Reconstruction, step ordering, and frustration on vicinal GaAs surfaces

L. Leprince, F. Houzay, and J. M. Moison
France Telecom, Centre National d'Etudes des Télécommunications, Paris B, Laboratoire de Bagneux, 196 avenue Henri Ravera, Boîte Postale 107, F-92225 Bagneux Cedex, France
(Received 27 April 1993)

The interaction between the step array and the terrace reconstruction of vicinal surfaces is evaluated by measuring with reflection high-energy electron diffraction the variation with the misorientation angle of the regularity of the steps and of the stability of the reconstruction. We demonstrate on GaAs≈(001) surfaces that the configurations for which the mean terrace width is a multiple of the reconstruction unit mesh, predicted by theoretical calculations to be energy minima, are a sharp optimum for *both* the step array and the reconstruction. A few minutes of arc away from this optimum, the steps modify the domain of existence and the coherence length of the reconstruction and the reconstruction modifies the step arrangement. This is clear experimental evidence that such configurations also minimize the mutual frustration of the two-ordered structures, step array, and terrace reconstruction.

The ordered defects involved in vicinal surfaces slightly misoriented with respect to a low-index plane are of great theoretical and practical[1–5] interest, especially on semiconductors. Unfortunately, little is known on the energetics which govern their structure. In addition to the energy of the low-index terrace including reconstruction terms, step and step-step energies must be considered, which makes theoretical analysis quite complicated. Experimentally, evidence has been given that those two terms can conflict. Reconstructions can reorient or roughen ledges up to one-dimensional (1D) faceting along reconstruction axes; the interstep order can be disorganized,[6] leading ultimately to step bunching or 2D faceting.[7,8] Actions of the step on the reconstruction have also been observed: *ad hoc* steps can select between reconstruction domains,[9] move reconstruction phase transition domains, or even induce new reconstructions.[10] All these effects may be viewed as manifestations of the mutual frustration of the two surface arrays—steps and reconstruction—whose periods are in general not commensurate and can anyway be modified easily through surface phase transitions or changes of the misorientation angle α, i.e., of the natural step period.

Conversely, on the basis of sparse experimental data[11,12] correlated to minimum-energy calculations, it has been suggested[1] that vicinal surfaces whose terrace width is a multiple of the unit mesh of the terrace reconstruction ("magic surfaces") correspond to energy minima and are more stable than others. In other words, the frustration is clearly smaller on these surfaces than on others since the step order is not frustrated at all and the residual frustration of the reconstruction is merely due to the finite domain size. The reduction of this residual frustration by domain, i.e., terrace enlarging, would be made at the expense of the step array order. If this is unfavorable on the whole, such surfaces are also stable minimal-frustration configurations.

Testing experimentally such a simple and elegant approach to minimal frustration, and determining the gain obtained in terms of stability and surface ordering, requires a model system. Metal surfaces display either very small or quite complex reconstruction unit meshes.[1] Even though silicon is by far the most studied semiconductor surface,[13–15] it has some drawbacks for these purposes, such as again too small or too large reconstruction unit meshes which enhance entropy terms,[16] or step height variations[8] which also generate different domains on adjacent terraces. GaAs≈(001) surfaces display $(n\times4)$ adequate reconstructions whose transitions are easily governed by the As coverage, and their double step height ensures a similar orientation of the reconstruction on all terraces.[17] Furthermore, molecular-beam-epitaxy (MBE) growth can provide a large surface mobility and hence a fast approach to equilibrium even at moderate temperatures. In this paper, by measuring the regularity and stability of vicinal GaAs≈(001) surfaces cut according to the "magic surface" or minimal-frustration criterion as compared to nearby ones, we show that such surfaces represent a sharp optimum from *both* the point of views of the step array regularity and the stability of the reconstruction.

