Reflectance-difference spectroscopy of mixed arsenic-rich phases of gallium arsenide (001)

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The relationship between the reflectance difference spectra and the atomic structure of arsenic-rich reconstructions of GaAs(001) has been investigated. Scanning tunneling micrographs reveal that a roughening process occurs as the surface structure changes with decreasing arsenic coverage from 1.75 to 0.75 monolayers (ML). At 1.65 ML As, small pits, one bilayer in depth and having the same $c(4\times4)$ reconstruction as the top layer, form in the terraces. At the same time, gallium atoms are liberated to the surface, disrupting the $c(4\times4)$ ordering. At about 1.4 ML As, (2×4) domains nucleate and grow on top of the $c(4\times4)$. Further desorption of arsenic causes the underlying layer to gradually decompose into a metastable $(2\times n)$ phase ($n=2$, 3, or 4), and finally into the (2×4). In the reflectance difference spectra, negative peaks at 2.25 and 2.8 eV correlate with the $c(4\times4)$-type arsenic dimers. However, the intensity of the latter feature strongly depends on the presence of adsorbates, such as alkyl groups and gallium adatoms. By contrast, the intensity of the positive peak at 2.9 eV is directly proportional to the density of (2×4)-type dimers.

I. INTRODUCTION

Reflectance difference spectroscopy (RDS) is an effective *in situ* probe of the surface reconstructions of compound semiconductors during growth by molecular-beam epitaxy, chemical beam epitaxy, and metalorganic vapor-phase epitaxy.[1–4] The technique determines the relative difference in the near-normal reflectance of light polarized along the two principle axes of the surface, and since the bulk crystals are isotropic, the spectra quantify the optical anisotropy of the first few atomic layers. The reconstructions of gallium arsenide (001) have been widely studied using this technique.[5–7] While the line shapes of the $c(4\times4)$, $(2\times4)/c(2\times8)$, and $(4\times2)/c(8\times2)$ surfaces have been correlated with electron diffraction patterns [reflection high-energy electron diffraction and low-energy electron diffraction],[1] the physical origin of the reflectance anisotropy has not been conclusively determined. Uncertainty remains as to whether the reflectance anisotropy is a result of bulk-surface transitions,[8–12] or by transitions among the molecular orbitals of the surface dimers.[13,14]

Early efforts to account for the reflectance anisotropy involved the calculation of the surface dielectric function of simplified GaAs surfaces.[13,14] In these studies, (2×1) and (1×2) dimerized surfaces were used to approximate the arsenic-rich (2×4) and gallium-rich (4×2) reconstructions. The authors identified transitions within the dimer structures as the source of the RDS spectra, and obtained rough qualitative agreement with the experimental data. Based on more recent, first-principles calculations, other researchers have concluded that transitions between bulk valence states and unoccupied surface states are primarily responsible for the reflectance anisotropy.[9–12] While these results more closely match the RDS data, discrepancies in the energies and magnitudes of spectral features remain. The best agreement between theory and experiment has been achieved through tight-binding calculations of the dimer structures.[8] The influence of both discrete dimer structures and surface-modified bulk wave functions were cited as contributing factors to the observed anisotropy.

In this paper, we report on the reflectance difference spectra of a series of gallium arsenide (001) reconstructions at arsenic coverages ranging from 1.75 to 0.75 monolayers (ML). The main structural features on these surfaces are arsenic dimers, which are bonded to either a sublayer of As or Ga atoms. These two kinds of arsenic dimers are referred to as ''$c(4\times4)$-type'' and ''(2×4)-type,'' respectively. To aid in the interpretation of the spectra, a direct comparison has been made between the RDS line shapes and the atomic structures as seen by scanning tunneling microscopy (STM). We have found that surface roughening occurs as a result of the nucleation and growth of the (2×4) phase on top of the $c(4\times4)$ phase. This process is accompanied by gallium out diffusion into the $c(4\times4)$ layer. Furthermore, we have discovered that the reflectance anisotropy is affected not only by the types of arsenic dimers, but also by the presence of transitional structures, such as a new $(2\times n)$ phase, and by adsorbates, including alkyl species and gallium adatoms.

