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Enabling electron diffraction as a tool for determining substrate temperature and surface morphology

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The reconstruction transitions for the GaAs(001) surface have been identified as a function of the band gap-derived substrate temperature and As_4 beam equivalent pressure. Surface morphology measurements using *in situ* scanning tunneling microscopy reveal that the surface spontaneously forms a random distribution of two-dimensional islands. The onset of island formation is coincident with the reflected high-energy electron diffraction pattern changing from the β to α subphase of the (2×4) reconstruction. An electron diffraction-based method for determining the substrate temperature and engineering the surface morphology with a desired amount of roughness is presented. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1416477]

Numerous technological and fundamental breakthroughs have occurred simply due to the ability to deposit one material on top of another. A significant leap forward in the quality of the deposited layers came with the introduction of *in situ* electron diffraction.[1] Initially, it was used to uncover the periodic properties of the surface atomic structure.[2–4] Later on, the peak intensities were found to oscillate in time during growth which allowed the measurement of the rate material was being deposited.[5] As the understanding of the relationship between the atomic-scale surface morphology and the diffracted electrons has grown, so has the quality and capabilities of the growth process.[6,7] The complex nature of the growth process can still benefit from further refinements in the understanding of the relationship between the diffraction pattern, the surface morphology, the substrate temperature, and the elemental flux.

The III–V(001) surfaces exhibit a plethora of surface reconstructions, all with different stoichiometries and symmetries. Most of the well-ordered structures can be understood with thermodynamic models, indicating that the reconstructions are a fundamental property of the equilibrium state.[8] Specifically, first-principles theory calculations are routinely carried out, and tend to accurately reflect experimental observations.[9–12] Experimentally, the transitions between these reconstructions occur as a function of temperature and group-V flux.[13–16] Determining the precise temperature at which these transitions occur is problematic due to the temperature measurement schemes commonly employed in molecular-beam epitaxy (MBE) systems. For example, differences as great as 140 °C between the actual temperature and the thermocouple (TC) derived temperature have been observed.[17] To mitigate this problem, a common practice for the GaAs(001) surface is to calibrate the TC to read 580 °C when the oxide is first removed. Even though this procedure is transferable from lab-to-lab, it can be performed only at one temperature and only at one particular time. In this letter, the ability to use electron diffraction as an *in situ* tool to measure the substrate temperature as well as to determine the aperiodic surface morphology is presented.

Experiments were carried out in an ultrahigh vacuum (UHV) multichamber facility ($5-8\times10^{-11}$ Torr throughout) which contains a solid-source MBE growth chamber (Riber 32P), incorporating a reflection high-energy electron diffraction (RHEED) system and a highly accurate (±2 °C), optical transmission thermometry system for substrate temperature determination.[18] The temperature is determined by first measuring the fundamental band gap of the substrate, and then comparing this to a database to convert the band gap to the temperature.[19] The MBE chamber also has an all UHV connection to a surface analysis chamber, which contains a custom-integrated commercially available (Omicron) scanning tunneling microscope (STM) for surface morphology measurements.[20]

The RHEED measurements were performed on commercially available, ‘‘epiready,’’ *n*-type (Si doped $10^{18}/cm^3$) 2-in. GaAs(001) ±0.05° substrates that were loaded into the MBE system without any chemical cleaning. The surface oxide layer was removed at 590 °C while exposing the surface to a 10 μTorr As_4 beam equivalent pressure (BEP) from a solid-source valve-controlled cell. A 1.5-μm-thick GaAs buffer layer was grown at 580 °C using a growth rate of 1.0 μm/h as determined by RHEED oscillations, and an As_4 to Ga BEP ratio of 15. After growth, the surface was annealed under an As_4 BEP of 1 μTorr for 15 min at 600 °C followed by another at 570 °C under the same conditions. This procedure improves the RHEED pattern and prepares the surface for RHEED measurements. Surface reconstructions for a fixed As_4 BEP were identified by either heating or cooling the substrate in 10 °C increments, waiting 15 min, and recording the RHEED pattern in the [110], $[1\bar{1}0]$, and [100] directions. This procedure was repeated for five different As_4 BEPs by adjusting the valve position on the arsenic cell. In addition, the RHEED patterns were measured without any As_4 BEP. This was done by first creating a $c(4\times4)$ pattern at low temperatures and low As_4 BEPs. Then, the As_4 BEP was eliminated which leaves the surface with a $c(4\times4)$ symmetry. By holding the substrate temperature below 350 °C, with no As_4 BEP for 30 min, the background As_4 was removed

