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Indium-rich 4x2 Reconstruction in Novel Growth of InAs on the GaAs(001)*

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Molecular beam epitaxy (MBE) of the lattice mismatched InAs/GaAs(001) system is studied by *in situ* scanning tunneling microscopy (STM) and reflection high energy electron diffraction (RHEED). We found that deposition of submonolayer (~0.6ML) In on the GaAs(001)-As-rich 2x4-β surface could result in a new 4x2 reconstruction, and that if the growing front maintains this reconstruction, the multilayer InAs grows two-dimensionally and the commonly observed three-dimensional islanding is completely surpressed. The atomic structure for this new 4x2 reconstruction is discussed on the basis of voltage-dependent STM images. In addition, a "domain wall" structure is discussed, representing a new type of strain relief mechanism in the layer-by-layer growth reported here.

KEYWORDS: molecular beam epitaxy, scanning tunneling microscopy, heterojunction, InAs, GaAs

1. Introduction

"Self-organization" in heteroepitaxially lattice-mismatched systems, arising from strain-induced coherent 3-D islanding has been of great importance for fabrication of 0-D confined nanostructures.[1-6] In a traditional "quantum wells" structure where electrons are confined only in one direction, one expects that the Stranski-Krastanov 2D-3D morphology transition can be delayed or surpressed, a more planar growth and thus both morphologically and compositionally abrupt interfaces can be achieved, which are crucial for the optimum performance of electronic and optoelectronic devices based on the superlattice and heterojunction.[7-14] For the InAs/GaAs with a lattice mismatch of 7.2%, in order to reduce the accumulated epitaxial strain energy in the grown layer, the 2D-3D transition occurs at approximately 2MLs (the so-called critical thickness) under standard MBE growth conditions.

MBE growth of any epitaxial film is inherently a nonequilibrium process, the morphology of a growing film and its evolution are governed by an interplay between kinetic and thermodynamic aspects of MBE growth.[11-14] This provides us with an opportunity to control growth mode by manipulating the growth conditions and kinetics. One of the examples is that by using extremely In-rich growth conditions, the usually observed 3-D morphology roughening could be significantly surpressed [7-10]. Schaffer et al. have reported a basically 2-D growth even for an epilayer thickness of 1800Å under such conditions,[7] which has resulted in superior interface quality and remarkably improved photoluminescence intensity.[8] For this novel growth, several questions raise: (1) how does the surface

