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# Interaction of selenium with the GaAs(001)-(2×4)/$c$(2×8) surface studied by scanning tunneling microscopy

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We have used the scanning tunneling microscope (STM) to study the deposition of Se onto the GaAs(001)-(2×4)/$c$(2×8) surface and the formation of the GaAs(001):Se-(2×1) surface reconstruction. The experiments were carried out in a combined STM–molecular-beam-epitaxy (MBE) system, allowing the surface to be imaged after deposition without transferring through air. Se was deposited onto well-characterized GaAs buffer layers grown *in situ* by MBE. We have imaged the surface at several stages during the deposition of up to two monolayers of Se, with depositions carried out at 300 °C and 470 °C. We have also looked at the effect of annealing the Se covered surface at over 520 °C, observing smoothing of the surface and desorption of Se from the surface. Se reacts with the GaAs surface at 470 °C, displacing the surface As and causing significant disruption to the GaAs surface. After approximately 0.25 monolayer of Se is deposited, the original (2×4) reconstruction is replaced by a largely disordered phase. With further Se deposition the Se-terminated (2×1) reconstruction is formed. The general surface morphology is similar to the original GaAs surface prior to Se deposition except that there are many small (2×1) islands on the terraces. When Se is deposited onto the GaAs surface at 300 °C a disordered Se overlayer is formed. The (2×1) reconstruction is only formed by subsequent annealing at higher temperature. The resulting surface morphology is similar to that formed by Se deposition at 470 °C. High-resolution STM images resolve the Se dimers that make up the (2×1) reconstruction; however, the individual atoms within one dimer have not been resolved. Small islands on the GaAs(001):Se-(2×1) surface can be removed by annealing at above 520 °C for up to 20 min. The resulting (2×1) surface has well-ordered flat planes several hundred angstroms across, which are comparable to those on the original GaAs buffer layer. The temperature at which the islands are removed is very close to the temperature at which Se is desorbed from the (2×1) terraces, suggesting that the removal of the islands involves desorption of Se and is not a simple diffusion process.

## I. INTRODUCTION

There has been recent interest in the deposition of Se onto the GaAs(001) surface, with several photoelectron spectroscopy (PES) studies being carried out.[1–4] The motivation for this work has been primarily directed to the use of Se as a passivating layer for the GaAs surface. Se is easily deposited *in situ* by molecular-beam epitaxy (MBE) during the growth of device structures. The Fermi level on both the clean and metal-covered GaAs(001) surface is pinned close to the middle of the band gap. This is often undesirable for device structures and contacts. It has been found that Se deposited onto the GaAs(001) surface can reduce the surface state density, and hence reduce the surface band bending. The Se-terminated GaAs(001) surface forms a very stable (2×1) reconstruction, which has been linked to the flattening of the surface bands.[5] Recent scanning tunneling microscope (STM) studies on the clean GaAs(001) surface have determined the mechanisms responsible for the Fermi-level pinning;[6–8] however the mechanism of Se passivation is not understood.

The deposition of Se onto GaAs is also important in the growth of ZnSe on GaAs. Following the demonstration of blue-green laser diodes fabricated from ZnSe-based II-VI semiconductors,[9,10] a great deal of attention has been paid to the growth of these materials. The laser diodes are grown by MBE on GaAs(001) substrates, and as a result they contain a III-V/II-VI heterovalent interface. The structure of this GaAs/ZnSe interface plays a critical role in the formation of defects in the II-VI epilayers, with different procedures for initiating growth of ZnSe on the GaAs surface having a major effect on the perfection of the ZnSe layer.[11] The structure of the GaAs/ZnSe interface can also affect the electrical transport across the interface, which is of crucial importance since this interface is one of the contacts to the laser structure. The interaction of Se on the GaAs(001) surface is one of the possible first steps in the formation of the GaAs/ZnSe interface, and so an understanding of the formation and structure of the GaAs(001):Se surface may lead to improved growth of ZnSe on GaAs.

The structure and chemical bonding of Se deposited by MBE onto the GaAs(001) surface has recently been studied by reflection high-energy electron diffraction[2] (RHEED) and PES.[2–4] It was shown that when Se is deposited onto an As-rich GaAs(001) surface, As is displaced by Se, and that Se also occupies all or some of the As sites on a few planes below the surface. A (2×1) reconstruction is formed on the surface. The details of the reaction of Se on the GaAs(001) surface are found to depend on the starting GaAs surface reconstruction and the temperature of deposition. At higher deposition temperatures (above 500 °C), other reconstructions including

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(2×3) and (4×3) have been observed.[2] These are probably Se deficient structures. It is clear that the interaction of Se with the GaAs(001) surface is complex, and that the details of the reaction mechanism and the structure of the Se-terminated surfaces are not well understood.

In this paper, we present STM images of Se deposited by MBE onto the As-terminated GaAs(001)-(2×4)/$c$(2×8) surface. We find that the formation of the (2×1) reconstruction occurs via an intermediate phase at low coverage, which is not well ordered. During the reaction there is considerable disruption of the (2×4)/$c$(2×8) surface reconstruction. The real-space imaging capability of the STM allows us to study the degree of ordering on the surface and observe the formation of islands, as a function of Se deposition and annealing temperature. It is possible to produce a highly ordered (2×1) surface with flat terraces of several hundred angstroms across. Although we are not able to make an unambiguous determination of the structure of the (2×1) surface, atomic resolution images are consistent with a top layer of Se dimers.

## II. EXPERIMENT

The experiments were carried out in a combined STM/MBE system allowing growth of GaAs buffer layers, Se deposition, and STM observation to be made without the sample leaving the UHV environment. The system consists of three connected chambers: an analytical chamber which includes low-energy electron diffraction (LEED) and the STM stage, linked via a preparation chamber to the MBE chamber, which contains elemental sources of Ga, As, Si (as a dopant), and Se. A more detailed description of the system can be found elsewhere.[12]

The Se depositions were made onto GaAs(001) epilayers grown *in situ*. The substrates were Si-doped $n^+$-type GaAs(001) wafers, that were etched in 2:5:40 solution of $H_2O_2$:$H_2NO_4$:$H_2O$ before being loaded into the UHV system. The substrate size was typically $1\times 1$ cm$^2$. After degassing at 350 °C–400 °C in the preparation chamber for 1 h, the substrate was introduced into the MBE chamber and the oxide desorbed in an $As_4$ flux at 600 °C as measured by an optical pyrometer. The GaAs epilayer was grown at 585 °C under As-rich conditions with a (2×4) surface reconstruction, and doped to $3\times 10^{18}$ cm$^{-3}$ with Si. The thickness of the epilayer was typically 0.4 $\mu$m. After completion of growth, the sample was left to recover under an As flux at growth temperature for 5 min to smooth out the surface, then rapidly withdrawn from the MBE chamber and transferred to the analytical chamber. This rapid quenching procedure preserves the (2×4)/$c$(2×8) reconstruction present during growth. All LEED and STM analysis was carried out at room temperature.

