

## Role of aperiodic surface defects on the intensity of electron diffraction spots

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(Received 30 September 2002; accepted 19 February 2003)

A random distribution of two-dimensional gallium arsenide (GaAs) islands is found to effect the intensity of the electron diffraction pattern from the GaAs(001) surface. By utilizing the spontaneous island formation phenomenon as well as submonolayer deposition, the island coverage is systematically changed. It is found that the intensities of the one-, two-, and three-quarter-order diffraction spots of the  $[1\bar{1}0]$  azimuth decrease as the concentration of islands increases. In addition, only in the presence of islands, does the intensity of the half-order spot decrease as the grazing angle of the electron beam is decreased. A simple quantitative model is developed that provides insight into how an aperiodic arrangement of islands effects the electron diffraction patterns. © 2003 American Institute of Physics. [DOI: 10.1063/1.1568161]

The quality of growth on the GaAs(001)-(2×4) reconstructed surface plays a critical role in optoelectronic device fabrication.<sup>1</sup> Electron diffraction is the most widely used tool to monitor the quality of the surface during growth. Interpreting the electron diffraction patterns can be difficult, since a large variety of different surface structures can yield similar diffraction patterns. Within the low-energy electron diffraction (LEED) community, obtaining the structure from a LEED pattern is known to be complicated and typically requires a sequence of patterns taken at various beam energies as well as extensive computational efforts.<sup>2-4</sup>

In molecular-beam epitaxy (MBE), typically only a monoenergetic reflection high-energy electron diffraction (RHEED) experiment is used to monitor the surface during growth. When a surface is well ordered, kinematic RHEED calculations are often used to predict the surface structure.<sup>5</sup> However, without an energy-dependent data set (e.g., like the LEED data), the surface structure prediction is likely erroneous. It is even more complex to get the crystal structure using RHEED when aperiodic features exist on the surface. This situation exists during crystal growth, where it has been observed that the half-order diffraction spot is absent from the diffraction pattern when the electron beam is directed along the  $[1\bar{1}0]$  direction (i.e., the “4-by” direction).<sup>6</sup> Historically, this was evidence that as islands form on the surface, the electron diffraction pattern is effected. Later, RHEED spot intensities were observed to oscillate in time and in phase with each monolayer of material deposited.<sup>7</sup>

Congruently, many studies on the GaAs(001) surface have documented a systematic approach to forming an aperiodic distribution of monolayer high islands.<sup>8-10</sup> These studies show that by simply altering the As<sub>4</sub> overpressure or changing the substrate temperature, any desired island coverage can be reversibly tuned without changing the local atomic structure.

In this letter, we take advantage of the well-documented crystal structure and surface morphology of the GaAs(001)-(2×4) reconstructed surface to learn about its role in electron diffraction. Specifically, the RHEED spot intensities are measured as the island coverage and grazing angle of the electron beam are systematically changed. A simple quantitative model is developed, which shows how islands on a surface give rise to a decrease in the intensities of the 1, 2, and 3 quarter-order diffraction spots from the  $[1\bar{1}0]$  azimuth.

Experiments were carried out in an ultrahigh vacuum (UHV) multichamber facility ( $5-8 \times 10^{-11}$  Torr throughout) which contains a solid-source MBE chamber (Riber 32P), incorporating a RHEED system (12 keV), a substrate temperature determination system accurate to  $\pm 2^\circ\text{C}$ ,<sup>11</sup> and an arsenic cell with an automated valve and controller. The MBE chamber also has an all UHV connection to a surface analysis chamber, which contains a custom integrated scanning tunneling microscope (STM).<sup>12</sup> All experiments were performed on commercially available, “epiready,” *n*-type (Si-doped  $10^{18}/\text{cm}^3$ ) GaAs(001)  $\pm 0.1^\circ$  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  $\mu\text{Torr}$  As<sub>4</sub> flux. A 1.5- $\mu\text{m}$ -thick GaAs buffer layer was grown at 580 °C using a growth rate of 1.0  $\mu\text{m}/\text{hr}$ . as determined by RHEED oscillations, and an As<sub>4</sub> to Ga beam equivalent pressure (BEP) ratio of 15.

