

Surface structure of As-stabilized GaAs(001): 2×4 , $c(2\times 8)$, and domain structures

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Structural models characterized by missing rows of surface As dimers are proposed for the 2×4 and $c(2\times 8)$ As-stabilized surfaces of GaAs. Experimental and theoretical support for these models and for a variable surface stoichiometry are examined. A model with full monolayer As coverage and either symmetric or asymmetric dimers is found to be inadequate. Total-energy calculations show that 2×4 or $c(2\times 8)$ unit cells are optimal for surface As coverages of $\Theta_{\text{As}}=0.5$ and $\Theta_{\text{As}}=0.75$. The proposed model can account in a simple way for the observed one-dimensional disorder and provides a good description for the amplitudes of fractional-order structure factors.

I. INTRODUCTION

The surface atomic structure of the GaAs(001)- 2×4 [or $c(2\times 8)$] reconstruction is still controversial. It is generally agreed that it is As-stabilized but it has not been possible to relate it to any specific surface composition (As-to-Ga ratio or fractional As coverage, Θ_{As}). There is considerable evidence that it exists over a compositional range.¹ With low-energy electron diffraction (LEED) a centered $c(2\times 8)$ structure has been found,² but this has never been observed with reflection high-energy electron diffraction (RHEED). The diffraction patterns obtained by this latter technique are usually referred to as the 2×4 structure.³ The differences between LEED and RHEED observations may be related to the occurrence of one-dimensional disorder⁴ and this may be an inherent property of this surface, as will be discussed further on in this paper.

A previous calculation of the surface energy band structure for a 2×1 reconstruction model based on tilted As-As dimers was found to be in good, although not complete, agreement with a surface band structure determined by angle-resolved photoemission measurements.⁵ On this basis an extension of the 2×1 to a 2×4 structure was proposed, assuming an As-terminated surface ($\Theta_{\text{As}}=1$). It was realized, however, that this model did not adequately explain the RHEED diffraction features and two possible modifications were suggested:⁶ (i) an ordered vacancy model and (ii) a model with twisted dimers maintaining the complete As termination ($\Theta_{\text{As}}=1$). Very recently one of us (D.J.C.) examined different vacancy models with symmetric As-As dimers by tight-binding total-energy calculations.⁷ Two models having a 2×4 symmetry were found to have low surface energies. The models are characterized by a missing row of As dimers per unit cell, corresponding to a surface As coverage of $\Theta_{\text{As}}=0.75$.

In this paper we analyze and discuss the proposed surface structural models. First we show in Sec. II that ex-

perimental results from both RHEED and LEED data demonstrate the persistence of disorder for this reconstruction. This is followed by a discussion of the surface stoichiometry and photoemission data. On the basis of a number of experimental criteria the various surface models are then analyzed. A 2×4 structure model with a missing row of As-As dimers seems to provide the best basic model which can be expanded to allow for both a variable surface stoichiometry and for the presence of one-dimensional disorder. These conclusions are supported by total-energy calculations.

II. EXPERIMENTAL DATA

A. Electron diffraction

GaAs(001) surfaces are conveniently prepared by molecular-beam epitaxy (MBE). The ability to monitor growth by RHEED has led to the situation where most diffraction studies on MBE-grown surfaces are carried out by RHEED rather than LEED. Both techniques are able to provide information on the surface structure, with LEED being more established in quantitative studies. An important difference between the two is the instrumental coherence length. For a standard LEED instrument this is of the order of 100 Å diameter. For RHEED the coherence length in the direction of the incident beam is of the order of thousands of Å's due to the glancing angle of incidence while it is similar to LEED values in the direction perpendicular to the beam. In comparisons between LEED and RHEED results, this aspect must be taken into account. In the following we first discuss the RHEED results.

During growth under As-stable conditions and for substrate temperatures below ~ 875 K the so-called 2×4 structure is observed.^{3,8} The diffraction pattern is then found to be streaky, but when the growth is interrupted by closing the Ga flux a recovery towards a less streaky pattern and even one with sharp diffraction spots is

found. This is particularly observable for the specularly diffracted intensity on the 00 rod for which the temperature-dependent time dependence has been studied by Neave *et al.*⁹ The growth-induced intensity oscillations observed during RHEED are closely related to these phenomena.^{9,10}