Se depositions were carried out the day following the growth of the GaAs buffer layer. This allowed the background As in the growth chamber to pump away, leaving a base pressure of $2\times 10^{-10}$ Torr. The amount of Se incident on the surface was determined from the incoming flux. The flux was measured by a nude ion gauge placed behind the sample position with the sample moved to one side. The deposition rate that was used in these experiments, assuming unity sticking coefficient, corresponded to 1 monolayer (ML) in 7 s. The coverages quoted in this paper are calculated from the deposition time, assuming unity sticking coefficient. However, this may not give an accurate coverage determination as the Se sticking coefficient may differ from unity. In addition, there are errors in the absolute value of the flux as the ion gauge sensitivity is not accurately known. However, the Se exposure for different depositions can be compared. The substrate temperature during deposition was measured with an optical pyrometer calibrated from the oxide desorption from the GaAs substrate. The resulting surface periodicities were monitored during deposition by RHEED and determined from LEED in the analytical chamber.

The STM images presented in this paper are of filled states and were collected in the constant current mode with a tunneling current of 0.1 nA, and a sample bias of between −2.5 and −3 V. On each sample studied, several STM images were taken from different regions of the surface to ensure that the surface structure and morphology were uniform. In no case was any significant variation found at different locations on the same sample. Since the sample has to be returned to the MBE chamber for further Se deposition, the STM images are representative of the particular sample, but do not show exactly the same region of surface for different Se coverages. All the

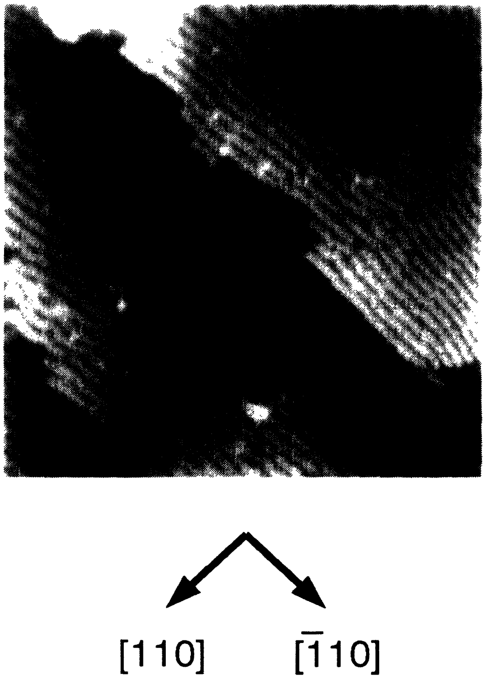

FIG. 1. An STM image of the MBE-grown GaAs(001)-(2×4)/$c$(2×8) buffer layer prior to Se deposition. The crystallographic orientation for all the STM images shown in this paper is marked. This image, together with those in Figs. 2, 3, and 5, shows an area 600 ×600 Å$^2$.

STM images shown have the same crystallographic orientation (shown in Fig. 1), and with the exception of Fig. 4, all show an area of surface approximately $600\times600$ Å$^2$.

### III. REACTION OF Se ON THE GaAs(001)-$(2\times4)/c(2\times8)$ SURFACE

In these experiments Se was deposited onto a well-characterized As-terminated GaAs(001)-$(2\times4)/c(2\times8)$ surface. An STM image of a clean GaAs surface, prior to Se deposition, is shown in Fig 1. The typical terrace size is 300–500 Å, with no small islands on the terraces. In this image there are two steps, each 2.8 Å high, running in the $[\bar{1}10]$ direction. The $(2\times4)$ reconstruction arises from the formation of As dimers along the $[\bar{1}10]$ direction, with the $4\times$ periodicity in the $[110]$ direction due to a regular array of dimer vacancies.[13] These dimer vacancies line up to form dimer-vacancy rows along the $[\bar{1}10]$ direction. These dimer-vacancy rows can be seen in Fig. 1 as dark lines in the $[\bar{1}10]$ direction spaced 16 Å apart, running parallel with the two step edges that are seen in this image. There are two different ways that these $(2\times4)$ unit cells can be arranged on the surface leading to either true $(2\times4)$ periodicity or $c(2\times8)$ periodicity. In practice, the surface has a mixture of these two periodicities.[13,14] For the purposes of this paper, we will refer to this structure simply as the $(2\times4)$ reconstruction.

The first series of Se depositions was carried out at a substrate temperature of 300 °C, which corresponds to a typical ZnSe growth temperature. This resulted in a disordered overlayer on the GaAs surface. After the deposition of 0.25 ML of Se, a random distribution of small bright features is seen by STM on the GaAs surface [Fig. 2(a)]. The underlying $(2\times4)$ reconstruction appears undisturbed. With further Se deposited onto the surface, the density of these disordered features increases, until at about 1 ML of deposited Se, the entire GaAs surface appears to be covered, as seen in Fig. 2(b). In this image, one 2.8-Å-high step from the underlying GaAs surface can be seen running in the $[\bar{1}10]$ direction. Although the LEED pattern of this surface is predominantly $(1\times1)$, there are very weak $\frac{1}{4}$-order spots, showing that the $4\times$ periodicity of the $(2\times4)$ reconstruction is preserved underneath the Se overlayer. Since the STM images do not show disruption of the $(2\times4)$ surface at low coverage, we conclude that at 300 °C, Se simply bonds to the surface As atoms forming a disordered Se overlayer. The surface Se could be individual atoms or molecules—we are unable to distinguish between these two possibilities from the STM images. Our results are consistent with the PES results of Scimeca *et al.*[3] who found that after depositing Se onto the GaAs(001) As-terminated surface at room temperature and annealing at 250 °C, Se was bonded to As and not Ga.