II. EXPERIMENTAL METHODS

Gallium arsenide films, approximately 1 μm in thickness, were grown on nominally flat GaAs (001) substrates in a horizontal flow metalorganic vapor-phase epitaxy (MOVPE) reactor. The substrates were doped *n*-type with 1×10^{17} Si atoms/cm^3. The wafer temperature during growth was $550\pm25\,°C$, and the organometallic reagents, triisobutylgallium (TIBGa) and tertiarybutylarsine (TBAs), were used at concentrations of 5 and 50 ppm, respectively. Palladium-diffused hydrogen was the carrier gas, and the total reactor pressure was 20 torr. The wafers were cooled immediately following growth with the TBAs and H$_2$ supplies maintained until room temperature was reached. This ensured that the surface would have the maximum arsenic coverage possible. The samples were transferred to an ultrahigh vacuum cluster tool via a turbo-pumped interface cham-
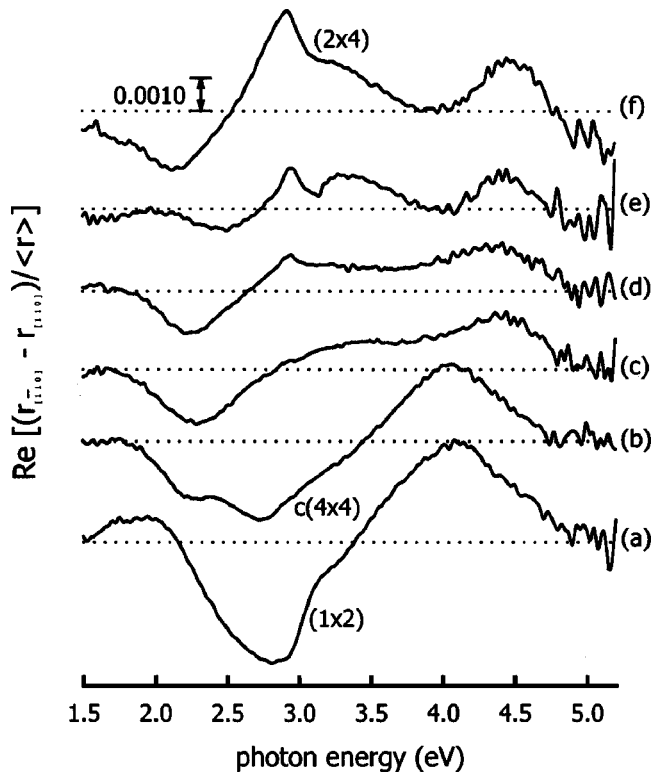
FIG. 1. Reflectance difference spectra of GaAs (001) surfaces with arsenic coverages of (a) >1.75 ML, (b) 1.75 ML, (c) 1.65 ML, (d) 1.4 ML, (e) 1.2 ML, and (f) 0.75 ML. The spectra corresponding to the (1×2), $c(4\times4)$ and (2×4) surfaces are indicated.

ber. Details of this system have been described previously.[15]

A commercially available ISA/J-Y Nisel reflectance difference spectrometer was used for our measurements of the GaAs surfaces. The spectral range varied from 1.5 to 5.2 eV, with increments of 0.025 eV and integration times of 1000 ms. Three scans were recorded consecutively and then averaged to minimize the noise. Real-space images of the reconstructions were obtained using a Park Autoprobe/VP scanning tunneling microscope. Tunneling was out of filled states with a sample bias of −3.0 to −4.0 V and with a tunneling current of 1 nA.

