Grazing incidence small-angle X-ray scattering from laterally ordered triangular pyramidal Ge islands on Si(111)

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We present a structural study of nearly perfect triangular pyramidal Ge islands grown on Boron-terminated Si(111). Using atomic force microscopy we show that these islands are strongly ordered in the direction perpendicular to the miscut induced terrace steps of the substrate. We use *grazing incidence small-angle X-ray scattering* to assess the island shape and size distribution. In this geometry the scattered wave amplitude is composed of four terms, including all combinations of scattering from the islands and reflection from the substrate. We finally show that this technique is able to determine the complete symmetry of arbitrary shaped objects on a substrate.

1. Introduction

The growth of heteroepitaxial layers with a small lattice mismatch to the substrate has been of great interest recently, due to the possibility to grow nanometer-sized structures. Following the development of Ge hut clusters on Si(100) (Mo et al., 1990), several types of islands have been observed in different systems (Legoues et al., 1994; Leonard et al., 1993 and Rasmussen et al., 1997). For better device performance of quantum-dot structures, the objective now is to obtain islands of the same size, shape and strain-state, and to find a mechanism for self-organized ordering (Tersoff et al., 1996). The size and shape of islands are usually determined using microscopy techniques, which provide us with information about their symmetry and faceting. In the case of buried islands, however, these techniques are of limited use. X-ray scattering is able to determine the island shape (averaging over a much larger number of islands) as well as strain-state (Kegel et al., 1998).

We present here a structural study of Ge islands on Si(111) using atomic force microscopy (AFM) and grazing-incidence small-angle X-ray scattering (GISAXS). Similar systems have already been studied (Voigtländer and Zinner, 1993 and Köhler et al., 1991), but usually truncated or shapeless structures are obtained. We have grown nearly perfect triangular pyramidal Ge islands, which are aligned along the miscut induced terraces of the Si substrate. Their intrinsic symmetry allows a much better shape control of the islands. Whereas (100) islands can be elongated in two independent directions, (111) triangular islands can only be rescaled. These triangular islands were formed by the deposition of Ge on Si(111) using boron as a surfactant. With AFM we observed a strong tendency of ordering of the islands in the direction perpendicular to the terrace steps. We used GISAXS to determine the average shape and size distribution of the islands. We show that the X-ray wave scattered by the islands, described by the Distorted-Wave Born Approximation (DWBA), is composed of four terms, including possibilities of scattering from the nanostructures and reflection from the substrate. We demonstrate that GISAXS allows us to determine the *full symmetry* of the islands, a task that transmission small-angle X-ray scattering can hardly accomplish.

2. Experimental

The Ge/Si(111) sample was prepared by molecular beam epitaxy as described in Rupp et al. (1996). The substrate (kept at 530^0C) was a 0.45^0 miscut Si(111) wafer, on which a 1500Å Si buffer layer was grown, followed by a 0.33 monolayer of boron, which is known to induce a $\sqrt{3}x\sqrt{3}$ reconstruction on the Si(111) surface (Zotov et al., 1996). A 150Å thick Ge layer was subsequently deposited at 0.25Å/s yielding nearly perfect triangular pyramidal islands. The deposition of 0.33 monolayer of boron saturates all dangling bonds of the Si substrate and enhances the lateral diffusion of Ge, allowing the formation of perfect structures. For comparison, a sample grown without boron did not yield pyramidal islands (Kovats, 1999).

3. Results

The inset of fig.1 shows the detailed shape and size of a few Ge islands, determined using AFM. They are completely pyramidal with well-defined (113) facets with a base side L ~ 2500Å and a height H ~ 350Å, and some exhibit a very small (111) terrace on top with L_{top} ~ 100Å. We find the three baselines of the pyramids to be oriented along the equivalent {1-10} directions (arrows). The lateral organization of the islands can be seen in fig. 1, where a tendency of arrangement is visually clear. The islands are organized in rows perpendicular to the miscut of the substrate (arrow pointing to $\omega = 45^0$, where $\omega = 0^0$ corresponds to the {1-10} direction), which was determined using high-resolution X-ray diffraction. This means that the islands are aligned parallel to the terrace steps of the substrate.

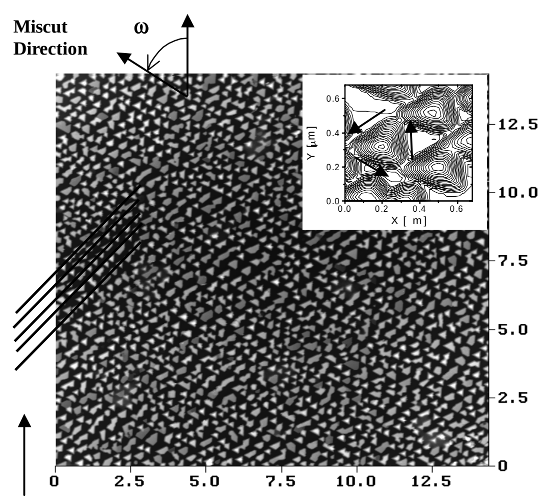

Figure 1 <1 –1 0> <1 1 –2>
Atomic force microscopy of Ge islands on Si(111). Notice the alignment of the islands given by the parallel lines. The inset shows their triangular shape and the alignment along the {1-10} directions as indicated by the arrows.