At 300 °C no $(2\times1)$ reconstruction is formed when 1–2 ML of Se are deposited onto the GaAs(001)-$(2\times4)$ surface. However, Takatani, Kikawa, and Nakazawa[2] find from RHEED studies that the $(2\times4)$ reconstructed GaAs surface does transform into the $(2\times1)$ reconstruction after prolonged Se exposure (up to 2 min) at 400 °C. The disordered Se-terminated surface shown in Fig. 2(b)

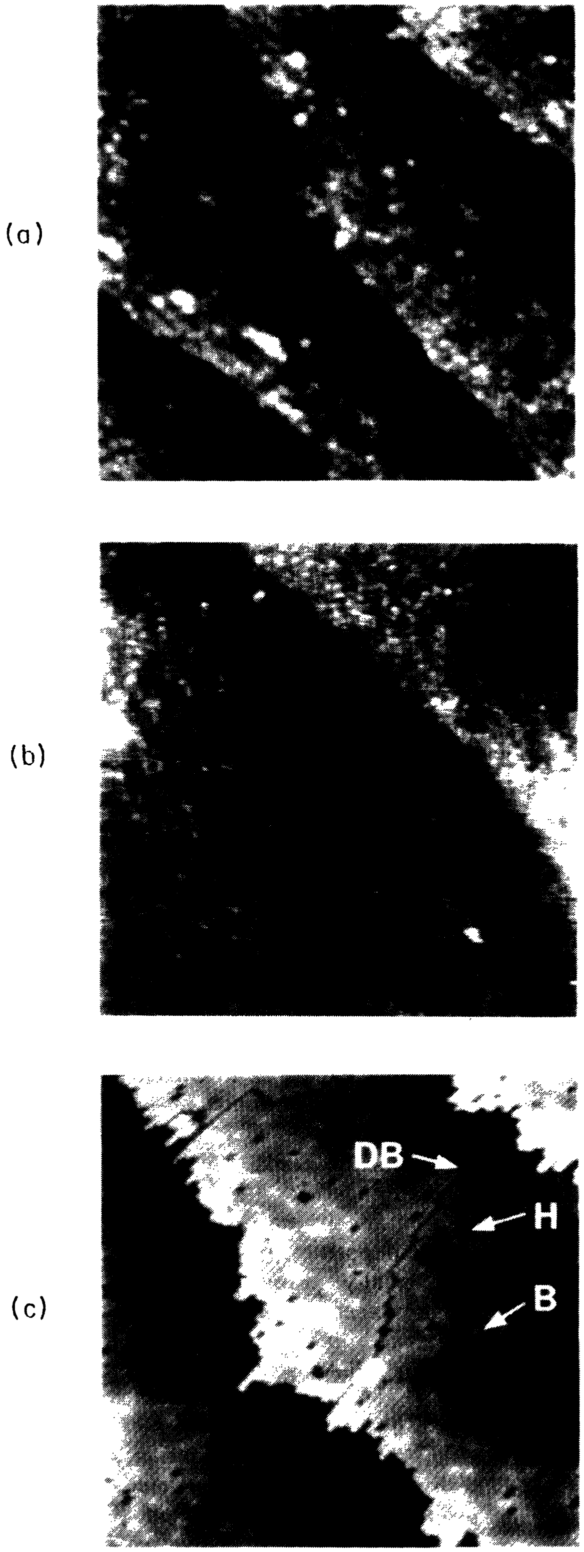

FIG. 2. STM images showing the deposition of Se at 300 °C onto the GaAs(001)-$(2\times4)/c(2\times8)$ surface for (a) 0.25 ML of Se, (b) 1 ML of Se, and (c) 1 ML of Se and annealing above 520 °C for 20 min. At this deposition temperature an ordered $(2\times1)$ surface (c) is only obtained after higher-temperature annealing. The three types of defects discussed in the text are marked in (c): domain boundaries (DB), holes ($H$), and bright spots ($B$).

can be transformed into a $(2\times 1)$ reconstructed surface by annealing at a temperature close to 500 °C. For short annealing times (1–2 min), the STM shows that the resulting $(2\times 1)$ surface is not well ordered. After prolonged annealing above 520 °C for up to 20 min, a highly ordered $(2\times 1)$ surface, shown in Fig. 2(c), can be obtained. The details of this annealing procedure are described in Sec. V. Rows are seen in the STM image [Fig. 2(c)] running in the [110] direction (perpendicular to the step edges) which are separated by 8 Å (the 2× spacing) in the [$\bar{1}$10] direction. The morphology of this GaAs:Se surface is comparable to the as-grown GaAs(001)-$(2\times 4)$ surface: atomically flat terraces a few hundreds of angstroms in width, with the steps running predominantly along the [$\bar{1}$10] direction. There are very few small islands on the flat terraces [only one can be seen in Fig. 2(c)]. The atomic structure of this $(2\times 1)$ surface will be discussed in more detail below.

In order to follow the reaction of Se with the GaAs surface, Se was deposited at a higher temperature (approximately 470 °C) so that no additional annealing step was required to form the $(2\times 1)$ reconstruction. This is close to the temperature at which the $(2\times 4)$ reconstruction on the clean GaAs surface starts to change over to a more Ga-rich structure in UHV. At this temperature, the incident Se immediately reacts with the GaAs surface, and we can follow the change from the $(2\times 4)$ As-terminated reconstruction to the $(2\times 1)$ Se-terminated reconstruction. At a low Se coverage of 0.1 ML, the Se is seen to create holes in the GaAs(001)-$(2\times 4)$ surface [Fig. 3(a)]. These holes are several $(2\times 4)$ unit cells in size and always involve complete $(2\times 4)$ unit cells. In addition, some small areas of modified structure appear on top of the $(2\times 4)$ unit cells. There is still a large part of the $(2\times 4)$ reconstruction that is unaffected. To ensure that these holes resulted from the Se deposition and not as a result of heating the GaAs, a clean GaAs(001)-$(2\times 4)$ surface was heated for several minutes at 470 °C. STM images showed that this had no effect on the GaAs surface. As the Se deposition proceeds, the number and size of these holes increase together with the modified regions on top of the $(2\times 4)$ unit cells, until the surface in Fig. 3(b) is reached with the deposition of 0.25 ML of Se. At this stage, there is virtually no remnant of the $(2\times 4)$ reconstruction. It can be seen that each terrace on the original GaAs surface has been broken up into two levels. A step on the underlying GaAs surface ran along the [$\bar{1}$10] direction from the top-left corner to the bottom-right corner of Fig. 3(b). The structure on each of these two levels is the same, and has a predominance of 2× spacings along both axes but is not well ordered. We have not yet been able to make a determination of the detailed atomic structure on this surface. Further Se deposition leads to the formation of the $(2\times 1)$ reconstruction. Figure 3(c) shows the surface after depositing 0.6 ML of Se. The $(2\times 1)$ structure is clearly visible, but the rows along the [110] direction are rather short, with many holes in the $(2\times 1)$ surface, and many small $(2\times 1)$ islands on the main terrace. Further Se deposition largely removes the holes in the main $(2\times 1)$ terrace, but many small islands remain. These islands can only be removed by appropri-

