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GaAs(001) (2 × 4) to c(4 × 4) transformation observed in situ by STM during As flux irradiation

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ABSTRACT

Atomic resolution scanning tunnelling microscopy (STM) has been used to study in situ the As-terminated reconstructions formed on GaAs(001) surfaces in the presence of an As_4 flux. The relationship between the As-rich (2 × 4) and c(4 × 4) surfaces is observed throughout the gradual evolution of the reconstruction transformation. The results suggest that during the initial stage of the transformation, Ga-rich As-terminated variations of the c(4 × 4) form in order to accommodate excess mobile Ga produced by pit formation. These transient structures later planarize, as excess Ga is incorporated at step/island edges. Successive imaging of the same sample area during As_4 irradiation allows point-by-point adatom binding to be analysed in a way inaccessible to MBE–STM systems relying on sample quenching and transfer.

1. Introduction

GaAs(001) exhibits a myriad of reconstructions depending on As overpressure, Ga:As flux ratio and sample temperature [1], however the (2 × 4) and c(4 × 4) reconstructions have received that greatest attention [2,3] due to their existence within the temperature and flux range typically used for high quality GaAs and InGaAs growth. An improved fundamental understanding of these surfaces could play a significant role in future device fabrication, especially for self-assembled InGaAs quantum dots, since here a partial transition from (2 × 4) to c(4 × 4) can be observed in the early stages of growth just before an InAs wetting layer forms.

STM has primarily been employed post growth after sample quenching has been performed [4–6]. Performing STM outside the MBE growth environment clearly prevents access to growth dynamics and introduces uncertainty over the precise nature of the quenching process, for which there has been very little work to validate. This is specifically problematic for the GaAs system where complex As molecular binding underpins epilayer growth [5].

More recently, attempts have been made to combine MBE and STM in the same chamber, in both group IV [7] and group III–V systems [8]. In the latter case an STM stage has been customised to operate in a pre-existing MBE chamber. In this work MBE sources are integrated into an STM chamber in a commercially available Omicron MBSTM system. The system supports sample heating and As_4 irradiation during sample imaging, providing a mechanism to observe As molecule interchange dynamically on the surface.

Studies of the (2 × 4) to c(4 × 4) transformation have identified $\beta 2$ as the generally accepted (2 × 4) model (Fig. 1a) but the exact composition of the c(4 × 4) reconstruction is still under debate. The simplistic model of As–As dimers simply back-bonded upon the existing dimer rows of the (2 × 4) (Fig. 1b) perhaps over simplifies the actual atomic binding configuration, for which Ga incorporation has also been speculated [9,10].

The transition from (2 × 4) to c(4 × 4) normally takes place between 450 °C and 550 °C, depending on the applied As flux. At these relatively low temperatures, Ga surface atoms will be relatively immobile due to limited thermal kinetics. This transport of Ga would be expected to play a key role in the reconstruction transformation, with local-range Ga transport responsible for pit and island formation during the formation of a c(4 × 4) surface on a (2 × 4) and vice versa [10]. Simplistically a single unit cell of $\beta 2$(2 × 4) can be transformed into a (1 × 1) layer by either the addition of 0.25 ML of Ga and 0.5 ML of As or the removal of 0.75 ML Ga and 0.5 ML As, allowing a (1 × 1) to be created either in line with the existing (2 × 4) or 1 ML below, respectively. The (1 × 1) layer forms the nucleus of the traditional c(4 × 4) model (Fig. 1b) and would require an additional 0.75 ML of As to produce the reconstruction. Since islanding requires only a third of the Ga produced by the formation of an identical sized pit, it can be assumed that a two-level c(4 × 4) surface would exhibit a 3:1 island:pit ratio. It has been observed experimentally that this is

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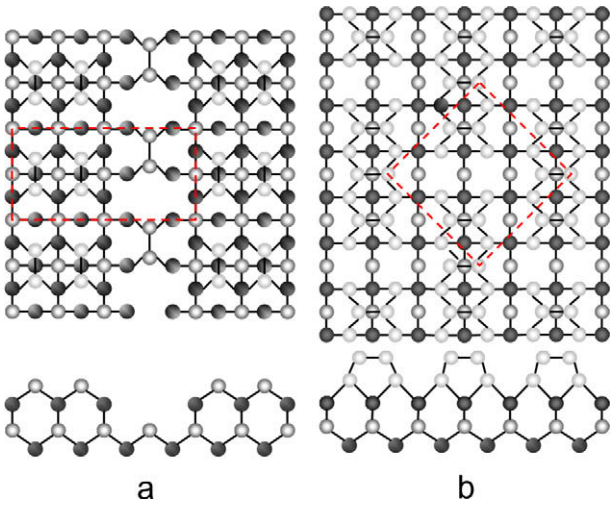

Fig. 1. Model of GaAs(001) As-rich reconstructions. (a) $\beta2(2\times4)$ with 1/2 ML of As in the upper layer and 3/4 ML of Ga in the second layer, (b) $c(4\times4)$ with 3/4 ML of As back-bonded onto a full ML of As.

not the case [2,11], so the question remains: where is the excess Ga incorporated? The presence of hetro-dimers [10,12] have been proposed to explain the excess Ga incorporation, a structure easily detectable with STM due to the sensitivity to empty and filled states. Furthermore, the reported coexistence of (2×4) and $c(4\times4)$ [13] raises a question to the presence and stability of the theoretical $\gamma(2\times4)$ between the established $\beta2(2\times4)$ and the $c(4\times4)$.