In Fig. 1 we show the diffraction patterns in the $[\bar{1}10]$, $[010]$, and $[110]$ azimuths of the As-stabilized GaAs(001) surface, taken at a substrate temperature (T_s) of 550°C after growth termination (i.e., with an incident As_2 flux of $\sim 10^{14} \text{ mol cm}^{-2} \text{ s}^{-1}$, but no Ga flux) and after full recovery (this means waiting ≥ 20 min). The substrate was accurately oriented, having a misorientation of less than 0.05° from the $[001]$ direction. It should be noted first that the patterns seen in Fig. 1(a) show high-intensity spots (or elongated spots) lying on a semicircle, the zeroth-order Laue circle. This is what would be expected for a well-ordered surface (the intersection of the reciprocal lattice rods of the surface lattice mesh and the Ewald sphere give points on a circle, i.e., the Laue circle). The periodicity corresponds to a quadrupling of the surface unit cell in the $[110]$ direction (i.e., in the direction perpendicular to the azimuthal direction of the incident beam). One also observes the presence of streaks having the same periodicity but with much lower intensity. Turning to the $[010]$ azimuth [Fig. 1(b)] and the $[110]$ azimuth [Fig. 1(c)] spots are observed for integral-order beams but not for half-order beams and here the intensity of streaks is relatively higher. The spots are again found to lie close to semicircles and an analysis of the data shows diffraction spots corresponding to the zeroth-, $\frac{1}{4}$ -, $\frac{2}{4}$ -, and $\frac{3}{4}$ -order Laue circle for the $[010]$ azimuth and up to the $\frac{2}{4}$ -order Laue circle for the $[110]$ azimuth. For Fig. 1(a) only spots on the zeroth-order Laue circle are observed and the lack of a diffraction pattern on the half-order Laue circle is in agreement with the lack of half-order spots in Figs. 1(b) and 1(c).

The reciprocal surface lattice derived from the observed Bragg diffraction spots is shown in Fig. 2. All the lattice points lie on a 1×4 lattice. We further indicate the streaks, most prominently seen in the half-order beams [Fig. 1(b)] by dashed lines. The presence of these streaks was discussed by Joyce *et al.* and ascribed to the presence of one-dimensional disorder in a 2×4 lattice.⁴ The absence of Bragg diffraction features in the half-order beams of the $[110]$ azimuth has also been noticed before.⁶ The present results provide strong confirmation of the earlier interpretation, except that it is not possible to conclude whether the basic unit cell is a 2×4 or a $c(2 \times 8)$ structure. On the basis of the tilted dimer model proposed in Ref. 5, it was suggested that the one-dimensional disorder was related to a randomness in the sequencing of dimer chains.⁴ However, the idea that the surface reconstruction is caused by an ordering of dimers is questionable. An important observation pointing in this direction is the following: When a 2×4 surface is exposed to a H_2 plasma, the streaks related to the fourfold periodicity in the $[\bar{1}10]$ azimuth persist for a much larger exposure than the half-order streaks in the $[110]$ azimuth.¹¹ This indicates that the fourfold periodicity is independent of the twofold periodicity, i.e., it cannot be