The degree of ordering was evaluated from the AFM measurement by calculating the height-height correlation function of the islands as shown in fig. 2 for directions parallel and perpendicular to the terrace steps. After a pronounced first maximum at R ~ 3600Å, the correlation function parallel to the terrace steps decays slowly and is eventually flat. In the miscut direction (i.e., perpendicular to the terrace steps), however, we observe a clear oscillation of the correlation function up to the ninth neighbor. Thus we conclude that the islands show a tendency of ordering, where the period of this oscillation corresponds to the average spacing of the islands of 3600Å. A mechanism for this alignment of islands may be the confinement of the islands to a single terrace of the substrate. In this case the distribution of islands follows the terrace configuration. Considering the miscut of 0.45^0 of the Si(111) substrate and assuming that the terraces are separated by one single Si(111) bilayer (≅ 3.14Å), the average terrace width would be ~ 400Å, and therefore, considerably smaller than the average distance of 3600Å between islands. This indicates the presence of step bunching, resulting in larger terraces (Suzuki et al., 1996). Such a mechanism of self-organization of islands opens the possibility of obtaining ordered arrays of Ge dots directly on the substrate.

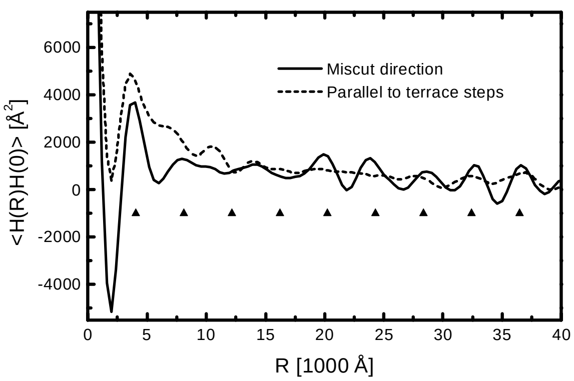

Figure 2
Height-height correlation function of the Ge islands for the directions parallel and perpendicular to the terrace steps (miscut direction). The triangles indicate the peak positions if the lined up islands would be equally spaced.

We turn now to a grazing incidence small angle x-ray scattering experiment, where the same Ge/Si(111) sample was taken to the X-ray beamline ID-3 at the European Synchrotron Radiation Facility (ESRF), in Grenoble, France. As shown in fig. 3a, the sample surface was illuminated at a grazing angle α_i with a collimated X-ray beam of wavelength $\lambda = 1.22$ Å. The scattering angle α_f was set to 0.7^0 to collect diffuse intensity. The island induced diffuse scattering was measured with a position-sensitive detector parallel to the sample surface as a function of $q_{//} \cong (2\pi/\lambda) \cos \alpha_f \sin 2\theta$. The scattering was measured for several ω with constant $\alpha_i = 0.44^0$ and $\alpha_f = 0.7^0$. Fig. 3b shows scans in this scattering geometry, where the symmetry properties of the scattering patterns clearly change with the azimuthal angle ω. At $\omega = -30^0$ the scattering pattern is symmetrical. For $\omega = 0^0$ the scattered intensity exhibits a shoulder on the right side of the central peak. This shoulder moves to the left when $\omega = 60^0$, and returns to its original position for $\omega = 120^0$ (data not shown). The patterns are symmetrical when $\omega = -30^0$, 30^0, 90^0, 150^0, etc. The symmetry properties of these scattering patterns prove the triangular symmetry of the islands.

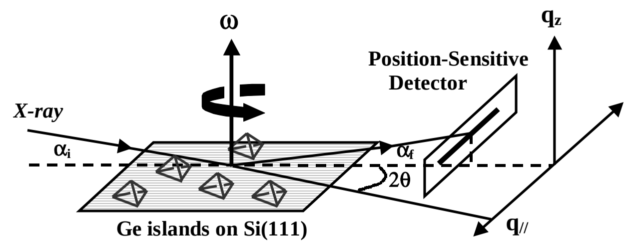

Figure 3a
Scattering geometry of GISAXS used to study the triangular Ge islands.