To create transitional surfaces, the samples were heated on a manipulator stage at temperatures ranging from 300 to 500 °C, while monitoring the RDS signal at a constant energy of 2.8 eV. The signal increased from a starting value of −0.004 to a maximum value of +0.003. Transitional line shapes were obtained by interrupting heating when the signal

was between −0.0005 and +0.0010. The pressure in the chamber did not exceed 2×10^{-9} torr during heating or cooling. All spectra reported in the paper were taken after the sample had cooled to 20 °C.

III. RESULTS

Presented in Fig. 1 are reflectance difference spectra of gallium arsenide (001) surfaces with arsenic coverages varying from 1.75 to 0.75 ML. The $(1\times2)/d(4\times4)$, $c(4\times4)$, and (2×4) line shapes are labeled (a), (b), and (f), respectively, and are consistent with previously published spectra. The intermediate line shapes are labeled (c), (d), and (e). The (1×2) RDS spectrum was recorded on GaAs crystals right after removal from the reactor, while the other spectra were obtained following annealing of the crystals. Note that the magnitude of the 2.8 eV feature in the RDS spectrum of the (1×2) is -0.0040 ± 0.0002.

The most noticeable changes that occur with the decrease in arsenic coverage are the disappearance of the negative peak at 2.8 eV and the emergence of the positive peak at 2.9 eV. However, more subtle changes may be detected as well. For example, the negative band in spectra (a) through (c) is composed of two minima at 2.25 and 2.8 eV. The former feature remains after the latter one has disappeared, as seen in line shape (c). There is also an abrupt shift in the position of the positive band in the higher-energy range from curve (b) to curve (c). The positive peak for the $c(4\times4)$ surface is centered at 4.0 eV, while for the first transitional phase, it is at 4.4 eV. Comparison of spectra (d) through (f) reveals two overlapping bands at 2.9 and 3.25 eV. In addition, the negative band at 2.25 eV in (d) disappears in (e) and reappears at 2.2 eV in (f). None of the spectral features in Fig. 1 appears to be related to the linear electro-optic effect.[16–18] This is supported by secondary-ion-mass spectroscopy data on the GaAs epitaxial films, which yielded impurity concentrations ranging from 5×10^{-15} to 5×10^{-16} cm^{-3}.

For each RDS spectra in Figs. 1(a), 1(b), and 1(c), Figs. 2(a), 2(b), and 2(c) displays the corresponding scanning tunneling micrographs. Image (a) is of the (1×2) reconstruction. This surface consists of a random distribution of arsenic dimers, alkyl groups, and hydrogen atoms adsorbed on top of a complete layer of arsenic atoms.[15] The As dimer bonds are oriented along the [110] direction, yielding local $\times2$ periodicity. In image (b), the alkyl groups and H atoms have desorbed, and well-ordered groups of three arsenic dimers are observed (i.e., the gray rectangles). This pattern is characteristic of the $c(4\times4)$ reconstruction.[15,19] The $c(4\times4)$ was
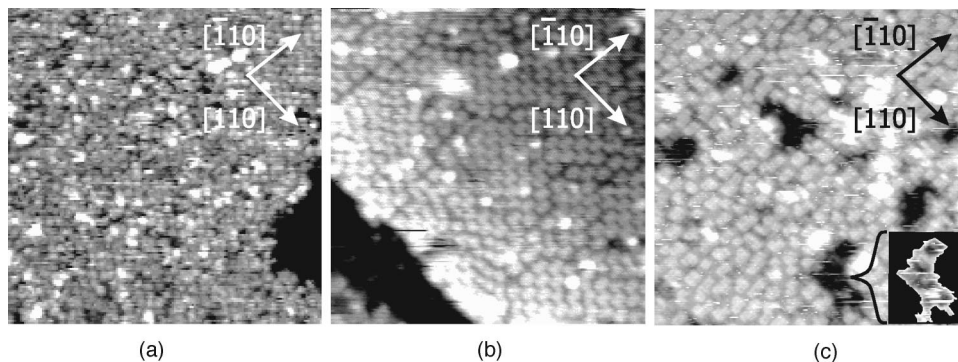

FIG. 2. Scanning tunneling micrographs of GaAs (001) surfaces with arsenic coverages of (a) >1.75 ML, (b) 1.75 ML, and (c) 1.65 ML. The image sizes are 400×400 Å^2, 320×320 Å^2, and 300×300 Å^2, respectively.