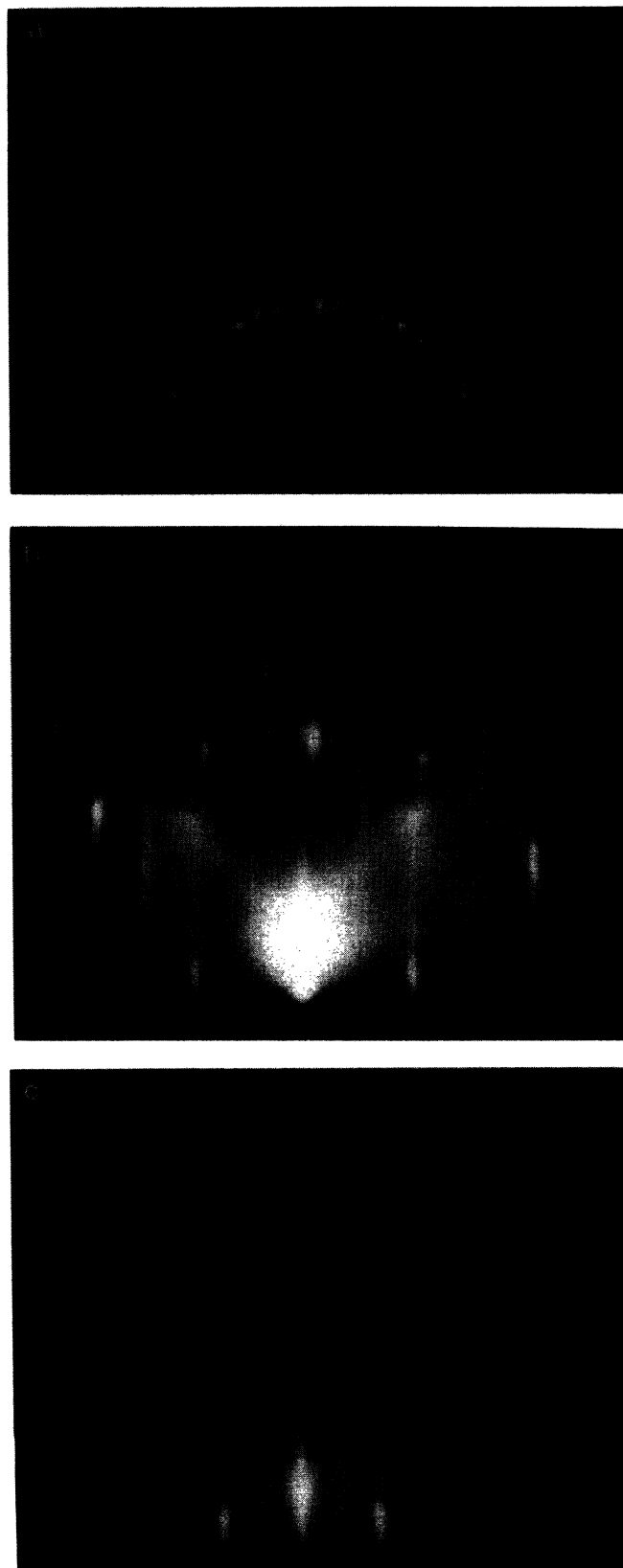


FIG. 1. RHEED patterns from a GaAs(001)- 2×4 reconstructed surface, 12.5 keV electron beam energy, $T_s = 550^\circ\text{C}$. (a) $[\bar{1}10]$ azimuth, $\Theta_i = 3.6^\circ$. (b) $[010]$ azimuth, $\Theta_i = 1.8^\circ$. (c) $[110]$ azimuth, $\Theta_i = 1.8^\circ$.

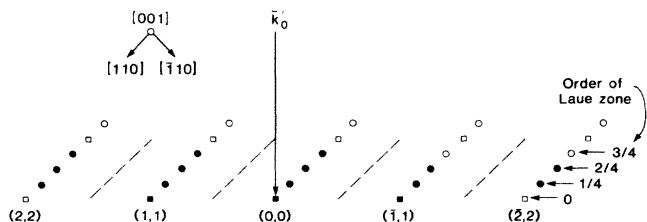


FIG. 2. Reciprocal surface lattice derived from the Bragg diffraction spots observed in Fig. 1. The squares and circles indicate integer- and fractional-order spots, respectively. Solid signs mark observed spots. $\vec{k}_0$ is the projection of the incident electron wave vector.

due to an ordering of dimer chains. The hydrogen exposure is believed to break the dimer bonds by chemisorption of hydrogen atoms to the dangling bonds of the surface atoms.¹¹⁻¹³

Few LEED results have been reported for GaAs(001). Drahten *et al.* prepared the As-stable surface by a combination of ion bombardment and annealing in an incident As flux and observed a pattern which provides clear evidence for a $c(2 \times 8)$ structure.² The only other pattern published for this surface (to our knowledge) was obtained by van Bommel *et al.*¹⁴ who heated GaAs(001) to 400°C in an AsH₃ ambient of pressure 10^{-5} torr. This pattern is shown in Fig. 3. It is seen that all diffracted spots lie on a 1×4 lattice and that there are no spots in $\frac{1}{2}$ -order positions, but instead a streak indicating one-dimensional disorder. This indicates that along the [110] direction the dimension of single domains can be considerably smaller than the coherence length of the LEED beam (~ 100 Å).

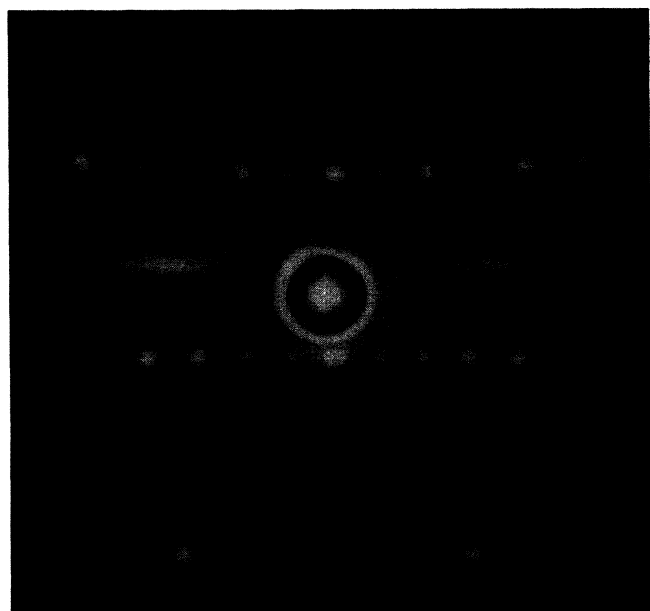


FIG. 3. LEED pattern of the As-stable GaAs(001) surface (from Ref. 14, courtesy of J. E. Crombeen).

B. Surface stoichiometry

There have been a number of attempts to determine the surface stoichiometry of GaAs(001) surfaces from measurements of As-to-Ga intensity ratios (R) using low-energy Auger transitions^{1,2,14,15} [e.g., As(31 eV) $M_{4,5}VV$ and Ga(55 eV) $M_{2,3}M_{4,5}M_{4,5}$] or core-level (As 3d and Ga 3d) photoemission peak areas.¹⁶ The surface stoichiometry (Θ_{As}) is derived by an electron escape model assuming a value for the escape depth λ and normalizing the relative intensities from As and Ga atoms by measuring a surface of known stoichiometry [i.e., the (110) cleavage surface] under otherwise identical experimental conditions.