Samples were prepared with either spontaneously formed islands or deposited islands. After the oxide was removed and the buffer layer grown, the following procedure was used to form the spontaneous two-dimensional (2D) islands on the surface.<sup>9</sup> Samples were annealed for 15 min at a specific temperature and As<sub>4</sub> BEP to form the desired island coverage. In addition to the spontaneously formed islands, a submonolayer coverage of GaAs islands was obtained by depositing GaAs. These samples are prepared by annealing at 560 °C under a 10  $\mu\text{Torr}$  As<sub>4</sub> BEP to produce a flat well-ordered (2×4) surface. Next, the substrate temperature was

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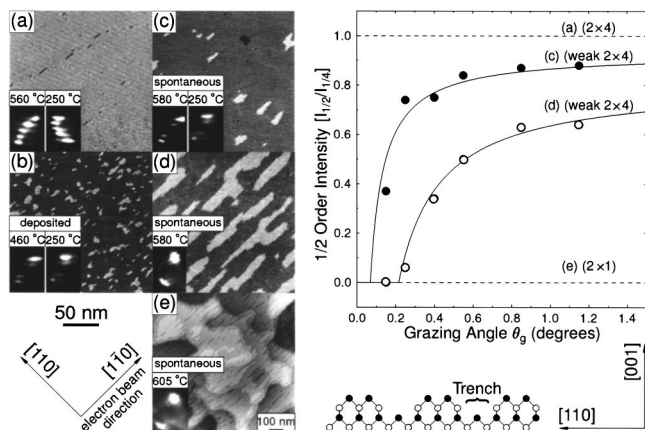


FIG. 1. (a)–(d) Filled-state STM images ( $-3.0$  V and  $0.1$  nA) with corresponding RHEED patterns taken along the  $[1\bar{1}0]$  direction while annealing at the indicated temperatures. (a) ( $560$  °C,  $10.0$   $\mu$ Torr  $\text{As}_4$  flux) Flat surface showing 4-by RHEED patterns. (b) Surface covered with 10% of deposited one-monolayer-high islands with weak 4-by RHEED patterns. (c) ( $580$  °C,  $1.1$   $\mu$ Torr  $\text{As}_4$  flux) Flat surface covered with 10% of spontaneously formed one-monolayer-high islands with weak 4-by RHEED patterns. (d) ( $585$  °C,  $1.1$   $\mu$ Torr  $\text{As}_4$  flux) Flat surface covered with 25% of spontaneously formed one-monolayer-high islands with weak 4-by RHEED patterns. (e) ( $605$  °C,  $0.35$   $\mu$ Torr  $\text{As}_4$  flux) Filled-state STM image ( $-3.0$  V and  $0.1$  nA) with 1-by RHEED pattern. Upper right-hand side: Plot of the RHEED peak intensity ratio vs grazing angle for the different annealing conditions. The solid lines are intensity vs grazing angle theory curves, using an island length of  $35$  nm and four different coverages as seen in the STM images. To help bracket the extremes, two dashed lines are also drawn. Lower right-hand side: Side view of the atomic structural model for the GaAs(001)-(2 $\times$ 4) reconstructed surface.

reduced to  $460$  °C and then 10% of a plane of Ga was deposited under an  $\text{As}_4$  flux of  $1.0$   $\mu$ Torr. Finally, the substrate was quenched to room temperature. Additional details of the STM experiments have been explained elsewhere.<sup>8,13</sup>

For both types of samples, the peak intensity of the half-order and quarter-order RHEED diffraction spots from the  $[1\bar{1}0]$  azimuth were measured as a function of grazing angle of the electron beam (beam energy of  $12$  keV was used). This measurement was accomplished by translating the sample on a micrometer driven manipulator. The change in  $\text{As}_4$  flux due to the translation of the sample was measured to be less than 5%. In addition to the grazing angle measurements, RHEED patterns were recorded with the electron beam running along the  $[1\bar{1}0]$  direction at high temperatures directly before the cool down ( $\sim 1.5$  °C/s). An additional set of RHEED measurements were performed at low temperatures ( $\sim 250$  °C) for both the 10% spontaneous and 10% deposited samples.

