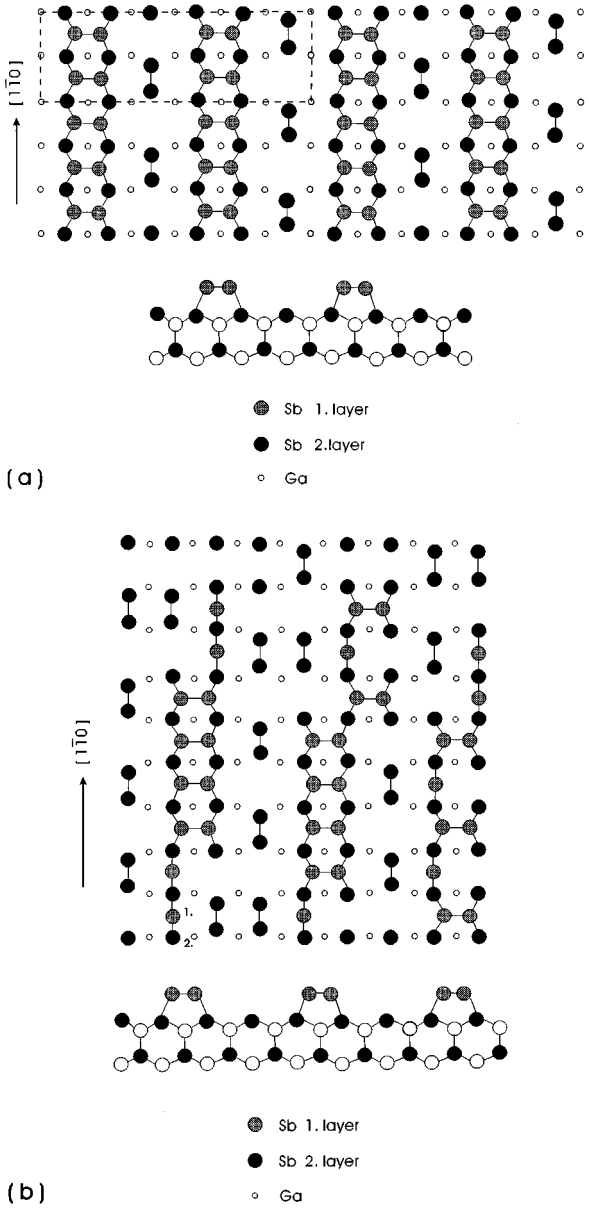

FIG. 4. Model for the $c(2\times6)$ reconstructed GaSb (001) surface. The marked centered surface unit mesh contains four Sb dimers along the [110] direction in the topmost layer and two Sb dimers one double layer below, perpendicular to those of the topmost layer. (a) shows the ideal arrangement, and (b) the real structure, as observed in STM.

rupted by a shift of the following dimers by one lattice constant in the [011] direction. Since the shifts of neighboring rows are not in phase, local regions are formed where the dimer rows are separated by four lattice constants instead of three. Another source of disorder arises from the fact that not all of the Sb atoms of the outermost layer are dimerized. At many locations, the protrusions in the rows are not elongated but round shaped. These round protrusions are apparently related to single Sb atoms in the outermost layer, occurring as a consequence of a missing Sb atom required for the dimerization. Between the Sb-dimer rows and at the step edge the second atomic layer, also consisting of Sb atoms, is exposed. Especially at the step edge and at that regions on the terrace where the distance between the topmost dimer rows is larger than three lattice constants, pairs of elongated protrusions along the [110] direction are clearly visible in the STM images. These pairs of protrusions indicate the existence of a double row of Sb dimers in the second layer, oriented along the [110] direction. In some STM images (not shown here) local regions at step edges are imaged where the second-layer Sb dimers of the double row are shifted by one lattice constant with respect to each other, clearly revealing the orientation of the dimers along the [110] direction. Apart from the step edges, also on the terraces the dimerization of the second layer Sb is clearly evident (see Fig. 3). These second-layer dimers give rise to the twofold periodicity along the [110] direction found in the LEED pattern, whereas the separation of the Sb-dimer rows of the outermost layer build up the threefold periodicity along the [110] direction. A centered arrangement is derived by a shift of neighboring second-layer Sb-dimer rows by one lattice constant along the [110] direction. In fact, in the STM images (see Fig. 3) locations revealing a shift of neighboring second-layer Sb dimers can be identified. All these STM results corroborate thus with the structure model depicted in Fig. 4, keeping in mind that some degree of disorder is also part of the microscopic surface structure.