The results show that the 2×4 and/or $c(2 \times 8)$ surface is arsenic rich but it is not possible to relate it to a specific stoichiometry. Massies *et al.*¹ report Auger ratios R varying from 2.3 to 3.7, and the presence of a $c(4 \times 4)$ phase for ratios higher than 3.2. For the same Auger transitions other authors give ratios of 2.25,² 3.0,¹⁴ and 2.12.¹⁵ These ratios may be subject to different experimental conditions, e.g., a different energy dependence of analyzer transmission function. The ratio for the (110) cleavage surface has been measured to be 2.10 ± 0.1 .² If this ratio is used as a reference then for $2.12 \leq R \leq 3.2$ and a value for λ in the range 4.5–6.0 Å, one finds $\Theta_{As} \approx 0.5$ –1.0. The photoemission measurements by Bachrach *et al.*¹⁶ give a value of $\Theta_{As} = 0.89$ although an interpretation of their measurements with an energy-dependent escape depth was found to lead to a value $\Theta_{As} = 0.99$.¹⁷

An alternative way of determining the stoichiometry is to deconvolute the line shape of a core-level photoemission peak into a bulk and a surface contribution. The latter part is then associated with atoms in the outermost layer. Both Ludeke *et al.*¹⁸ and van der Veen *et al.*¹⁹ find for the As 3d level, using similar photon energies (80 and 72.7 eV, respectively), a surface-to-total signal contribution ratio of 0.36. Using a value $\lambda = 3.9$ Å Ludeke *et al.*¹⁸ estimate $\Theta_{As} \approx 0.62$. In this method the calculated coverage depends strongly on the value of λ . Using the value of $\lambda(E) \approx 5.5$ Å from Ref. 17 we find $\Theta_{As} \approx 0.87$. The uncertainties of this method are further accentuated by the different published results of deconvolution of both the As 3d and Ga 3d core levels.¹⁸⁻²⁰ This may partly be a result of a real variation in the surface stoichiometry.

C. Angle-resolved photoemission

The surface electronic structure of GaAs(001)- 2×4 has been measured with angle-resolved photoemission.⁵ It is found to be nonmetallic and this puts severe restrictions on possible surface atomic structures as discussed in Sec. III. Two states S_1 and S_2 having initial energies ($E_{VBM} = 0$) $E_i \approx -0.5$ eV and $E_i \approx -0.6$ to -0.9 eV, respectively, are found to be mainly sp_z -like, i.e., they are dangling-bond related. A state S_3 is found to be intense for surface parallel wave vectors ($\vec{k}_{||}$) close to the $J_{2 \times 1}$ symmetry point (see Ref. 5) having energies in the range -1.3 to -1.6 eV. A fourth surface state S_4 is found to be nearly dispersionless and is observed for wave vectors

near the $J_{2 \times 1} - K_{2 \times 1}$ symmetry points at $E_i \simeq -3.15$ eV.

In Fig. 4(a) we show the energy dispersion of surface state S_2 . In Fig. 4(b) the surface Brillouin zones (SBZ's) of the 2×4 and $c(2 \times 8)$ structures, respectively, are shown together with the 1×1 SBZ. The $[\bar{1}10]$ direction in which the measurements were made corresponds to the $\Gamma J'_{1 \times 1}$ direction and obviously the periodicity of the dispersion $E = E_i(\bar{k}_{\parallel})$ is the double of what would be expected for an unreconstructed 1×1 SBZ. A comparison with Fig. 4(b) shows that the observed dispersion is in direct agreement with the 2×4 unit cell. For the $c(2 \times 8)$ SBZ there is not a doubling of the periodicity along the $\Gamma J_{1 \times 1}$ direction compared to the 1×1 SBZ, since the $J_{1 \times 1}$ point in this case is equivalent to the $J_{c(2 \times 8)}$ point and not the Γ point. If the energy in the $J_{c(2 \times 8)}$ point is close to the Γ point, however, this would be consistent with an energy of the $J'_{1 \times 1}$ point close to that of Γ and the $c(2 \times 8)$ unit cell would also be consistent with the observed periodicity. From Fig. 10 of Ref. 5 one finds that this is indeed the case, i.e., the observed dispersion is in agreement with both a 2×4 and a $c(2 \times 8)$ unit cell.