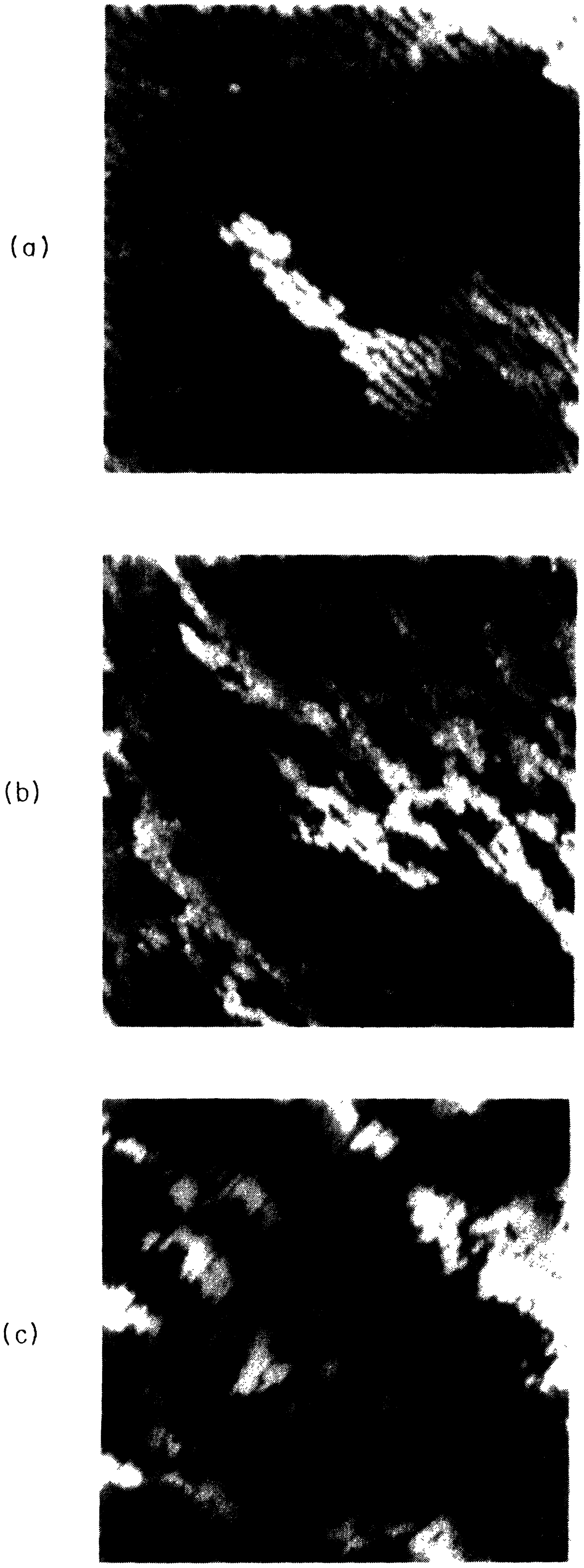

FIG. 3. STM images showing the deposition of Se at 470 °C onto the GaAs(001)-$(2\times 4)/c(2\times 8)$ surface for (a) 0.1 ML, (b) 0.25 ML, and (c) 0.6 ML. At this deposition temperature, Se immediately reacts with the GaAs surface. The $(2\times 1)$ reconstruction (c) is formed via a largely disordered intermediate phase (b).

ate annealing above 520 °C, when it is possible to obtain a well-ordered surface comparable to that shown in Fig. 2(c). The annealing of the (2×1) structure will be discussed below.

The images shown in Fig. 3 demonstrate that the formation of the GaAs(001):Se-(2×1) surface is a complex process involving considerable disruption to the original GaAs(001)-(2×4) surface. The reaction of Se with the GaAs surface does not occur until the temperature is close to the temperature at which As starts to desorb from the clean GaAs surface. This indicates that the Se reaction involves the displacement of As. The surface disruption also indicates that Se does not simply produce an ordered overlayer on top of the GaAs surface, but that more than one layer of the GaAs may be modified by the Se. These conclusions are supported by the results of PES experiments,[2–4] where it is found that on the GaAs:Se(001)-(2×1) surface, there is no bonding between As and Se, and that Se exists in two states. PES results also show that Se may replace As up to three layers below the surface. We note here that quantitative determination by PES of the number of layers that the Se occupies may be complicated by the relatively rough (2×1) surface that is obtained without annealing.

## IV. THE GaAs(001):Se-(2×1) SURFACE

The structure of the GaAs(001):Se-(2×1) surface has not yet been determined, although it is generally considered to involve a surface layer of Se dimers as well as Se substituting for As on planes below the surface.[2,5] The STM cannot directly give structural information on subsurface layers, although as discussed above, the complexity of the reaction of Se with the GaAs surface points to a modification of more than just the surface layer. The STM image of the (2×1) surface [Fig. 2(c)] clearly shows the 2× periodicity as rows along the [110] direction which are 8 Å apart, but shows no detail along the rows. Figure 4(a) shows a higher resolution image of a region of (2×1) reconstruction, which shows the individual (2×1) unit cells; both the 2× and 1× periodicities are clearly visible. The single bright feature associated with each unit cell is most likely to be a Se dimer. This cannot be confirmed as the individual atoms of the dimer have not been resolved, however this STM image is very similar to the STM images that have been obtained of Si dimers on the Si(100)-(2×1) surface.[15] Further consideration of the detailed structure of the GaAs(001):Se-(2×1) surface will be published elsewhere.[16]

The STM clearly shows that with the appropriate annealing conditions the GaAs:Se-(2×1) surface is well ordered [Fig. 2(c)]. The terrace size is determined by the terrace size of the GaAs surface that the Se was deposited onto. Apart from the steps at the terrace edges, there are three different types of defect that can be seen: dark lines running parallel with the dimer rows, small "holes" in the (2×1) surface, and bright spots on the (2×1) terraces. The origin of these bright spots is unknown, but their apparent height is small making it unlikely that they are atoms on top of the Se dimers. It is more likely that they are an electronic effect due to some form of defect underneath the top Se layer. They occur at quite a high density—approximately $10^{12}$ cm$^{-2}$ in Fig. 2(c). The small "holes" in the (2×1) surface are clusters of a few missing Se dimers.