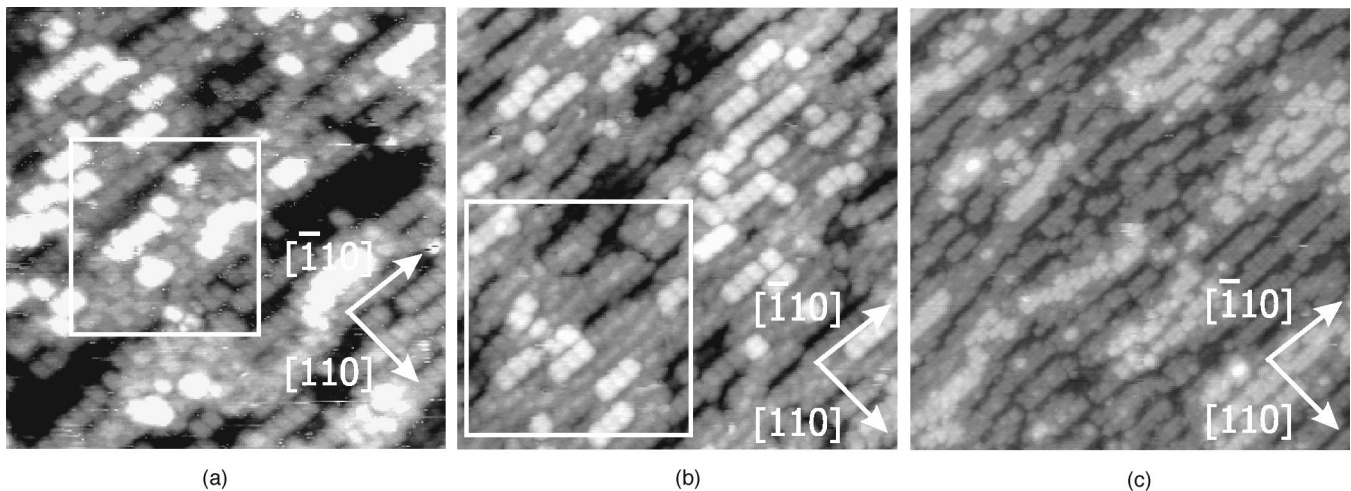(a)   (b)   (c)

FIG. 3. Scanning tunneling micrographs of GaAs (001) surfaces with arsenic coverages of (a) 1.4 ML, (b) 1.2 ML, and (c) 0.75 ML. The image sizes are 250×250 Å^2, 210×210 Å^2, and 270×270 Å^2, respectively.

produced by heating the sample until the reflectance-difference signal at 2.8 eV had decreased from -0.004 to -0.003.

After further heating of the GaAs sample to reduce the intensity of the 2.8 eV peak to zero, the surface becomes covered with pits one bilayer in depth, as shown in image (c). By increasing the brightness of the image, it can be seen that the reconstruction within these pits is the same as the top layer. In the inset picture, a pit has been extracted onto a black background, and the image contrast increased to reveal the surface structure. For these pits to form with the same structure as the main surface, arsenic atoms in the second layer must exchange with gallium atoms in the third layer. Since the area of the pits was measured to be approximately 10% of the total area, the arsenic lost is estimated at about 0.1 ML. This small decrease in arsenic coverage causes the once well-ordered groups of three arsenic dimers to become distorted in the [110] and [110] directions. However, despite the disordering, the entire surface is covered with structural groups that closely resemble the $c(4 \times 4)$ phase. These results demonstrate that the negative feature at 2.8 eV is not solely an indicator of the coverage of $c(4 \times 4)$-type dimers, as has been previously proposed.[1,8]