It is interesting to note that the fourfold symmetry along the $[110]$ direction which is observed in electron diffraction measurements is not found in any energy dispersion of surface states. This fourfold symmetry has been demonstrated in photoemission in one case; however, by monitoring the photoemission intensity at a constant initial energy $E_i = -0.8$ eV as a function of $\bar{k}_{\parallel}$ in the azimuth.²¹

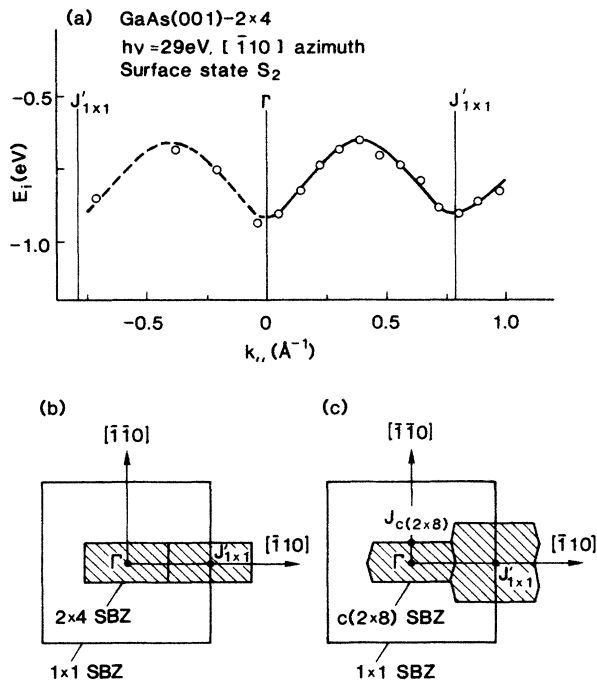


FIG. 4. (a) Energy dispersion of surface state S_2 of GaAs(001)- 2×4 . (b) Surface Brillouin zones of the 2×4 and 1×1 unit cells. (c) Surface Brillouin zones of the $c(2 \times 8)$ and 1×1 unit cells.

III. SURFACE STRUCTURAL MODELS

The discussion in the previous section has presented a number of criteria which should be met by a surface model. The results of measurements of the surface stoichiometry ($0.5 < \Theta_{As} < 1.0$) show that a vacancy model is the most realistic choice. The existence of this large compositional range shows furthermore that a set of models which can be naturally derived from one another by changing the surface composition is needed.

In the following we first discuss the models which so far have been proposed for the 2×4 structure. In Sec. II A we have seen that a 1×4 rather than a 2×4 structure has been observed experimentally by electron diffraction techniques and this may be related to an unavoidable presence of one-dimensional disorder. The photoemission results provide clear evidence for a twofold periodicity and, based on the small probing range of this technique, they are in direct agreement with a 2×4 structure, although a $c(2 \times 8)$ structure cannot be rejected. The twofold periodicity is in general believed to arise from the formation of As dimers leading to a lowering of the surface energy.^{5,7,22} Depending on the model considered, dimers have been assumed to be symmetric or asymmetric and twisted as well. Obviously, the total energy of any realistic surface model should be minimal and the calculated energies of the surface electronic structure should agree with the measured electronic structure.

An extra criterion which we shall apply in the selection of models is that a calculation of the structure amplitude F for the fractional-order beams should give nonvanishing amplitudes. This calculation is essentially kinematic and although it is known that dynamic diffraction is very important for GaAs(001),^{23,24} one would not expect any intensity in a beam for which the structure amplitude is zero. F is calculated from²⁵

$$F = \sum_m f_m e^{2\pi i(\bar{k} - \bar{k}_0) \cdot \bar{r}_m}, \quad (1)$$

where f_m is the atomic scattering amplitude of the atom on site $\bar{r}_m$ and $\bar{k}_0$ and $\bar{k}$ are the incident and diffracted wave vectors of the electrons. The summation is carried out over the atoms of the surface unit cell. Writing $\bar{k} - \bar{k}_0 = (\bar{q}_{\parallel}, q_z)$, the parallel wave vectors $\bar{q}_{\parallel}$ are determined by the surface reciprocal-lattice vectors, $\bar{q}_{\parallel} = G^{r,s}$. Here $G^{r,s}$ defines the lattice rod indexed by (r,s) with $G^{r,s} = 2\sqrt{2}(r^2 + s^2)^{1/2}/a$, where a is the lattice constant.²³ The indices r and s are integers for an unreconstructed surface and can be fractions for a reconstructed surface. For the 2×4 surface the reciprocal-lattice vectors along the $[\bar{1}10]$ direction are determined by $s=0$ and $r = \pm\frac{1}{2}, \pm 1, \pm\frac{3}{2}, \dots$, while along the $[110]$ direction we have $r=0$ and $s = \pm\frac{1}{4}, \pm\frac{2}{4}, \dots$.