In this paper we report observations of in situ STM in which an As flux is utilised during a high temperature measurement. We are able to monitor the evolution of a single $150\times150\ nm^2$ area, in order to observe atomic binding during the reconstruction transition. This unique measurement allows a direct comparison of the same atomic sites in a manner which would be impossible for conventional MBE, followed by quenching and then STM. With the ability to watch a step-by-step evolution of the surface, the presence and relative density of transient reconstructions can be monitored.

Perfectly ordered (2×4) dimer rows are readily prepared in an MBE reactor. With the assistance of RHEED intensity observations, the preferred $\beta2(2\times4)$ can be preserved during either rapid [14] or slow/compensation [15] quenching. More recently surface stress analysis has investigated the dimer bonding model for GaAs(001) reconstruction [16]. These techniques concentrate on obtaining an average across a relatively large area. However the ability to observe atomic scale interactions gives a more detailed insight into the mechanics of the reconstruction transformations.

Only small fluctuations in As flux are necessary for broken dimers or adsorbed As to appear randomly across the surface. Whether the $\alpha(2\times4)$ and $\gamma(2\times4)$ are reconstructions in there own right or RHEED intensity alterations as a consequence of coexisting reconstructions is still debateable, however what is clear is that the (2×4) pattern has stoichiometric variations depending on the preparation parameters. It is equally probable that the arguably more complicated $c(4\times4)$ reconstruction has an equally diverse nature.

In this paper, a well ordered (2×4) surface is quenched and then, after transfer to the STM chamber, the sample is heated under an As flux. The goal being, firstly, to investigate the pit and island formation of $c(4\times4)$ in order to observe and better understand their origins and secondly to observe the surface occupancy of the $\gamma(2\times4)$ phase to confirm whether it is widespread before the formation of $c(4\times4)$ or merely a consequence of coexisting phases. Samples prepared *in situ* have been used to explore the transition to $c(4\times4)$ and have purported that the $c(4\times4)$ forms both above and below the initial (2×4) [17,18]. In this paper an As_4 flux has been applied to a stable (2×4) and the transition is observed dynamically for the first time using STM.

2. Experimental details

This work was performed on a commercially available Omicron MBSTM system. The system comprises of an interconnected conventional MBE chamber and an MBSTM chamber. The former chamber is fitted with standard III–V sources and RHEED for dynamic surface analysis. The latter chamber includes a Omicron VT-STM head and a small source loaded with metallic arsenic. This provides an As_4 flux directly onto the sample during STM surface imaging. Full details of the system are discussed elsewhere [19].

A Si-doped vicinal GaAs(001) epi-ready wafer was cleaved to $10\times3.9\ mm^2$ in order to accommodate the sample mounting. Heating is achieved through a heating plate, whereby a current is passed through a piece of PBN in close proximity to the sample. This plate has been shown to provide uniform heating with 2 °C variation across the entire sample when observed by thermography camera. The sample and plate form a single unit that can be passed between the MBE and STM chambers, where identical contacts provide a heating current in each case.

With no additional ex situ cleaning, standard oxide removal was performed the MBE chamber under a 3.0E-6 mbar As_4 beam equivalent pressure (BEP) at 580 °C and then a 0.5 μm buffer layer was grown at this temperature with As:Ga flux ratio of 30:1. This resulted in strong a (2×4) RHEED pattern and STM observations showed a smooth surface comprised of single bilayer steps.

The sample was then quenched to ~420 °C in a similar manner to that outlined by Yang [15]. The RHEED pattern was monitored throughout the quenching, with particular attention given to the intensity of the 2nd order diffraction rods. With the sample held at 420 °C all additional As was pumped from the growth chamber before the sample heating was removed and the sample was swiftly transferred into the MBSTM chamber.

STM tips were prepared with a 2 step process. The first involved electro-chemically etching high purity <1 1 1> 0.4 mm W wire in 3 M NaOH solution. Tips that showed short shanks and sharp apices under light optical microscopy were then placed in a custom designed heating plate and inserted into the vacuum system. Six 10 s heating cycles were then applied to the apex at 900 °C, subliming the oxide. Initial STM scanning at room temperature revealed a highly ordered $\beta2(2\times4)$ reconstruction across the entire surface. The sample was then ramped to ~400 °C within the STM stage and left for 5 h to stabilise. Scanning commenced and continued until the *z*-drift was negligible and the *xy*-drift was in the order of 0.05 nm/s and could be compensated.

Whilst 400 °C is lower than the temperatures normally used for GaAs/GaAs (570–590 °C) or InAs/GaAs (480–510 °C) it is necessary in this instance to choose a lower temperature. We believe that As-rich (2×4) reconstruction is only stable in the absence of an As flux from 400;°C to 430 °C [11]. Since the sample temperature must be allowed to stabilise for several hours prior to imaging in the absence of an As flux, this temperature was chosen to maintain a highly ordered (2×4) surface. Since this represents the lowest temperature at which (2×4) is stable in the absence of As, any external flux will instigate a transformation to $c(4\times4)$. This allows a very low flux to be applied (As BEP = 2.0E-8 mBar), hence prolonging the transition to accommodate multiple STM images.

At this stage a suitable area was selected and the As e-beam source was operated to achieve a stable flux of 200 nA (~2.0E-8 mBar BEP). The cell shutter was then opened and maintained as such throughout. The images presented in this study each take 400 s to capture and comprise of the image immediately before