With the emergence of the positive feature at 2.9 eV in the RDS spectra, (2×4) unit cells appear on the surface. Presented in Figs. 3(a), 3(b), and 3(c) are STM images showing increasing numbers of (2×4) domains. These micrographs correspond to the spectra of Figs. 1(d), 1(e), and 1(f), respectively. The distance traversed from white to black in the STM images is approximately 5.4 Å, or a single GaAs lattice constant. The (2×4) unit cells appear as white or gray rectangles, with the long axis in the [110] direction. Multiple (2×4) unit cells are aligned in rows along the [110] direction, and are delineated by the dark dimer vacancies separating each row.[19] Three atomic bilayers, corresponding to white, gray, and black, are exposed on these surfaces because of the phase transition. In Fig. 4, the intensity of the 2.9 eV peak in the RDS spectrum is plotted against the areal density of (2×4) unit cells. A linear correlation is observed between the peak intensity and the coverage of (2×4)-type dimers.

In Fig. 3(a) and 3(b), the (2×4) unit cells are visible in the topmost layer. However, in the layer below, a predominantly $c(4 \times 4)$ reconstruction is seen in (a), while a new transitional structure is observed in (b). The boxed regions of each of these images have been enlarged and are displayed in Figs. 5(a) and 5(b). In the left-hand picture, the gray rectangular patches, highlighted with white outlines, are groups of two and three arsenic dimers. The periodicity in the [110] direction is disrupted, but this phase is nevertheless recognizable as $c(4 \times 4)$. In the right-hand picture, the structure below the white (2×4) unit cells has $2 \times$ periodicity in the [110] direction, whereas the repeat spacing in the [110] direction varies from $\times 2$ (8 Å) to $\times 3$ (12 Å) to $\times 4$ (16 Å) (as indicated by the highlighted white rectangles). Consequently, we denote this phase as $(2 \times n)$. Referring back to spectrum (e) in Fig. 1, the appearance of the $(2 \times n)$ in the STM image corresponds with the emergence of a positive RDS peak at 3.25 eV.

Line scans through the STM images in Fig. 5 have revealed that the gray rectangles for the $c(4 \times 4)$ and the light gray ridges for the $(2 \times n)$ are at the same height. They are both $0.35a$ below the white rectangles of the (2×4) in the top layer, where a is the GaAs lattice constant. Based on these data, we conclude that the uppermost ridges of the $(2 \times n)$ must be due to arsenic dimers bonded to arsenic atoms
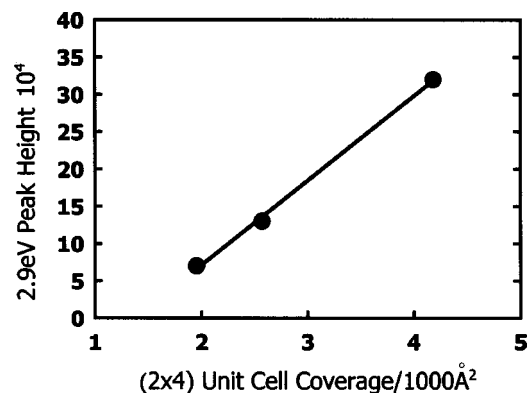