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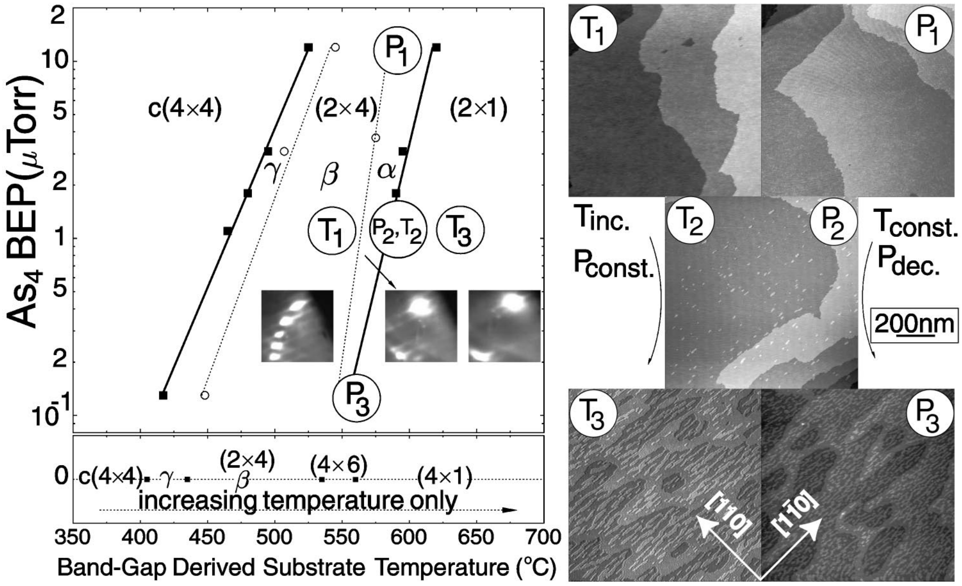

FIG. 1. (Left-hand side panel): RHEED-derived transition pressures (As_4 BEP) and temperatures for the GaAs(001) surface reconstructions. The lines are drawn as a guide for the eye. (Right-hand side panel): 1 μm $\times 1$ μm filled-state STM images taken at a sample bias of -3.0 V. The images labeled T_1 to T_3 represent an increasing-temperature, constant-pressure data series, while the images labeled P_1 to P_3 represent a decreasing-pressure, constant-temperature data series. The pressure-temperature annealing conditions for the STM samples are cross referenced in the RHEED diagram.

from the chamber by the ion pump. RHEED patterns were then recorded in 10 °C increments as just described.

The STM measurements used identical but smaller substrates, the oxide was removed, a buffer layer was grown, and the surface was prepared as in the RHEED experiments. After preparation, the sample was then annealed at a desired As_4 BEP and temperature condition. While quenching to room temperature at 1.5 °C/s, the RHEED pattern was monitored to ensure it did not change. The sample was then transferred to the STM without breaking the UHV and imaged at room temperature. To confirm the high-temperature annealing procedure produced a sample that had reached equilibrium, the anneal times were successively increased until the surface morphology, as viewed with STM, remained unchanged (~10 min). For the equilibrated samples, multiple filled-state STM images were acquired using tips made from single crystal ⟨111⟩-oriented tungsten wire, a sample bias of -3.0 V, and tunneling current between 0.05–0.2 nA.

Pressure and temperature conditions for the transitions between various surface reconstructions as observed by RHEED are shown on the left-hand side of Fig. 1. Increasing the substrate temperature at any nonzero As_4 BEP results in the surface reconstruction changing from the $c(4\times4)$ to the (2×4) to the (2×1), as indicated by the solid lines.[16] Decreasing the substrate temperature reverses the reconstruction sequence at the same temperatures. Within the (2×4) phase, three separate RHEED patterns were identified as the α, β, and γ subphases, and the transitions between these are identified with dashed lines. The β subphase RHEED pattern in the "4-by" direction consists of a specular, 1/4, 1/2, 3/4, and primary-order spots from top to bottom all with equal intensity, as illustrated by the photograph displayed in the region labeled β.[16] The α subphase pattern, is similar to the β except the 1/2-order spot is missing, as also illustrated in Fig. 1.[16] A characteristic (2×1) RHEED pattern in the "1-by" direction is also shown. The zero As_4 BEP data series is displayed on a separate graph directly below the logarithmic plot. The transition temperatures are marked with solid squares, and they happen only when the substrate temperature is increased from the $c(4\times4)$ phase. The pattern then changes to the (2×4) the (4×6), and finally to the (4×1).