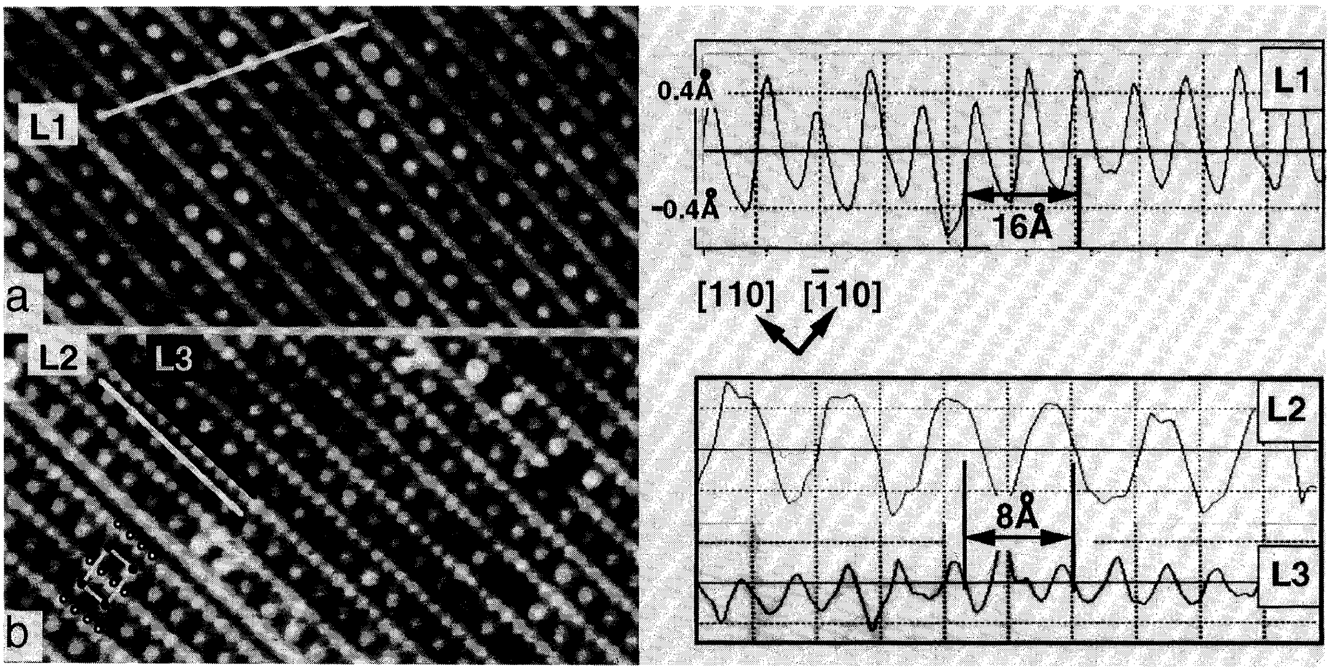

Fig. 1. Filled states STM images of the new In-terminated 4x2 surface. (a) Vs = -2.2V; (b) Vs = -1.6V.

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morphology really like? (2) what kind of surface structure/reconstruction results? (3) what type of strain relaxation mechanism works and how does it work? (4) it is due to limited kinetics or energetically favorable? In the present study, by using a high-performance *in-situ* MBE-STM system, we have investigated the evolution of the surface morphology and structure as a function of the III-V flux ratio and epilayer thickness in the present layer-by-layer growth of InAs/GaAs(001), aiming at understanding the questions (at least partially) above at the atomic scale.

2. Experiment

The MBE-STM system used in the study and the preparation of the GaAs(001)-2x4 substrates have been described elsewhere.[15] The starting InAs wetting layer was prepared by depositing submonolayer (approximately 0.5ML) In on the As-rich 2x4 surface at a substrate temperature of 450°C. After deposition, the sample was allowed to maintain at 450°C for about 5min. The surfaces prepared in this way display a very sharp 4x2 RHEED pattern.[7] The subsequent multilayer InAs growth was carried out by using migration enhanced epitaxy (MEE) to eliminate kinetic complications. Our "quench-and-look" STM experiment reveals that the As/In flux ratio and shutter switching time in MEE are critical for the layer-by-layer mode and that a smooth surface cannot grow unless the resulting growth front reproduces the 4x2 symmetry for the wetting layer.[16] These observations demonstrate the very special function of the phase serving as template for the planar growth.

3. Results and discussion

Shown in Fig. 1(a) are typical filled-states STM images of the 4x2 surface. Fig. 1(a) was recorded at a sample bias of -2.2V while Fig. 1(b) at -1.6V. The cross-section profiles L1, L2 and L3 are shown in the lower panel of the figure. The STM image exhibits a high degree of perfection of this surface, without any kink defects. This surface is quite different from the GaAs(100) As-rich 2x4, Ga-rich-4x2 and InAs(001)-4x2,[17, 18] and consists of straight line and evenly spaced humps along the [110] direction with a unit cell consisting of 16.0Å x8.0 (±0.23)Å (the 4x2 unit cell is highlighted by a rectangle in Fig.1(b). The 4x along the [-110] direction and the 2x along the [110] direction translational symmetries are more clearly indicated by the cross-section profiles L1 and L2, respectively. In these filled states images, we don't see much contrast change for the line and the hump. When the bias voltage becomes smaller, we did observe a distinct 1x periodicity along the line built up by smaller humps, as indicated by the cross section profile L3. If studying the Fig. 1(b) more carefully, we find that the small humps forming the 1x periodicity always straddle the larger 2x humps along the [110] direction, which are highlighted by the empty and the solid circles, respectively, revealing that the 2x and 1x humps should tunnel from different species. All these basic features are quite different from what observed for the homoepitaxially grown InAs (001) In-rich 4x2 surface [17, 18], and also for the 2x4 substrate,[15] therefore, they should have different structures. Because the 2x direction agrees with the In dimerization direction, we conclude that these 2x (larger) humps are due to tunneling from In-dimers.