GaAs≈(001) surfaces cut towards (111)Ga or (111)As, are called "Ga" and "As," respectively, according to the atoms forming the $[\bar{1}10]$ (respectively, [110]) edges of their As-terminated terraces. Even after growth optimization relying on *in situ* reflection high-energy electron diffraction (RHEED) observations[18–21] or *ex situ* analysis of step-guided ultrasmall deposits,[5] scanning tunneling microscopy (STM) still reveals disorder both along and perpendicular to the ledge.[22–24] RHEED and STM data both indicate that As surfaces display the better step ordering and Ga surfaces the better ledge profile, so we will consider here the rather straight Ga ledges and address their interaction with terrace reconstruction. For ordered surfaces, step-to-step distances are $(n+\frac{1}{2})a/\sqrt{2}$, where n is an integer step index and a the lattice parameter, terrace widths are $na/\sqrt{2}$ along [110], and step height is $a/2$. The two main reconstructions observed on both (001) and ≈(001) are (2×4) and (3×1), following Wood's notation along $[\bar{1}10]$ and [110]. The common unit for both step width and reconstruction domain is then $a/\sqrt{2}$.

The relationship between interstep distance and reconstruction period can be changed first through α, i.e., n. Since the (2×4) is the stable phase during growth stops, we define minimally frustrated surfaces or MFS (respectively, frustrated surfaces FS) as having terrace widths multiples (respectively, not multiples) of the (2×4) unit mesh normal to the ledges, i.e., step indices multiples (respectively, nonmultiples) of 4. Surfaces chosen here (Fig. 1) have neighboring step indices of 12 (MFS-12, three unit meshes in the terrace, $\alpha=3°14'$), 11 (FS-11, $\alpha=3°$), and 13 (FS-13, $\alpha=3°31'$); dense (001) surfaces (DS, i.e., MFS-∞, $\alpha=0$) have also been considered. Second, for a given n, we can change the reconstruction by crossing the boundaries of $(2\times4)\Longleftrightarrow(3\times1)$ reversible phase transition via changes of either As pressure P_{As} or substrate temperature T. In the (3×1) region, all surfaces become MFS ones since the "$\times1$" unit mesh along [110] can accommodate any terrace width.

Samples were obtained by conventional MBE growth. The orientation of the vicinal substrates were checked by x-ray diffraction to be accurate to within $\approx5'$, i.e., ≈0.3 step index. Non-intentionally doped GaAs was grown on them at 0.5 μm/h. Under growth conditions corresponding to the step-propagation growth mode,[21,25,26] an excellent stationary morphology was obtained for the first 500 Å. Micrometer-thick samples could then be grown without degradation of the surface morphology, and growth "accidents" could be cured, which demonstrates the good self-organization of the steps. The reconstruction was (3×1) during growth and returned to (2×4) following growth stops during which RHEED analyses were performed. Typical patterns are shown in Fig. 2. RHEED yielded key information on both the step periodicity and the terrace reconstruction.

The fluctuations within the step lattice were monitored[20,21] by the full width at half maximum W of the doublet specular beams (Fig. 3), with the incident beam perpendicular to the ledges "going down" the steps at an "out-of-phase" angle,[20] i.e., in the $[110]^*$ plane. MFS-12 displays sharp beams ($W\approx3.5$ mrad) while FS-11 and FS-13 display larger beams with inner structures depending on the incidence angle. By assuming that the several terrace widths which coexist on the surface as shown by STM images[22,23] have significant coherence domains, we may draw on Fig. 3 the peak positions expected for terraces corresponding to various integer step indices, through a simple kinematical model.[20,27] With this indexing, it may be seen qualitatively (Fig. 3) that the MFS surface mostly displays the planned terrace size $n=12$ while the FS surfaces involve terraces with $n=12$ besides terraces with the planned size $n=11$ or 13. FS step periods are therefore not stable in presence of the reconstruction: they tend to convert to the more stable neighboring MFS morphology but cannot fully do so due to the macroscopic cut angle, thus degrading the step periodicity. Upon crossing the $(2\times4)\Longleftrightarrow(3\times1)$ phase transition, W increases weakly with increasing T for FS-13, FS-11, and some MFS-12 samples, while for the other ones, it sharply increases to reach the FS value at the phase transition temperature (Fig. 4). The difference between MFS samples could not be assigned to any growth or diffraction procedure, and could be due to minute α variations. This increase of W does not correspond to the appearance of other definite step periods but rather to a statistical disordering, which disappears reversibly when the (2×4) is restored. It indicates the locking effect of the (2×4) which favors steps with $n=12$, contrary to the (3×1) which cannot have any such effect. This effect can