A. 2×4 surface models

1. 2×4 asymmetric dimer models

Asymmetric dimer models have been proposed without⁵ and with twist.⁶ They describe a complete As-terminated surface ($\Theta_{As} = 1$) and the dominant recon-

struction mechanism is the formation of asymmetric dimers. Calculations of the surface electronic structure of a 2×1 asymmetric dimer model show partial agreement with photoemission data explaining the surface states S_1 , S_2 , and S_4 , but not S_3 .⁵ The observation of a persistence of the fourfold symmetry after "hydrogen exposure" which breaks the dimer bonds indicates that the dimerization is not causing the fourfold periodicity as supposed in Ref. 5. The structure amplitudes calculated for the fractional-order beams given by $(r,s)=(\frac{1}{2},0)$, $(0,\frac{1}{4})$, $(0,\frac{2}{4})$, and $(0,\frac{3}{4})$ are all found to be zero for the model without twist, both for $\bar{q}_z=0$ and $\bar{q}_z \neq 0$. Our calculations for twisted-dimer models of the 2×4 structure assuming that the magnitude of the twist is the same for all dimers show in general very small values of F for the fractional quarter-order beams. This result is confirmed in dynamical calculations of beam intensities for GaAs(001)- 2×4 assuming asymmetric dimers with twist.²⁶

A further important point for the discussion of full monolayer coverage models is the result of recent *ab initio* self-consistent pseudopotential calculations.²⁷ These show that the electronic structure of dimer models for the complete coverage is metallic, which is in contradiction to the observed nonmetallic behavior.

2. 2×4 vacancy models

In Fig. 5 three vacancy models which were proposed in Ref. 7 are shown. Two of the structures, Figs. 5(b) and 5(c), are nearly degenerate in energy, being more stable than the structure shown in Fig. 5(a) for which the total energy is higher by 0.2 eV per 1×1 unit cell. The calculated surface-state energies of the models in Figs. 5(b) and 5(c) correspond well to the experimental photoemission data of Ref. 5.⁷ For all three models the structure amplitudes are nonzero for fractional-order beams, although F for the $(r,s)=(0,\frac{2}{4})$ beam is quite small for the models in Figs. 5(a) and 5(b) if only atoms in the first surface layer are taken into account.

Total-energy calculations⁷ for a half-monolayer As

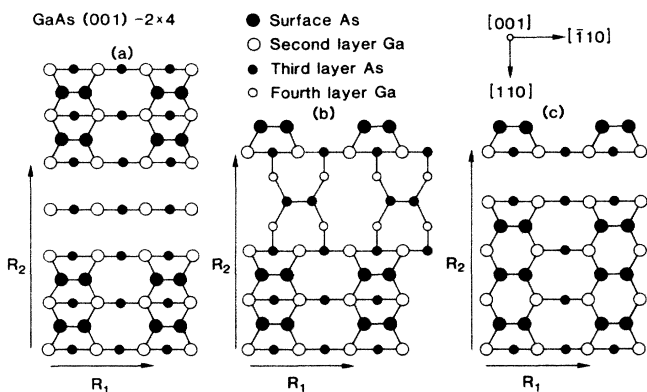


FIG. 5. Top views of three models (a)–(c) of 2×4 structures involving As dimers and missing dimers (a), (c) and also missing Ga atoms (b). From Ref. 7.

coverage show that even for this case a 2×4 [or $c(2 \times 8)$] unit cell is energetically far more favorable than a 2×2 unit cell. The energy difference of approximately 0.3 eV per 1×1 cell results from a rehybridization of Ga dangling bonds induced by structural modifications which are possible in the 2×4 [or $c(2 \times 8)$] structure but not in a 2×2 unit cell. These theoretical results suggest that a 2×4 unit cell is the optimum reconstruction for As coverage of 0.5 and 0.75, with the latter coverage leading to a lower surface energy.

It is possible to combine the models in Figs. 5(a) and 5(c) to obtain a variable stoichiometry because these structures can be derived from each other by just adding or taking away two As atoms per 2×4 unit cell. The two structures have As coverages $\Theta_{As}=0.5$ and 0.75, respectively, and for a surface having a combination of these two structures the surface stoichiometry must lie in the range $0.5 \leq \Theta_{As} \leq 0.75$. It should be noted that in preparation of the GaAs(001)- 2×4 surface by MBE, the surface is grown in an overpressure of arsenic with the incident arsenic flux typically a few times higher than the Ga flux. The arsenic flux is kept on after termination of growth (by closing the Ga cell) to compensate for evaporation of As from the surface (i.e., at growth temperatures) and the actual value of Θ_{As} must depend on incident fluxes and temperature. If we now turn to the model shown in Fig. 5(b) it is not possible to combine this with other simple models by just adding or subtracting As atoms, because Ga atoms are also missing. We therefore disregard this structure in the following.

One can also add As atoms to the structure of Fig. 5(c) and with two additional As atoms per 2×4 unit cell the As coverage will be complete, $\Theta_{As}=1$. We discussed in Sec. III A 1 above that such a structure cannot account for the experimental diffraction intensity data, but as long as Θ_{As} is not too close to 1 there can still be agreement with experimental data (excluding the disorder; see Sec. III B). For example, if $\Theta_{As}=0.95$, 20% of the surface might have the structure of Fig. 5(c) and this may be enough to account for the observed strong quarter-order fractional beams in electron diffraction which cannot be explained by a $\Theta_{As}=1$ model; see Sec. III A 1. However, the surface would also be metallic instead of nonmetallic, and this has not been observed yet.