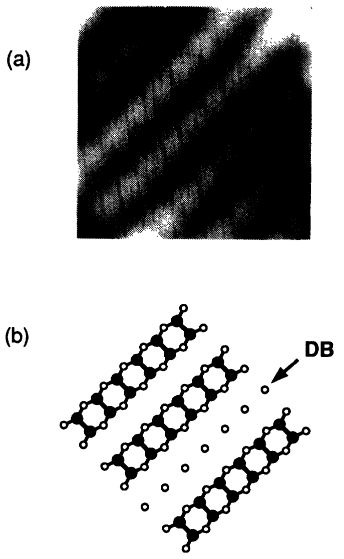

FIG. 4. (a) STM image of a 35×35 Å$^2$ area of the GaAs(001):Se-(2×1) surface. The individual dimers that make up the (2×1) unit cell (8×4 Å$^2$) are easily resolved. (b) A schematic of the surface layer structure of the GaAs(001):Se-(2×1) surface showing a domain boundary (DB) as seen in Fig. 2(c). The surface Se atoms are shown as filled circles and the first-layer Ga atoms as empty circles.

In the large area image of Fig. 2(c) there are three broader dark lines on the terraces (the two on the middle terrace are running approximately in the [110] direction). Measurements made from the STM images show that these dark lines are a result of two adjacent dimer rows being 12 Å apart rather than the normal 8 Å. At the segments of these dark lines that run in the [$\bar{1}$10] direction it can easily be seen that they are boundaries between two different domains of the (2×1) reconstruction. The phase of the dimerization of the surface Se atoms in the [$\bar{1}$10] direction changes across the boundary. This is illustrated schematically in Fig. 4(b). The dark line of the domain boundary corresponds to a row of missing Se atoms. Typical domain widths in the [$\bar{1}$10] direction range from 100 Å to several hundred angstroms, and the domains are typically larger in the [110] direction. These domain boundaries might be expected on a (2×1) dimer surface. It is interesting to note that similar domain boundaries are not generally observed on the Si(100)-(2×1) surface, which may be due to the higher temperatures usually used to clean the Si(100) or reflect a stronger Se dimer bond, which does not allow the dimers to reorder and eliminate domain boundaries.

## V. SURFACE MORPHOLOGY RESULTING FROM ANNEALING

We have already shown that appropriate annealing of the disordered Se overlayer produced by deposition at

300 °C results in a well-ordered (2×1) reconstructed surface [Fig. 2(c)]. A similar result is obtained by appropriate annealing of the (2×1) surface produced directly by Se deposition at 470 °C. There is no observable difference in the annealing behavior for Se deposited at 300 °C and 470 °C. In both cases the surface morphology is dependent on the precise annealing conditions. The removal of small islands on the terraces occurs at a temperature close to that at which Se starts to desorb from the terrace. Although we have not carried out a complete study of the effect of annealing on the surface morphology, our preliminary study does show some clear trends.

After depositing approximately 1 ML of Se at 470 °C, the (2×1) surface has a large number of small islands on the terraces, and the step edges are relatively ragged. This is very similar to the surface morphology obtained after Se deposition at 300 °C followed by brief annealing at about 500 °C. Annealing at no more than 520 °C for 20 min has very little effect on the surface morphology. It may improve the ordering along the step edge, but has little effect on the island size and density on the terraces. The surface after this annealing step is shown in Fig. 5(a). Further annealing at 540 °C for 20 min eliminates most of these small islands as shown in Fig. 5(b). However, in this case, holes have also been created in the (2×1) terraces. These holes are similar in size to the islands that were previously present on the surface and are evenly distributed on the terraces. The step edges are also more ragged as a result of the higher-temperature anneal. It is apparent that Se has been lost from the surface. The Se has either gone down below the surface or has evaporated. PES data show that heating to 550 °C and above does remove Se from the surface, and eventually the Se can be completely desorbed.[2,3] It is therefore reasonable to assume that we are seeing the result of Se desorption from the GaAs:Se-(2×1) surface. The question then arises: are the islands seen in Fig. 5(a) removed by diffusion or by desorption? Since the islands are 2.8 Å high (the GaAs bilayer height) and so consist of a Se and Ga layer, desorption of the Se would leave Ga on the surface. The Ga would then have to diffuse across the surface, possibly attaching at step edges. Thus, the possible mechanisms for island removal are either diffusion of both Se and Ga to the step edges or desorption of Se followed by diffusion of Ga to the step edges. We are not able to differentiate between these mechanisms, but since the islands are only removed at temperatures close to the onset of Se desorption it is likely that desorption of Se from the islands occurs. However, we see no indication of excess Ga on a well-ordered (2×1) surface. When the sample is heated so that Se desorbs from the (2×1) terraces, the Ga probably remains in the holes that are created.