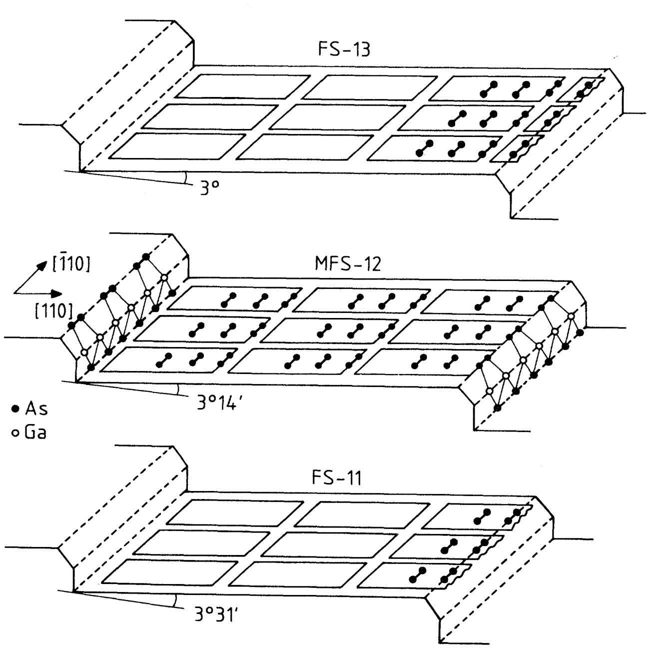

FIG. 1. Schematic view of the vicinal surfaces used in this work. The (2×4) reconstruction unit meshes are schematically pictured by rectangles involving three As dimers (short lines) and a missing dimer (Refs. 22 and 32).

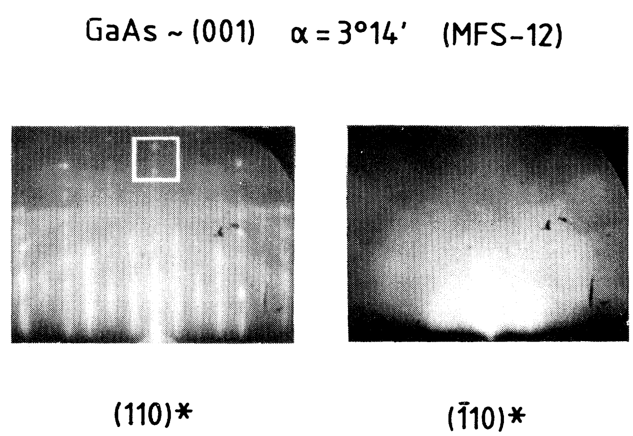

FIG. 2. RHEED patterns on MFS-12$\approx$(100)GaAs, misoriented 3°14′ towards (111)Ga. The specular doublet (white box) and the $\frac{1}{4}$ reconstruction streaks appear, respectively, on the $[110]^*$ and $[\bar{1}10]^*$ planes. Patterns were obtained at 580 °C and $P_{As}=3\times10^{-6}$ Torr, after the homoepitaxy of about 5000 Å on the base substrate. The divergence of the 10-keV primary beam was 1.5 mrad.

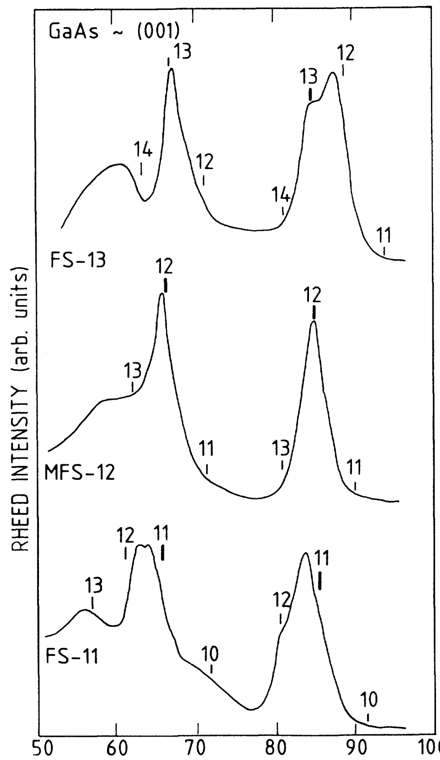

FIG. 3. RHEED intensity profiles along the specular streak for FS-11, MFS-12, and FS-13 showing the specular doublet. Peak positions corresponding to $n = 10, \ldots, 14$ step indices are calculated by a kinematical model.