B. $c(2 \times 8)$ vacancy models and domain formation

In the 2×4 vacancy models the phasing of the As–As dimers on either side of the missing row(s) of As atoms in the uppermost layer is the same. If the phasing of the dimers on either side of the missing rows is opposite, i.e., opposite in consecutive 2×4 unit cells along the $[110]$ direction [compare Figs. 6(a) and 6(b)] the structure has become a $c(2 \times 8)$ structure, as shown in Fig. 6, upper part. In this figure we took the structure of Fig. 5(c) as the basic unit. The formation of the $c(2 \times 8)$ structure leads to a small energy gain compared to the 2×4 structure. Total-energy calculations for a $c(2 \times 8)$ structure with $\Theta_{As}=0.75$, based on the structure of Fig. 5(c), give a smaller energy of the $c(2 \times 8)$ structure by 0.12 eV per

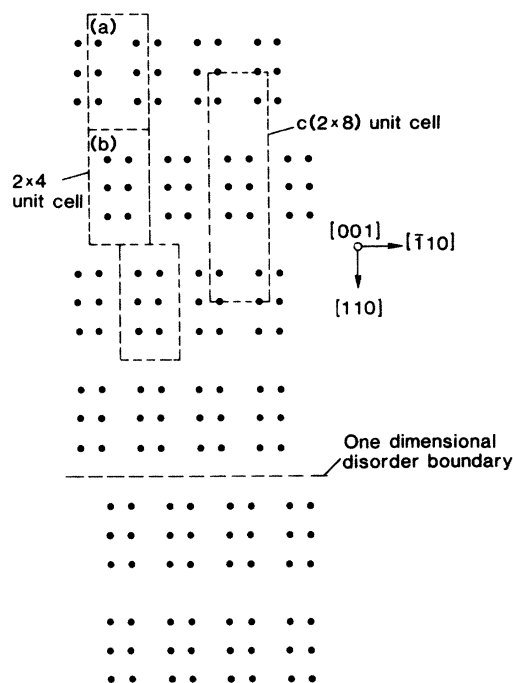


FIG. 6. Structure model of surface based on the 2×4 model of Fig. 5(c), with changes in the phasing of dimers in consecutive 2×4 unit cells along the $[110]$ direction (upper part) and with random changes in phase (lower part).

$c(2 \times 8)$ unit cell than the 2×4 structure. Experimentally, a $c(2 \times 8)$ unit cell has indeed been observed,² as mentioned in the Introduction.

In general, the electron diffraction studies show that the structure is disordered. This could be caused by an irregular phasing of the dimers in adjacent 2×4 unit cells along the $[110]$ direction, as indicated in the lower part of Fig. 6. In such a domain structure the half-order beams $(r, s) = (n/2, s)$, $n = \pm 1, \pm 3, \dots$ would be in antiphase for domains with opposite phasing of the As-As dimers and would be extinct. Such an extinction should occur for

the $r = n/2$ beams of the $c(2 \times 8)$ structure. The presence of one-dimensional disorder boundaries due to the randomness in the phasing of the dimers gives rise to the streaks observed in the half-order spots, and this was discussed in Ref. 4. The cause of the irregular phasing is directly related to the small energy difference between having the same or the opposite phasing of the As-As dimers in the 2×4 unit cells.

IV. CONCLUSIONS

The experimental evidence for a variable range of stoichiometries (i.e., As coverages) for the 2×4 and/or $c(2 \times 8)$ surface of GaAs(001) is supported by our total energy calculations which show that the optimal unit cell is 2×4 for both $\Theta_{\text{As}} = 0.50$ and $\Theta_{\text{As}} = 0.75$. The experimental results do not support the asymmetric twisted dimer model having $\Theta_{\text{As}} = 1$ but we cannot rule out the possibility of surface areas of different stoichiometry and a coverage approaching 1, e.g., $\Theta_{\text{As}} \leq 0.95$. For $\Theta_{\text{As}} = 0.50$ an alternative 2×2 unit cell can be ruled out because of its very high energy. The proposed structure for the 2×4 surface [Fig. 5(c), with $\Theta_{\text{As}} = 0.75$] gives surface electronic states in general agreement with photoemission data. A $c(2 \times 8)$ surface can easily be derived by a lateral shifting of surface As dimers from one unit cell to the next. The $c(2 \times 8)$ structure is found to have a very slightly lower energy than the 2×4 structure. This leads to the possibility of occurrence of both (2×4) and $c(2 \times 8)$ -like structures in nearly equal numbers, and explains the observations of one-dimensional disorder and a 1×4 rather than a 2×4 or $c(2 \times 8)$ structure in electron diffraction measurements.

Note added in proof. Studies of the arsenic rich GaAs(001) surface with scanning tunneling microscopy have confirmed that the fourth-order periodicity is due to a regular array of missing arsenic dimers (M. D. Pashley, Phys. Rev. Lett., to be published).