FIG. 4. The dependence of the height of the 2.9-eV-RDS peak on the areal coverage of (2×4) unit cells.
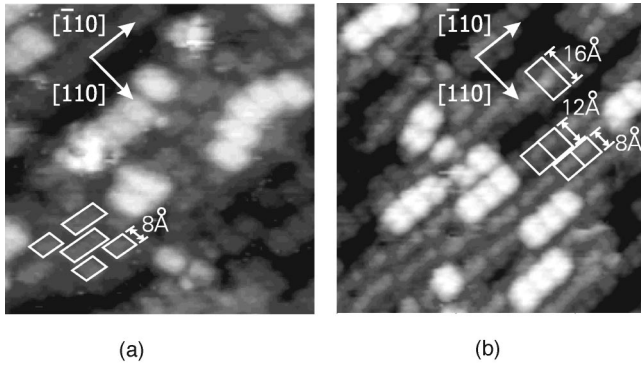
FIG. 5. Close-up scanning tunneling micrographs of GaAs (001) surfaces with arsenic coverages of (a) 1.4 ML and (b) 1.2 ML. Both images are 100×100 Å^2.
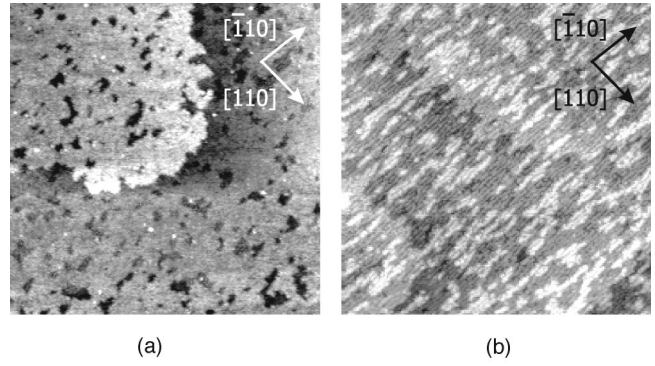

FIG. 6. Large-scale scanning tunneling micrographs of GaAs (001) surfaces with arsenic coverages of (a) 1.65 ML and (b) 0.75 ML, showing the initial and final stages of the $c(4 \times 4)$ to (2×4) phase transition.

in the second layer. For this to be the case, the dimer bonds must be parallel to the [110] crystal axis. However, the white spots along the ridges appear to be elongated in the [1̄10] direction, not the [110]. To reconcile this discrepancy, we propose that the $(2 \times n)$ ridges correspond to buckled dimers, in which the buckled up atom with the filled lone pair produces the light-gray spots. The buckled down atoms with a half-filled or empty lone pair merges with the dark-gray ledges adjacent to the brighter spots. Such a structure is akin to the (2×1) reconstruction of InP (001).[20] Since this is a metastable phase that occurs during the transition from the $c(4 \times 4)$ to the (2×4), it seems reasonable that the structure does not follow the electron counting model.[21] Nevertheless, more studies are needed of the $(2 \times n)$ phase to identify the arrangement of the atoms within each unit cell.

In order to more clearly illustrate the surface roughening that occurs with the $c(4 \times 4)$ to (2×4) phase transition, two large-scale micrographs are displayed in Figs. 6(a) and 6(b). These pictures correspond to arsenic coverages of 1.65 and 0.75 ML, respectively. The random formation of pits is evident in the first picture, and the roughness of the step edges indicates that arsenic desorption occurs there as well as on the terraces. In the latter picture, the terraces have two atomic bilayers exposed due to the nucleation and growth of small (2×4) islands on top of the original $c(4 \times 4)$ surface.

IV. DISCUSSION

While the nature of the homogeneous GaAs (001) (1×2), $c(4 \times 4)$, and (2×4) reconstructions has been well documented, including the reflectance difference spectra of these surfaces,[1–7,19,21–24] the origin of the optical anisotropy is not well understood. Theoretical calculations have reproduced to varying degrees the RDS spectra of the ''ideal'' reconstructed surfaces.[8,9,11] However, deviations in feature positions and intensities indicate that these models do not account for all of the factors influencing the optical transitions. In addition, the interpretation of RDS line shapes intermediate between those characteristic of the $c(4 \times 4)$ and (2×4) structures is incomplete, and could provide additional insight into the nature of these nonequilibrium semiconductor surfaces. Our method of comparing RDS line shapes directly to STM images of the surface allows us to examine how spectral features are related to the coverage and arrangement of dimer groups on the surface. Regardless

of the exact origin of the reflectance anisotropy, this benchmarking of spectra is essential if one is to use RDS as a practical tool for *in situ* monitoring during compound semiconductor crystal growth.