Characteristic 1 μm$\times 1$ μm STM images of the morphology of the GaAs(001) surface are shown on the right-hand side of Fig. 1. The labels for each image also mark the location on the RHEED reconstruction diagram where the anneal occurred. The "T" labels represent an increasing-temperature, constant-pressure (constant As_4 BEP) data set through the β, α, and (2×1) phases, while the "P" labels represent a decreasing-pressure, constant-temperature data set also through the three phases. The images are displayed in gray scale, and unprocessed except for having a (001) plane subtracted from them to make each terrace a distinct color separated from the other by a 0.28-nm-high step. The trend in all the images is that the coverage of the islands increases with either increasing temperature or decreasing pressure. The lowest temperature and highest pressure annealing conditions produce a β subphase RHEED pattern, and a surface with large well-ordered terraces free of islands, as shown in T_1 and P_1 of Fig. 1. The temperature-pressure condition that produces the α subphase RHEED pattern shows a surface with 10% of each terrace covered with one monolayer high GaAs islands, as shown in T_2 and P_2 of Fig. 1. These islands have spontaneously formed on the surface due to annealing (i.e., without depositing any material from the gallium effusion cell).[21] The pressure-temperature conditions that produces a (2×1) RHEED pattern shows a surface with even more islands, as shown in T_3 and P_3 of Fig. 1. Here, the surface roughness due to spontaneous island formation is so great that it makes it difficult to identify the original terraces. Notice, the islands still favor elongation in the $[1\bar{1}0]$ direction, and they avoid forming double height steps.

One of the most difficult problems within the MBE community is sharing knowledge of the substrate temperature for successful growth algorithms. This is due to poor lab-to-lab reproducibility in the thermocouple and pyrometer methods of substrate temperature determination.[17,18] It is possible to use *in situ* RHEED to measure the substrate temperature by referencing the RHEED diagram shown in Fig. 1. This can be accomplished by using one of two methods; first, by holding the thermocouple derived substrate temperature fixed and adjusting the As_4 BEP until a transition [e.g., $c(4\times4)$ to (2×4)] is observed, and then, by knowing the As_4 BEP at which the transition was observed the true substrate temperature can be read off the diagram in Fig. 1 and used to calibrate the thermocouple temperature. Alternatively, one can hold the As_4 BEP fixed and adjust the thermocouple-derived substrate temperature until a transition is observed. Using the surface reconstruction transition method provides a large dynamic range, since the RHEED diagram has a transition at any temperature between 400 °C–600 °C. In addition, this method of substrate temperature determination is not subject to lab-to-lab variations, since the reconstruction transitions are independent of the substrate properties, such as dopant type and concentration.[9,22,23] In fact, we speculate that the only requirement is that ~10 monolayers of single crystal GaAs(001) be present. This methodology can be carried out on other III–V(001) surfaces, as well.[24]

Not only can RHEED be used for determining the substrate temperature, it can also be used to monitor the aperiodic surface morphology. The STM images indicate the β to α RHEED transition is coincident with spontaneous island formation. That is, when a β pattern is present at low temperatures the surface is flat, while when an α pattern is present at low temperatures the surface is covered with two-dimensional, GaAs islands.[21] We believe the rapid cool down procedure does not affect the surface. The concentration of islands and their geometry can be tuned and the process is reversible. Therefore, it is possible to use *in situ* RHEED to control the surface morphology by referencing the RHEED diagram shown in Fig. 1. This can be accomplished by using one of two methods. First, increasing the substrate temperature while holding the As_4 BEP constant increases the island coverage. Alternatively, decreasing the As_4 BEP while holding the substrate temperature constant also increases the island coverage. This capability makes it possible to engineer interfaces with a tunable amount of roughness. Consequently, it may be possible to systematically study of the role of interface roughness on the transport or optical properties in reduced-dimensionality structures.[25]

Typically, diffraction patterns are used to predict atomic structural models simply from their symmetry. The $c(4\times4)$, (2×4), and (4×6) symmetries have been successfully modeled in this way. Interestingly, the other symmetries presented, namely the (2×1) and (4×1), have not been successfully modeled. In fact, it is believed that any structure with a 1-by feature would violate the electron counting model.[10] In these structures, it is believed that disorder is dominating the RHEED properties.[26] Nevertheless, little theoretical work has been done to understand the role of disorder on RHEED. A prescription for producing surfaces with a well known and tunable amount of disorder now exists for use as a test bed for such theory. These effects may play an important role in understanding the β to α RHEED transition, and possibly aid in understanding the general influence of disorder on diffraction.[27]

In conclusion, an algorithm for using RHEED on the GaAs(001) surface to determine the substrate temperature is presented. This method can be used in MBE machines even if they do not have access to a transmission thermometry system or on samples which are heavily doped or indium mounted. We believe this capability will aid in the lab-to-lab transfer of successful growth procedures. In addition, an algorithm for using RHEED to prepare a surface with a tunable amount of roughness is presented. We believe this capability may lead to a better understanding of the relationship between the interface roughness and the optical and transport properties in confined structures, as well as aid in the understanding of the role of disorder on diffraction.

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