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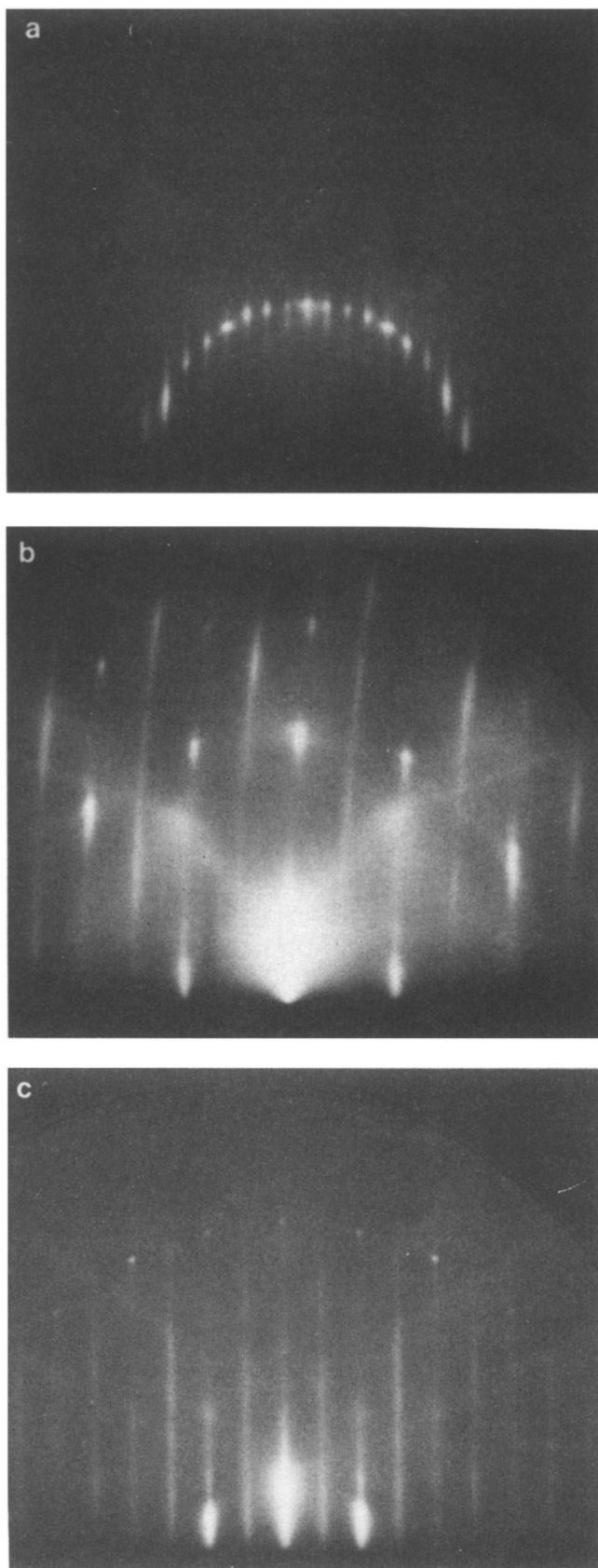


FIG. 1. RHEED patterns from a GaAs(001)- 2×4 reconstructed surface, 12.5 keV electron beam energy, $T_s = 550^\circ\text{C}$. (a) $[\bar{1}10]$ azimuth, $\Theta_i = 3.6^\circ$. (b) $[010]$ azimuth, $\Theta_i = 1.8^\circ$. (c) $[110]$ azimuth, $\Theta_i = 1.8^\circ$.

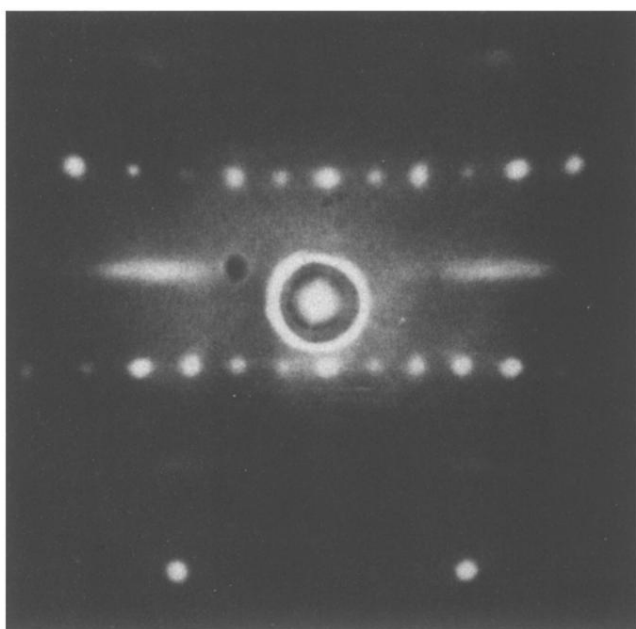


FIG. 3. LEED pattern of the As-stable GaAs(001) surface (from Ref. 14, courtesy of J. E. Crombeen).