An STM image of the surface prepared by annealing at  $560$  °C while under a  $10.0$   $\mu$ Torr  $\text{As}_4$  flux shows a flat (2 $\times$ 4)-reconstructed surface with a low density of defects and is displayed in Fig. 1(a). The image reveals lines running diagonally, which represent the “4-by” line spacing of the atomic structure.<sup>14</sup> Inset in this image are two RHEED patterns taken at the temperatures indicated. These RHEED patterns are similar and show a 4-by pattern which consists of the specular, quarter, half, three quarter, and primary-order spots from top to bottom all with equal intensity. An STM image of the surface prepared by depositing 10% of a plane of Ga shows a flat surface having about 10% of the surface covered with one-monolayer high GaAs islands and is dis-

played in Fig. 1(b). Again, the rows of the 4-by reconstruction can be seen, even on top of the islands. In the inset of Fig. 1(b) are the two RHEED patterns, which show a weak 4-by pattern missing the half-order spot. Prior to deposition the half-order diffraction spot was not missing and the RHEED pattern was identical to the RHEED pattern shown in Fig. 1(a).

An STM image of the surface prepared by annealing at  $580$  °C while under a  $1.1$   $\mu$ Torr  $\text{As}_4$  flux shows a flat surface having about 10% of the surface covered with one-monolayer high GaAs islands and is displayed in Fig. 1(c). These islands have spontaneously formed on the surface due to annealing at a high temperature (i.e., without depositing any material from the gallium effusion cell).<sup>8</sup> The inset of Fig. 1(c) are two RHEED patterns also showing a weak 4-by pattern. An STM image of the surface prepared by annealing at  $585$  °C while under a  $1.1$   $\mu$ Torr  $\text{As}_4$  flux shows a flat surface having about 25% of the surface covered with one-monolayer high spontaneously formed GaAs islands and is displayed in Fig. 1(d). The inset in Fig. 1(d) is one RHEED pattern, which shows a 4-by pattern. An STM image of the surface prepared by annealing at  $605$  °C while under a  $0.35$   $\mu$ Torr  $\text{As}_4$  flux shows a rougher surface with 2D GaAs islands on top of other 2D islands and is displayed in Fig. 1(e). Notice the scale of this image is different from the others, to show the large scale multilevel roughness. The inset of Fig. 1(e) is one RHEED pattern, which shows a 1-by pattern. Notice that the overall background intensity is brighter for this RHEED image than the others. This was done to enhance the details in the image. When this enhancement was required, we would define the pattern as a “1-by” instead of the usual 4-by.

The ratios of the half- to quarter-order peak intensities for the RHEED patterns discussed herein are plotted as a function of grazing angle and shown as line profiles on the right-hand side of Fig. 1. The ratio is calculated in order to eliminate any spurious effects that may cause the overall intensity of the beam to change. The intensity ratio remains constant at all grazing angles for a well-ordered surface [Fig. 1(a)], as shown in the line profile labeled (a). When a small concentration of islands is present, the intensity ratio increases from 0% to over 60% with grazing angle as shown in the line profiles labeled (c) and (d). Once the surface becomes three-dimensional, the half-order diffraction spot intensity goes away, as shown in the line profile label (e).

Everytime the GaAs(001) surface is flat a 4-by diffraction pattern is observed with equal intensity in all the spots. This pattern is a consequence of the long-range periodic order of the atomic structure. A side view of the atomic structural model is shown in Fig. 1 (lower right-hand side). Simple kinematic calculations show that diffraction from the trench (see Fig. 1) is a necessary requirement to having all the diffraction spots (e.g., the half-order spot). That is, by not including the atoms within the trench, the half-order spot would be missing.