We also obtained the empty states STM images (not shown here). All the basic hump-plus-line features are exactly the same as those in the filled states image, but the 2x humps become brighter. This contrast change corresponds to our identification that the 2x humps are due to tunneling from the In-atoms.[19] Based on these observations, we assign the individual 2x humps to be tunneling from the first layer In-dimers, the lines from the second layer As, and propose a tentative model for this new reconstruction in Fig. 2.

According to this model, the two-fold periodicity is due to the dimerization of the newly deposited In adatoms in the [110] direction, and the regular In-dimer missing gives rise to the four-fold periodicity along the [-110] direction. The exposed As adatoms in the missing rows dimerize to their neighbors and their uniform arrangement explains the 1x periodicity along the line as shown in Fig. 1(b). The model abides with the electron counting model and assures that all the arsenic dangling bonds fully filled with two electrons and all the In dangling bonds empty to leave no net charge at the surface, corresponding to a stable semiconducting surface.[20] The model gives a surface In coverage of 0.5ML, in good agreement with the experiment. The observed contrast differences between the filled- and empty-states images are accounted for by the different energy levels of the As dangling bonds and In dimer anti-bonding orbitals.[19, 20]

Once the substrate is wetted with the above 4x2 structure, the progressive InAs multilayer can be grown two-dimensionally under a very simple and strict condition; the In/As_4 flux ratio, substrate temperature and deposition rate must be chosen so that the growing front could reproduce the 4x2 symmetry. To demonstrate this point, in Fig. 3(a) and (b), we show two STM images obtained from the surfaces deposited with 4ML and 13ML (they are much thicker than the critical thickness of ~2ML) of InAs, respectively. No 3D island is observed at this stage and the surface is basically flat at the atomic scale, indicating a novel layer-by-layer growth.

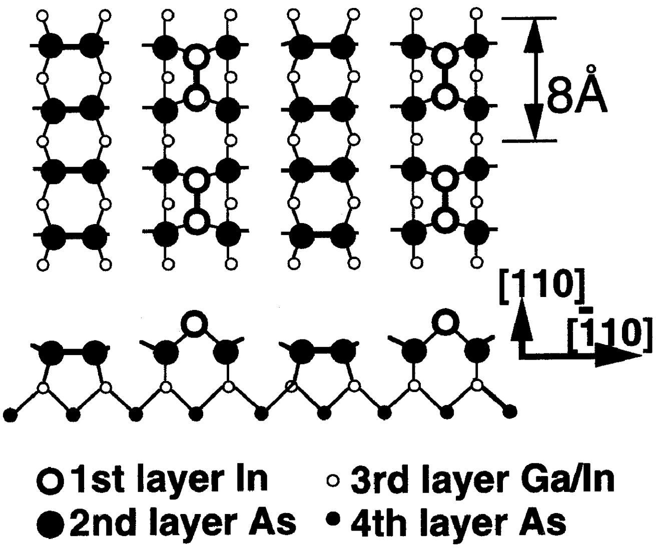

Fig.2. The ball-to-stick model proposed for the 4x2 surface. The black solid circles represent As atoms and the open circles the In atoms.

As the use of MEE and extremely In-rich conditions assure enough adatom diffusion, the change in the growth mode is obviously not due to limited kinetics. If it is this case, one would expect an earlier 2D-3D transition. We therefore agree with Snyder et al. that the layer-by-layer growth mode observed presently is due to increasing surface tension,[9] the In-rich reconstruction has much higher surface tension than that of the As-rich one. In the traditional Stranski-Krastanov growth of strained layer, 3-D islanding relieves the elastic energy, but increases the surface area and thereby costs surface energy. The relaxation of elastic energy is dominant and 2D-3D transition is favorable where the surface tension is small. With increasing surface tension, however, the driving force for such a picture diminishes. In order that the energy gain due to a strain reduction in the islands outweighs the surface energy increase, the 2D-3D transition must be delayed. The calculation of surface tension energy for this new 4x2 and the critical thickness for the In-rich growth condition forms an interesting topic for future theoretical investigation.