To reproduce the X-ray scattering patterns and thereby quantify the shape of the islands we have calculated the scattering cross section of triangular pyramids in GISAXS geometry in the distorted-wave Born approximation (Rauscher et al., 1999). As illustrated in fig. 4, due to the small angle of incidence of the X-ray beam, there is the possibility of reflection of the incident wave and subsequent scattering by the individual islands. As a result, the small angle scattering cross section is composed of four terms: scattering from the islands, scattering from the islands and subsequent reflection from the substrate, reflection from the substrate and scattering from the islands and double reflection from the substrate with island scattering in between. The scattered intensity is given by

$$I \propto |F(\mathbf{q}_{//},q_z)+R(\alpha_i)*F(\mathbf{q}_{//},p_z)+R(\alpha_f)*F(\mathbf{q}_{//},-p_z)+R(\alpha_i)*R(\alpha_f)*F(\mathbf{q}_{//},-q_z)|^2 \quad (1)$$

where R is the substrate reflectivity, $q_z = k_f - k_i = 2\pi/\lambda(\sin\alpha_f + \sin\alpha_i) = 0.103Å^{-1}$, $p_z = k_f + k_i = 2\pi/\lambda\ (\sin \alpha_f - \sin \alpha_i) = 0.0237Å^{-1}$. $F(\mathbf{q}_{//},q_z)$ is the structure factor of the pyramids, which is given by

$$F(\mathbf{q}_{//},q_z) = \int_0^{H(1-L_{top}/L)} F_\Delta\big(q_x,q_y,a(z)\big)e^{iq_z z}dz \quad (2)$$

where a(z) = (1-z/h) L and $F_\Delta(\mathbf{q}_{//},q_z,a)$ is the Fourier transform of a triangle of side a(z)

$$F_\Delta(q_x,q_y,a) = \frac{\sqrt{6}e^{-i\frac{\sqrt{3}q_y a}{6}}}{q_z(q_x^2-3q_y^2)}\left[q_x e^{i\frac{\sqrt{3}q_y a}{2}} - q_x\cos\left(\frac{q_x a}{2}\right) - i\sqrt{3}q_y\sin\left(\frac{q_x a}{2}\right)\right] \quad (3).$$

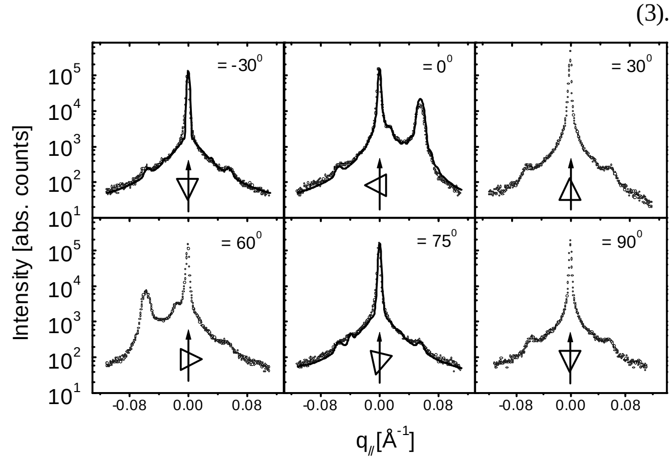

Figure 3b
X-ray scattering patterns from the islands. The triangles indicate the orientation of the islands with respect to the incident beam.

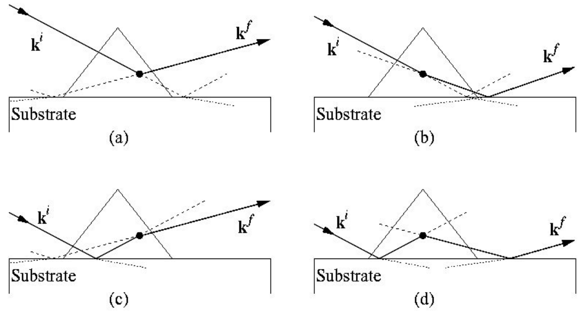

Figure 4
The four possibilities of scattering in GISAXS geometry: a) direct scattering by the island; b) scattering by the islands and subsequent reflection by the substrate; c) reflection by the substrate and scattering by the islands and d) double reflection by the substrate with the scattering in between.

As a result, the scattering patterns in fig. 3b are the sum of four different Fourier transforms. In fact, for the $\omega = 0^0$ pattern, the strong shoulder on the right corresponds to the first Fourier transform of eq. 1 with $q_z = 0.103Å^{-1}$, whereas the small shoulder near the central peak corresponds to the second transform, with $p_z = 0.0237Å^{-1}$.