After overheating and desorbing some Se, the well-ordered (2×1) surface can be restored by further deposition of a small amount of Se onto the surface followed by annealing. Figure 5(c) shows the result of depositing 0.5 ML of Se at 470 °C onto the surface shown in Fig. 5(b) and annealing at about 520 °C for 20 min. The resulting (2×1) surface is comparable to that shown in Fig. 2(c). There are no small islands on the terraces. It is possible to repeat the cycle of heating to desorb some Se followed

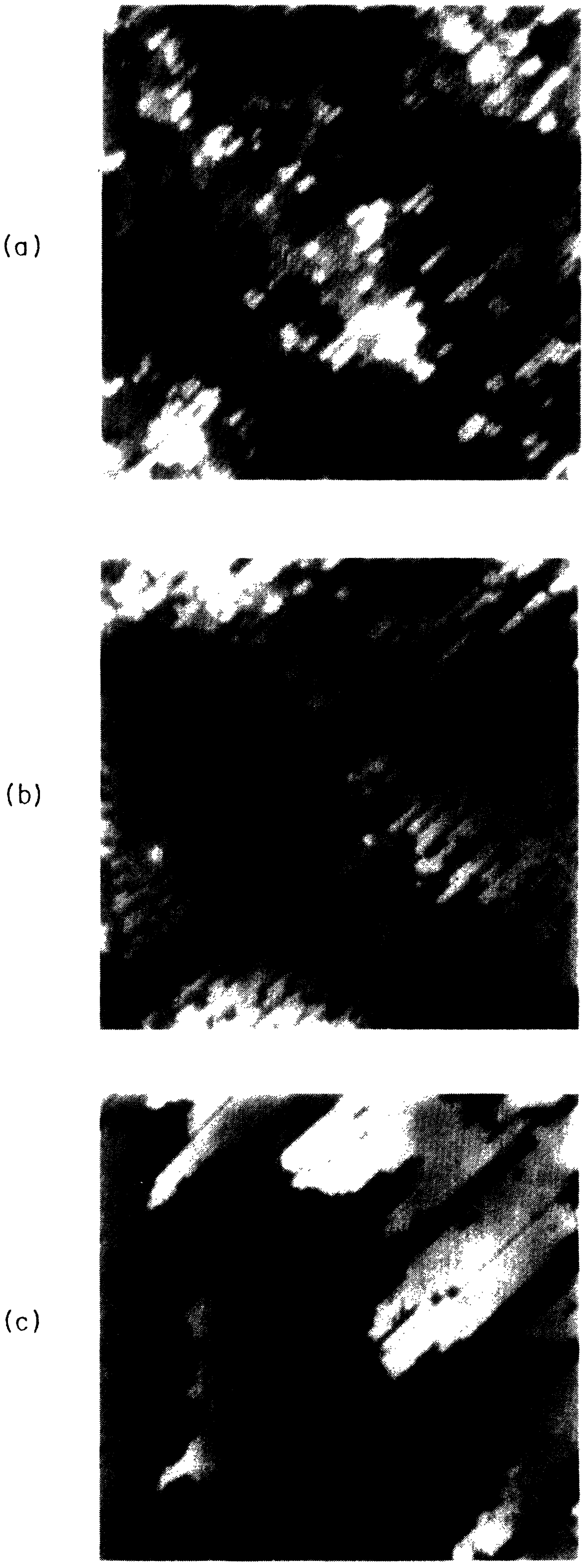

FIG. 5. STM images showing the effect of high-temperature annealing on the GaAs(001):Se-(2×1) surface. (a) After deposition of 1.5 ML of Se at 470 °C and annealing at approximately 520 °C for 20 min, (b) with further annealing at 540 °C for 20 min, and (c) with further 0.5 ML Se deposition at 470 °C and annealing at about 520 °C for 20 min.

by Se deposition and annealing, and maintain a well-ordered (2×1) surface. It is clear from the STM images shown in Fig. 5 that in order to produce a smooth, well-ordered (2×1) surface, it is necessary to control the annealing procedure quite carefully. Annealing at 520 °C or below does not remove small islands from the terraces. Annealing above 520 °C does remove these islands, but can also result in surface roughening from Se desorption. It is usually necessary to deposit additional Se and further anneal at about 520 °C in order to obtain highly ordered surfaces as shown in Figs. 2(c) and 5(c). The differences in the LEED and RHEED patterns for the different surfaces shown in Fig. 5 are very subtle (mostly a small change in the background intensity). The STM provides the most sensitive determination of the surface morphology.

In both Figs. 5(a) and 5(c), islands of the (2×1) structure can be seen on the terraces. In the first case, these are small islands which are only at most a few 2× spacings wide, and in the latter case, these are much larger islands. In both cases, they are mostly longer on the 1× ([110]) direction than they are in the 2× ([$\bar{1}$10]) direction. This anisotropy of island shape is a general feature of the GaAs:Se-(2×1) surface. We have observed islands that are only one dimer (8 Å) wide but extend up to 100 Å along the [110] direction.

## VI. CONCLUSIONS

Our STM results show that the formation of the (2×1) reconstruction on the Se covered GaAs(001)-(2×4) surface occurs via a complex reaction which involves the displacement of As from the GaAs surface. When Se is deposited onto the GaAs surface at 300 °C no reaction takes place, and a disordered Se overlayer is formed. The reaction leading to the (2×1) reconstruction occurs at temperatures close to the temperature at which As starts to desorb from the clean GaAs(001)-(2×4) surface in UHV. By depositing Se onto the GaAs surface at 470 °C, it is possible to follow the reaction. The STM images show that Se displaces As from complete (2×4) unit cells, resulting in small holes appearing in the (2×4) surface. These holes grow, and in addition, some regions on top of the As dimers of the (2×4) reconstruction are modified. After the deposition of approximately 0.25 ML of Se, the (2×4) reconstruction has almost completely disappeared, with each terrace being broken up into two levels. The details of this structure have not yet been determined. Further Se deposition leads to the formation of the Se-terminated (2×1) reconstruction.

The GaAs(001):Se-(2×1) surface formed either by depositing Se at 300 °C and brief annealing at up to 520 °C or by depositing Se at 470 °C has a large number of small islands on the main terraces. In order to remove these islands it is necessary to anneal the surface above 520° for up to 20 min. It is then possible to produce a very well-ordered (2×1) structure with flat terraces the same size as were present on the original GaAs surface. However, this annealing temperature is close to the temperature at which Se desorbs from the (2×1) surface, and so it is very easy to roughen the (2×1) terraces by desorbing Se at the same time as removing small islands. It is quite likely that small islands are actually removed by a mechanism that involves desorption of Se. If Se is desorbed from the (2×1) terraces, it can be replaced by further Se deposition and a well-ordered surface obtained by annealing.

The detailed structure of the GaAs(001)Se-(2×1) surface has not yet been fully determined. The STM data supports previously suggested models[2,5] involving a top layer of Se dimers. Our STM observations of the reaction of Se on the GaAs surface together with published PES data[2,3] show that Se replaces As on planes below the surface. The number of planes involved and the extent of this placement on each plane has yet to be determined.