A. The surface phase transition

The $c(4 \times 4)$ surface is covered with ordered rows of arsenic dimers above a complete monolayer of arsenic atoms, with the dimer bonds parallel to the [110] direction, while the (2×4) surface is comprised of arsenic dimers above a monolayer of gallium, with the dimer bonds oriented parallel to the [1̄10] direction.[19,22] The changes observed in the RDS spectra with decreasing arsenic coverage can be understood after first considering the phase transition as observed by STM. Starting with the (1×2), heating below 400 °C causes the hydrocarbons, hydrogen, and some arsenic to desorb, producing a well-ordered $c(4 \times 4)$ structure. A further reduction in the As coverage by only 0.1 ML generates a distorted $c(4 \times 4)$ with desorption pits appearing randomly throughout the terraces. With additional loss of about 0.25 ML As, (2×4) domains nucleate and grow on top of the $c(4 \times 4)$ phase. The (2×4) islands continue to grow as the underlying $c(4 \times 4)$ gradually decomposes into the $(2 \times n)$ and then the (2×4) reconstruction.

The transition observed in the STM images of Fig. 2 and 3, and described above, involves desorption of arsenic, exchange of As and Ga atoms, and migration of Ga atoms on the surface. The existence of the $c(4 \times 4)$ reconstruction in the desorption pits requires that the second-layer As atoms exchange with the Ga atoms below. This liberates gallium atoms, which then migrate over the $c(4 \times 4)$ surface. *Ab initio* calculations have been performed to identify the lowest-energy binding sites of Ga adatoms on the $c(4 \times 4)$ surface.[25] It was found that for dilute coverages, the adatoms occupy missing As-dimer sites and sites between dimer rows adjacent to the As dangling bonds. Gallium adatoms in these positions could account for the distortion of the $c(4 \times 4)$ rows, as observed in Fig. 2(c).

A mass balance was performed to estimate the number of gallium atoms liberated to the surface in Fig. 2(c). Between 6 and 8 Ga atoms must be replaced by As atoms to form a single group of three $c(4 \times 4)$-type dimers in the pits, while 6 to 8 Ga atoms must come into close proximity to form the

second layer of every (2×4) unit cell. For a total image area of 1652 nm^2, the area of the desorption pits is estimated to be 145 nm^2. The white patches, although not well resolved, are assumed to be (2×4) unit cells. By comparing the area of these unit cells to that of the desorption pits, we determined that at most 25 to 33% of the free gallium atoms reside in (2×4) cells. The remainder must be in the adatom positions. As more arsenic desorbs, the number of diffusing gallium atoms increases, and greater numbers of (2×4) unit cells nucleate on top of the $c(4\times4)$ structure.

Kanisawa and Yamaguchi[26] have examined the transition of GaAs (001) from the $c(4\times4)$ to the (2×4) reconstruction. In their study, they converted the $c(4\times4)$ to the (2×4) by heating the substrate under a constant As$_4$ flux. This phase transition could be reversed by cooling the sample in the arsenic beam. In agreement with our paper, these authors observed the formation of (2×4) domains on top of a $c(4\times4)$-like phase. However, they erroneously concluded that this process must involve gallium detachment from step edges and diffusion of long distances over the terraces. The STM image presented in Fig. 6(a) clearly shows pits resulting from the gallium out-diffusion from the terraces as well as from the step edges.

Morgan, Kratzer, and Scheffler[27] have made first-principles calculations of As$_2$ adsorption on the (2×4) surface. Their calculations indicate that As$_2$ adsorbs directly on top of the groups of two arsenic dimers terminating each (2×4) unit cell. They argued that this direct addition of arsenic to the (2×4) can lead to the nucleation and growth of the $c(4\times4)$ phase. Our results clearly demonstrate that the opposite situation holds, with the (2×4) nucleating and growing on top of the $c(4\times4)$.