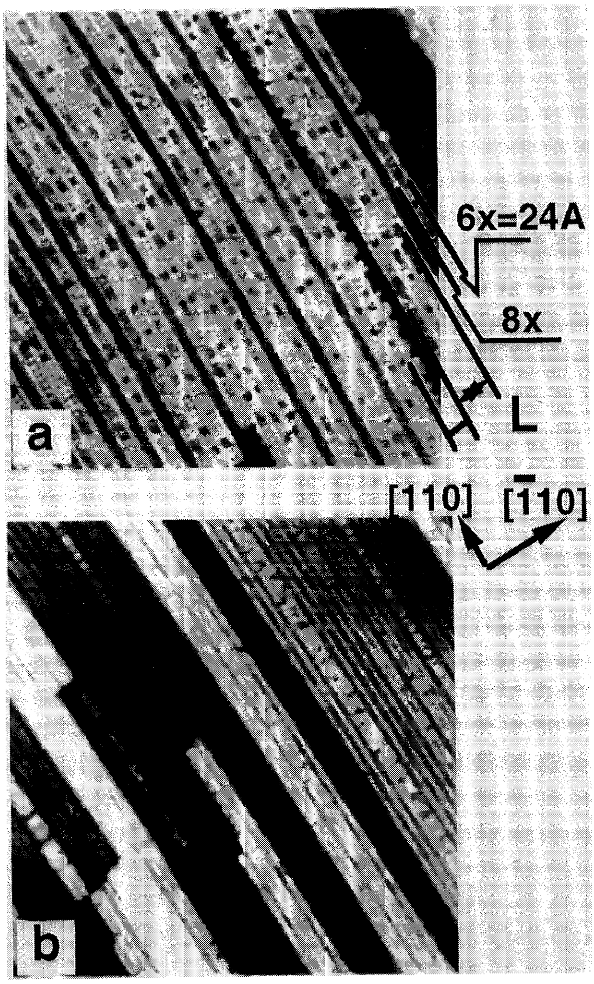

Fig.3. The surface morphologies after (a) 4ML, (b) 13ML InAs epilayer deposition on the GaAs(001) substrate under In-rich growth condition. It is evident that with increasing epilayer thickness, the formation of domain walls becomes more and more frequent.

Despite the smooth surface morphology, the STM images (Fig. 3(a) and (b)) do not show a perfect 4x2 ordering compared to Fig.1, although the whole RHEED displays an essential 4x2 symmetry. Now the surface mainly contains the 4x2 domain but is modulated with characteristic dark lines running from a step edge to another along the [110] direction, forming a unique "domain wall" structure with a characteristic wall separation of Na_0. Statistically, N=6 is dominant for both 4ML and 13ML cases. The "domain wall" structure appears at the same position in the empty states image, indicating its geometric origin. These features are similar to the 2xN structure on the strained Ge/Si interface and the vacancy defect bands induced by Ni on the Si(001) surface,[22-24] which has been attributed to be the consequences of surface strain relief. We believe that these regularly distributed "walls" may represent a new type of strain relaxation mechanism, and might be important and potential sites for the nucleation of misfit dislocations. Since no screw and other misfit dislocations were observed by the STM, their relationship is still unclear. A corresponding TEM experiment will be helpful to understand the question.

4. Conclusions

We have studied the novel 2-D growth of highly strained InAs on the GaAs(001) As-rich 2x4 substrate by *in situ* STM. We observe a new reconstruction that displays the 4x2 symmetry. Based on the voltage-dependent STM images, a tentative model characterizing the regular In-dimer rows and missing dimer rows, is proposed. We also document that the formation of "domain walls" structure is a result of strain relaxation in the novel 2-D growth

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