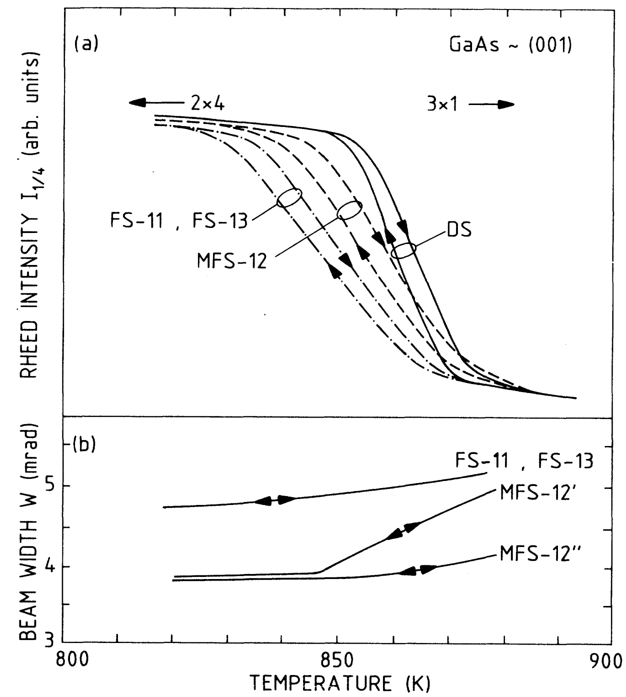

FIG. 4. Evolution during temperature cycles of (a) the intensity $I_{1/4}$ of the(2×4)-derived streaks, and (b) the full width at half maximum W of the specular doublet. MFS-12′ and MFS-12″ refer to the two behaviors observed for MFS-12. P_{As} is 5×10^{-7} Torr.

be observed only on the optimal MFS surface, and not on the two neighboring FS surfaces already disordered by the existence of $n = 12$ and $n \neq 12$ terraces. Finally, in the $[\bar{1}10]^*$ plane, i.e., for an incident beam parallel to the ledges, central streak profiles which give information on ledge roughness[21] are not significantly different for MFS and FS. Therefore, the coherence length in the ledge direction of $n = 12$ and $n \neq 12$ terraces, with, respectively, perfect and defected reconstructions, are similar for MFS and FS surfaces.

The surface reconstruction was monitored by the intensity $I_{1/4}$ of the $\frac{1}{4}$ or $\frac{3}{4}$ streak in the $[\bar{1}10]^*$ plane, with the beam parallel to the ledges. The disappearance of $\frac{1}{4}$ streaks at phase transition was observed with T scans (800 K $\rightarrow$900 K $\rightarrow$800 K). T scans performed at and below 10 K/s could be superimposed, while faster scans revealed a kinetic effect. At the chosen scan rate (≈ 1 K/s), these kinetic effects were safely avoided. Monitoring the intensity of the $\frac{1}{3}$ streak in the $[110]^*$ plane yielded curves inverted but similar though noisier. $I_{1/4}(T)$ curves obtained at $P_{\mathrm{As}} = 5\times10^{-7}$ Torr are shown in Fig. 4. Transition temperatures move with P_{As} according to an Arrhenius law $P_{\mathrm{As}} = P_0 \exp(-E/kT)$,[28,29] with P_0 increasing and E decreasing from 5 to 3.5 eV with increasing α. However, all curves obtained at various P_{As} have the same α-dependence trend. Curves for MFS and FS surfaces are similar, except that (i) the transition temperature T_{trans} (average of inflection temperatures $T_\uparrow$ and $T_\downarrow$ for increasing and decreasing temperature scans) on both FS-11 and FS-13 is lower by 10 ± 2 K than on MFS-12, and (ii) the width of the hysteresis cycle ΔT_{hys} $(=T_\uparrow - T_\downarrow)$ on both FS-11 and FS-13 is larger by 2 ± 0.5 K than on MFS-12. The presence of steps considerably changes the As adsorption-desorption regime,[29] as evidenced by the strong and monotonous decrease of E with α. It also decreases T_{trans} considerably: a 20 K difference corresponds to a change of the As pressure by nearly a decade. This corresponds to a decrease of the stability of the (2×4) phase with respect to the (3×1) phase due to the limitation of its spatial extension. It must be stressed that this stability does *not* decrease monotonously with α, but is higher for the MFS than for the *two* neighboring FS. When the (2×4) reconstruction is frustrated by the step array, its stability with respect to the nonfrustrated (3×1) is then decreased, and conversely the minimal-frustration configuration represents an optimum for the reconstruction energy term.