The previous structure model of Franklin *et al.*, in contrast, can be ruled out with certainty. First, it does not explain the $c(2\times6)$ LEED pattern which has been observed by several groups.[8,9] Even more convincing, furthermore, is the fact that the presence of single Ga atoms in between the outermost Sb-dimer rows can be ruled out unambiguously, since our high-resolution STM images show dimerized second-layer Sb there. In comparison with the much better characterized GaAs(001) surface structures we find that the GaSb $c(2\times6)$ structure is rather similar to the $c(4\times4)$ GaAs surface.[2] Both surfaces are composed of two atomic layers of group V atoms. In both case the outermost group V atoms form dimers. The main difference is the structural arrangement of the outermost dimers. In case of GaAs each As atom of the second layer has at least one bonding to the As dimers of the outermost layer, thus no dimerization of the second layer occurs. In case of GaSb $c(2\times6)$, instead, the second-layer Sb in the intermediate rows has two dangling bonds giving rise to a dimerization of these Sb atoms.

IV. CONCLUSION

In conclusion, we have investigated MBE grown GaSb(001) surfaces by LEED and STM under UHV conditions. We have shown that the Sb-rich $c(2\times6)$ surface of GaSb(001) consists of a Sb-dimer structure adsorbed on a second layer of Sb. The outermost Sb dimers form rows along the $[\bar{1}10]$-direction terminated by shifts of a single lattice constant in the [011] direction. The second Sb layer is dimerized where it is uncovered, i.e., between the rows of the outermost Sb dimers.

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[1] M.D. Pashley, Phys. Rev. B **40**, 10 481 (1989).
[2] D.K. Biegelsen, R.D. Bringans, J.E. Northtrup, and L.-E. Swartz, Phys. Rev. B **41**, 5701 (1990).
[3] J. Northrup and S. Froyen, Phys. Rev. B **50**, 2015 (1994).
[4] T. Hashizume, Q.K. Xue, J. Zhou, A. Ichimiya, and T. Sakurai, Phys. Rev. Lett. **73**, 2208 (1994).
[5] Q. Xue, T. Hashizume, J.M. Zhou, T. Sakata, T. Ohno, and T. Sakurai, Phys. Rev. Lett. **74**, 3177 (1995).
[6] A.R. Avery, C.M. Goringe, D.M. Holmes, J.L. Sudijono, and T.S. Jones, Phys. Rev. Lett. **76**, 3344 (1996).
[7] U. Resch-Esser, N. Esser, D.T. Wang, M. Kuball, J. Zegenhagen, B.O. Fimland, M. Cardona, and W. Richter, Surf. Sci. **352-354**, 71 (1996); U. Resch, N. Esser, Y.S. Raptis, W. Richter, J. Wasserfall, A. Förster, and D.I. Westwood, *ibid.* **269/270**, 797 (1992).
[8] M. Dumas, N. Nouaoura, N. Bertru, and L. Lassabatere, Surf. Sci. Lett. **262**, L91 (1992).
[9] C. Goletti, U. Resch-Esser, J. Foeller, N. Esser, W. Richter, B. Brar, and H. Kroemer, Surf. Sci. **352-354**, 771 (1996).
[10] G.E. Franklin, D.H. Rich, A. Samsavar, E.S. Hirschorn, F.M. Leibsle, T. Miller, and T.-C. Chiang, Phys. Rev. B **41**, 12 619 (1990).