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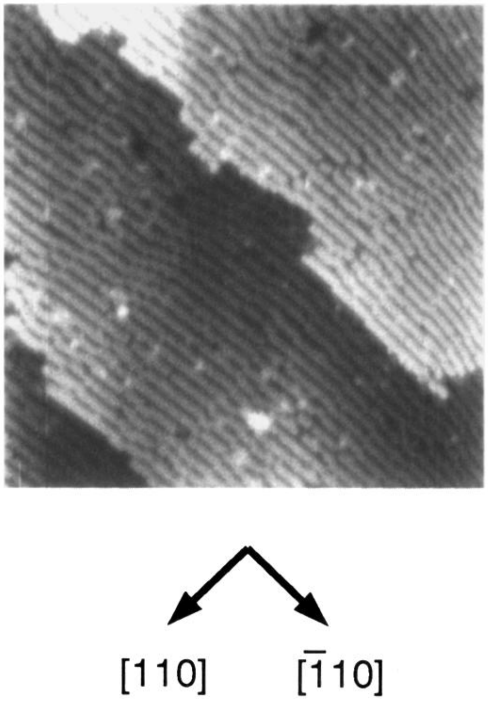

FIG. 1. An STM image of the MBE-grown GaAs(001)-$(2\times4)/c(2\times8)$ buffer layer prior to Se deposition. The crystallographic orientation for all the STM images shown in this paper is marked. This image, together with those in Figs. 2, 3, and 5, shows an area $600\times600$ Å$^2$.

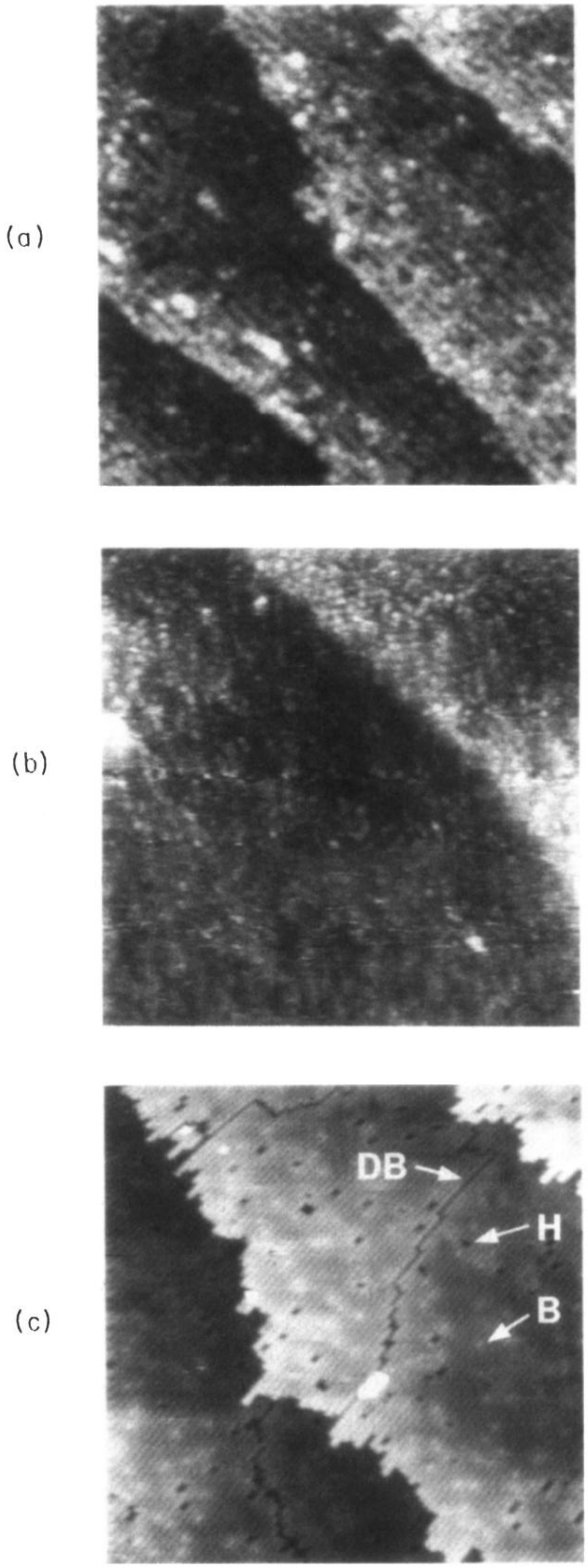

FIG. 2. STM images showing the deposition of Se at 300 °C onto the GaAs(001)-$(2\times4)/c(2\times8)$ surface for (a) 0.25 ML of Se, (b) 1 ML of Se, and (c) 1 ML of Se and annealing above 520 °C for 20 min. At this deposition temperature an ordered $(2\times1)$ surface (c) is only obtained after higher-temperature annealing. The three types of defects discussed in the text are marked in (c): domain boundaries (DB), holes ($H$), and bright spots ($B$).