After adding only a small amount of GaAs to the surface, either through deposition or spontaneously formed, a weak 4-by diffraction pattern occurs with a missing or weak half-order spot. This missing spot can reoccur simply by lowering the substrate temperature and/or raising the  $\text{As}_4$

BEP. Given the half-order diffraction spot is the first one effected and that this spot requires diffraction off the trench layers, it is reasonable to assume the islands somehow disrupt the trench layer diffraction process. Furthermore, as the surface gets rougher, all the quarter-order diffraction spots disappear and the reconstruction appears ( $2\times 1$ ). It has been shown that step edges adversely effect the diffraction intensity.<sup>15,16</sup> Here, the step edge density increases as the number of islands increases. Nevertheless, the STM images reveal that the RHEED-derived ( $2\times 1$ ) surface is locally well ordered in a ( $2\times 4$ ) arrangement, but without the long-range periodicity.<sup>8,9,14,17</sup>

When the surface shows a weak “4-by” pattern, it is interesting that simply changing the grazing angle of the electron beam makes the spot come and go. Clearly, there is no way that changing the angle of the beam can effect the surface crystal structure. Also, notice that the intensity of the half-order diffraction spot does not change with angle when there are no islands present [see line profile (a)]. This “control” sample shows that over this angular range, there are no spurious factors related to the scattering process that can change the intensity half-order diffraction spot as a function of grazing angle. All totaled, this suggests that when islands are present, the smaller grazing angles make diffraction off the trench atoms more difficult.

A mechanism that causes the half-order spot to disappear is uncovered by critically analyzing the intensity profiles displayed in the right half of Fig. 1. We propose that the islands geometrically block the electrons that would normally diffract off the trench atoms, resulting in the disappearance of the half-order spot. This is reasonable, due to the mean-free path of the electrons being much shorter than the average size of the islands.<sup>18</sup> In addition, as the angle of the beam is lowered, the distance the electron must travel through the surface increases. This model begins by considering an island of length  $\ell$  which fills in the trench with atoms and blocks all the electrons along a path of length ( $\mathcal{L} + \ell$ ), where  $\mathcal{L} = 2d/\tan(\theta_g)$  is the lateral distance that the electron is below the top layer of atoms,  $d$  is the vertical distance to the subsurface feature, and  $\theta_g$  is the grazing angle of the electron beam. The fraction of the surface that is not accessible for electron diffraction,  $F$ , due to the islands can be written as  $F = \rho(\mathcal{L}/\ell + 1)$ , where  $\rho$  is the fractional coverage and we assume the islands do not overlap. If the intensity,  $I$ , of the half-order spot is assumed to be proportional to the accessible fraction of the surface,  $(1 - F)$ , then it can be written as

$$I(\rho, \ell, \theta_g) = I_o \left[ 1 - \rho \left( \frac{2d}{\ell \tan(\theta_g)} + 1 \right) \right], \quad (1)$$

where  $I_o$  is the intensity without islands. This predicted intensity change with angle is plotted with the data points shown in Fig. 1 (solid lines). The most important fitting parameter in our model is the fractional coverage,  $\rho$ . The coverage parameter was set equal to the STM measured coverages of 0.1 and 0.25, for curves (c) and (d), respectively. The

island length fitting parameter,  $\ell$ , has only a small effect on the intensity for this angular range. A good fit to the data is obtained when an average island length of  $\ell = 35$  nm is used, which is also consistent with the STM images. Surprisingly, even a low coverage of islands can block a significant fraction of the surface. Equation (1) was derived for the  $[1\bar{1}0]$  direction, however, this approach can be generally applied to any azimuthal direction if one has an understanding of the relative top layer and subsurface layer contributions to the diffraction pattern. To help bracket the extremes, two dashed lines are also shown in Fig. 1. Without any islands, the RHEED pattern shows equal intensity in all orders, while with high coverages, the half-order spot is missing.

In summary, we have utilized the well-documented phenomenon of spontaneous island formation to discover how the RHEED diffraction pattern is effected by aperiodic defects. The half-order diffraction spot is found to be critically sensitive to small coverages of GaAs islands, as well as the angle the electron beam hits the surface. We quantified this relationship using a model that has the islands shadowing the diffraction process. Since electron diffraction is ubiquitously used to monitor the quality of the surface, this case study may prove critical to learning how aperiodic structures effect diffraction.

This work is supported by the National Science Foundation Grant No. FRG DMR-0102755 and C-SPiN MRSEC DMR-0080054.

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