In conclusion, we have examined the step arrangement regularity and the reconstruction stability of GaAs $\approx(001)$ vicinal surfaces, near to the "minimally frustrated" surfaces for which the planned terrace width is a multiple of the reconstruction unit mesh. Even though quantitative analysis of the data obtained remains out of reach at present and though the question of a genuine equilibrium situation cannot easily be addressed as indeed in most similar experimental situations, these surfaces clearly represent an optimum from the points of view of *both* the reconstruction and the step array, as suggested by theoretical calculations.[1] Most data pertaining to the step array and the reconstruction pass through an *abrupt*—a few minutes of arc—*extremum* at the minimal-frustration angle. Out of this optimum, the step modify, not the reconstruction itself, but its domain of existence or its coherence length, and the reconstruction modifies, not the step structure, but its arrangement. Even though confirmation by large-scale STM pictures is still needed, this is a clear check that a mutual frustration of these two-ordered structures occurs most often, and that only a very particular choice of natural interstep distance can minimize this frustration and lead to better organized surfaces, possibly on any material. Considering the residual roughness of such surfaces, it may be noted that the creation of elementary kinks on otherwise perfect ledges increases the frustration of the reconstruction on both adjacent terraces. STM pictures[13,22] have indeed shown a tendency for ledge irregularities to involve no longer atoms but whole reconstruction unit cells, larger and hence less mobile. This may lead in turn to a reduction of mobility along the ledges, which addresses the question whether surfaces can reach full equilibrium, even though no experimental evidence of a departure from equilibrium exists as long as growth provides the necessary mobility. All this clearly must be taken into account in cases where step regularity on the ultimate scale is needed, for instance, to guide an otherwise isotropic growth[30,31] and build extremely small and extremely regular structures such as quantum wires or boxes, later superlattices or serpentine superlattices.[5] From a more general point of view, the 2D frustration system formed by steps and terrace reconstruction is shown here to be experimentally speaking a fairly simple and well-defined one, in which the frustration can be adjusted around its local minimum by changing external parameters. This makes it a promising one for theoretical approaches and deeper studies on the mutual locking of step array and reconstruction, disordering by local strain, reconstruction stability, and phase transition in limited domains, etc.

The authors gratefully acknowledge fruitful discussions with C. A. Sébenne and J. M. Gerard. Vicinal surfaces were cut and polished by A. Daniel. X-ray measurements of the misorientation angles were performed by G. Le Roux. Laboratoire de Bagneux is "Unité Associée au Centre National de la Recherche Scientifique" (CNRS) UA250.

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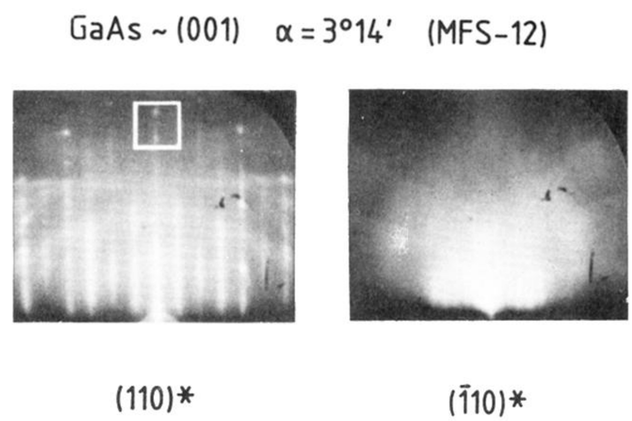

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