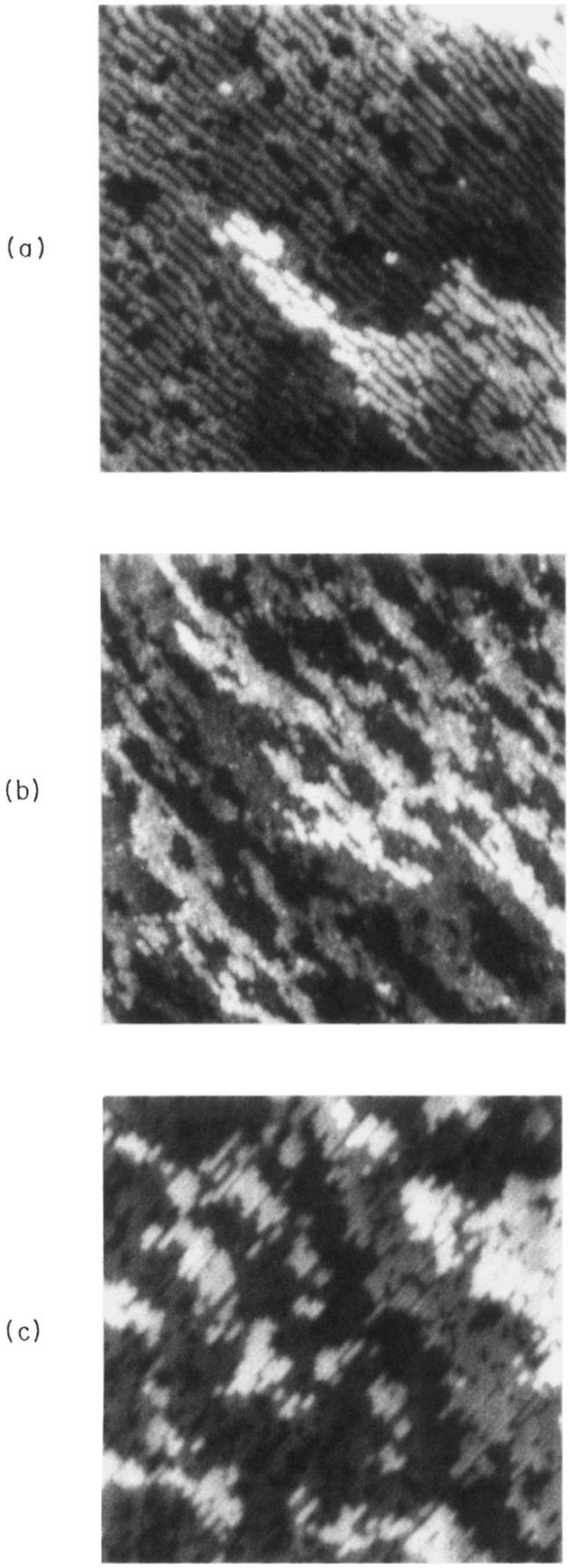

FIG. 3. STM images showing the deposition of Se at 470 °C onto the GaAs(001)-$(2\times4)/c(2\times8)$ surface for (a) 0.1 ML, (b) 0.25 ML, and (c) 0.6 ML. At this deposition temperature, Se immediately reacts with the GaAs surface. The $(2\times1)$ reconstruction (c) is formed via a largely disordered intermediate phase (b).

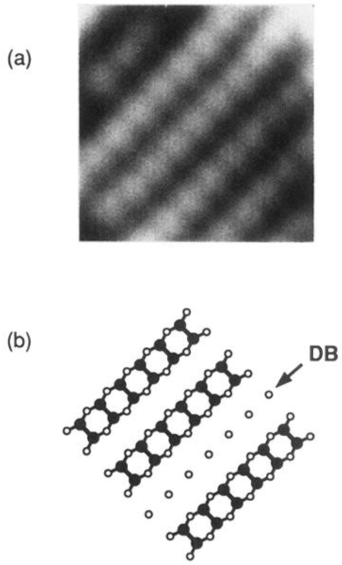

FIG. 4. (a) STM image of a $35\times35$ Å$^2$ area of the GaAs(001):Se-$(2\times1)$ surface. The individual dimers that make up the $(2\times1)$ unit cell ($8\times4$ Å$^2$) are easily resolved. (b) A schematic of the surface layer structure of the GaAs(001):Se-$(2\times1)$ surface showing a domain boundary (DB) as seen in Fig. 2(c). The surface Se atoms are shown as filled circles and the first-layer Ga atoms as empty circles.

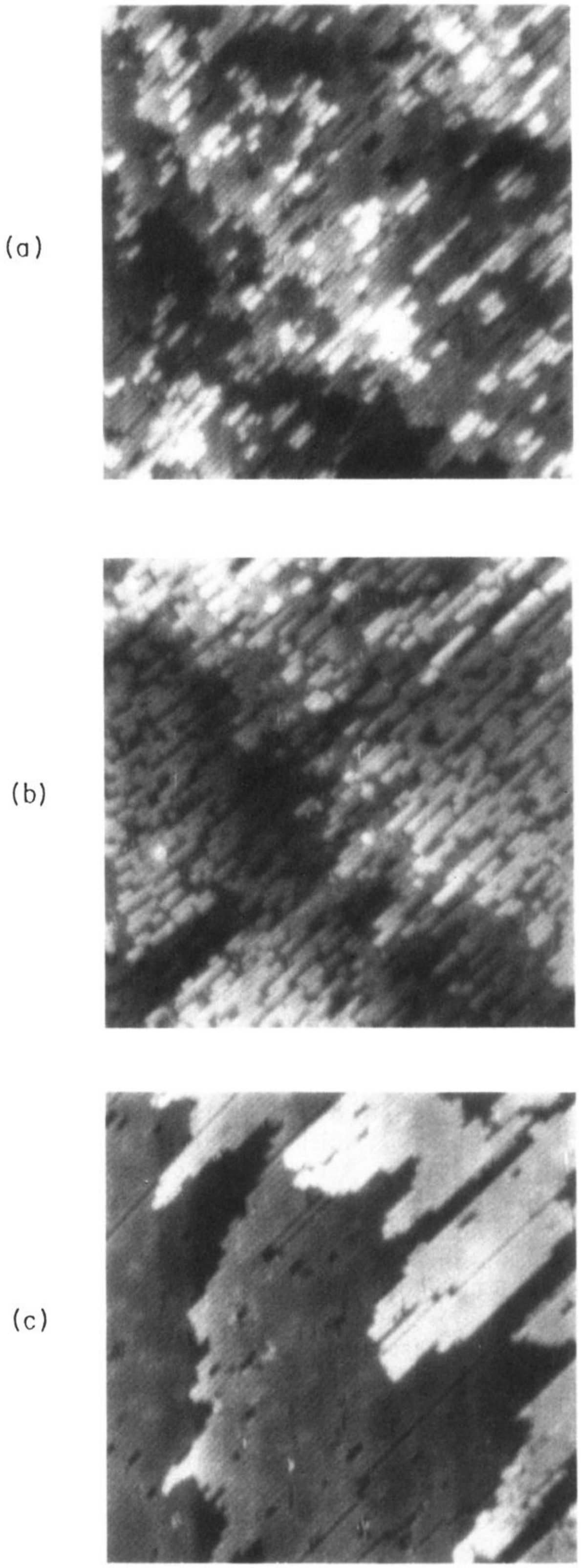

FIG. 5. STM images showing the effect of high-temperature annealing on the GaAs(001):Se-(2×1) surface. (a) After deposition of 1.5 ML of Se at 470 °C and annealing at approximately 520 °C for 20 min, (b) with further annealing at 540 °C for 20 min, and (c) with further 0.5 ML Se deposition at 470 °C and annealing at about 520 °C for 20 min.