B. Interpretation of RDS spectra

We now consider the changes in the RDS spectra in light of the STM images. In previous studies, the negative peak at 2.8 eV has been correlated with the presence of the $c(4\times4)$ reconstruction.[1] This peak can be seen in spectra (a) and (b) in Fig. 1, but not in spectrum (c). All three of these surfaces are covered with $c(4\times4)$-type dimers. The only real difference between them is that adsorbates are present on some of the surfaces: the (1×2) corresponding to RDS spectrum (a) is partially covered with alkyl groups and hydrogen; the $c(4\times4)$ corresponding to RDS spectrum (b) is clean; and the disordered $c(4\times4)$ corresponding to RDS spectrum (c) is partially covered with gallium adatoms. Therefore, it is concluded that the negative peak at 2.8 eV is due to optical transitions involving $c(4\times4)$-type dimers, but that these transitions are strongly influenced by the presence of other adsorbates.

An additional RDS feature that is recorded when $c(4\times4)$-type dimers occur on the surface is the negative peak at 2.25 eV. This peak is evident in spectra (b), (c), and (d) in Fig. 1. Examination of the matching STM pictures [Fig. 2(b) and 2(c) and Fig. 3(a)] reveals that all three of these surfaces

contain significant densities of $c(4\times4)$-type dimers. By comparison to the negative peak at 2.8 eV, this transition appears to be less affected by adsorbates.

It should be noted that the line shape shown in Fig. 1(c) very nearly matches the ''Type-2'' spectrum reported for *in situ* MOVPE growth of gallium arsenide.[6,28] The Type-2 spectrum was observed for conditions of higher gallium flux and lower temperature than those that yielded the RDS spectrum corresponding to the (1×2).[6] Such conditions would increase the gallium adatom coverage above that on the (1×2) surface. Thus, the Type-2 RDS spectrum reported in the MOVPE growth studies may arise from a disordered $c(4\times4)$ with a low coverage of gallium adatoms.

In RDS spectra (d), (e), and (f) in Fig. 1, a positive band at 2.9 eV is observed. This feature may be assigned to an optical transition involving (2×4)-type dimers. The corresponding STM images, Figs. 3(a), 3(b), and 3(c), all contain (2×4) unit cells in the first and second bilayers. Moreover, as shown in Fig. 4, the intensity of this peak is directly proportional to the coverage of the (2×4)-type dimers.

In RDS spectra (e) and (f), a positive band at 3.25 eV is observed. This feature may be related to the transitional $(2\times n)$ phase, which can be seen in both of the corresponding STM images [Figs. 3(b) and 3(c)]. In Fig. 3(c), the $(2\times n)$ phase is somewhat difficult to discern, but can be found in the borders surrounding the top-layer (2×4) islands. Nevertheless, further experimental and theoretical studies of the $(2\times n)$ are needed to definitively assign the 3.25-eV band to this structure.

V. CONCLUSIONS

On gallium arsenide (001) surfaces with As coverages ranging from 1.75 to 0.75 ML, we have directly correlated reflectance difference spectra to scanning tunneling micrographs. Upon arsenic desorption, a complex roughening process occurs with the formation of pits in the $c(4\times4)$ terraces. This liberates gallium adatoms onto the surface, which causes the $c(4\times4)$ to become disordered. Continued arsenic desorption results in the nucleation of (2×4) domains on top of the $c(4\times4)$, and the gradual conversion of this underlying structure into a metastable $(2\times n)$ and finally (2×4) phase. For surfaces covered primarily with $c(4\times4)$-type dimers, the intensity of the negative RDS band at 2.8 eV is not proportional to the dimer coverage, but instead is strongly influenced by the presence of alkyl, hydrogen, and gallium adsorbates. In contrast, the magnitude of the positive feature at 2.9 eV is linearly proportional to the coverage of (2×4)-type dimers.

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