Least-squares fits of 3 different patterns of fig. 3b were performed simultaneously, yielding $H = (390 \pm 30)$ Å, $L = (2600 \pm 200)$ Å, $L_{top} = (130 \pm 100)$ Å and a Gaussian size distribution (FWHM = 10%). A small (0.5%) contribution from arbitrarily oriented islands was added to simulate the outer shoulders which are due to the side faces of misoriented and shapeless islands. The fits also included a small diffuse scattering Lorentzian term (FWHM = $0.018Å^{-1}$) induced by a number of structures that have coalesced.

4. Discussion and conclusions

The grazing-incidence geometry (with $q_z \neq 0$) *was essential to determine the triangular symmetry* of the islands. If a similar experiment were performed with $q_z = 0$, (e.g. in transmission geometry) the diffuse scattering would exhibit a 6-fold symmetry instead of the 3-fold pattern. To illustrate this difference, in figs. 5a and 5b we show contour plots of the calculated scattering patterns for $q_z = 0$ and for $q_z = 0.1025Å^{-1}$ as a function of q_x and q_y. Clearly the $q_z = 0$ pattern has a 6-fold symmetry, and it is not possible to determine *neither the shape nor the orientation* of the triangular pyramids.

In conventional transmission small angle scattering geometry, since $q_z = 0$, the structure factor $F(\mathbf{q}_{//},0)$ is the same as $F(-\mathbf{q}_{//},0)$, ($F(\mathbf{q}) = F(-\mathbf{q})$, which corresponds to Friedel's law (Cowley, 1975), which says that the inversion of a crystal through a center of symmetry does not change the diffracted intensity. Therefore, for a triangular objet we obtain a six-fold scattering pattern. In the case of grazing incidence geometry, however, since $q_z \neq 0$, $F(\mathbf{q}_{//},q_z)$ is not necessarily the same as $F(-\mathbf{q}_{//},q_z)$ and we can circumvent this phase problem and observe the triangular symmetry of our islands.

We emphasize here that this feature of GISAXS can be used to determine the full shape and symmetry of an arbitrary nanostructure on a flat surface.

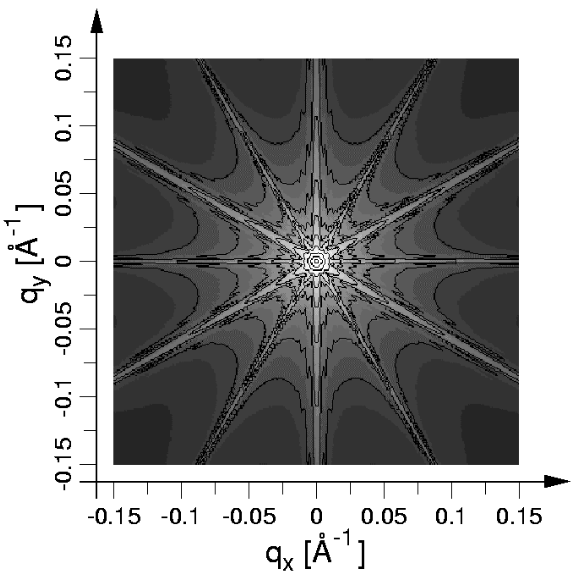

Figure 5a
Calculated scattering pattern from triangular islands for transmission small-angle scattering ($q_z = 0$) exhibiting a 6-fold symmetry.

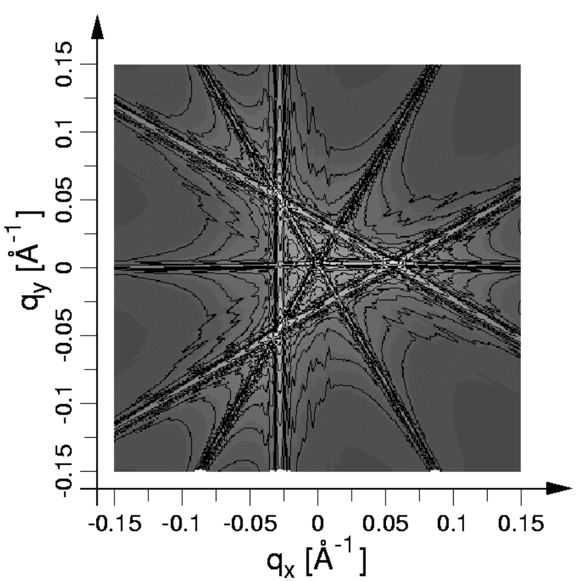

Figure 5b
GISAXS pattern ($q_z = 0.1025Å^{-1}$) from the same triangular islands showing 3-fold symmetry.

In summary, we have shown that triangular pyramidal Ge islands on boron-terminated Si(111) grow partially ordered. The degree and direction of ordering were determined using atomic force microscopy, and we have shown that the ordering features are caused by the miscut induced terraces of the substrate. The underlying growth mechanism opens the possibility to align islands using designed terraces to template the substrate surface. We have also shown that grazing-incidence small-angle X-ray scattering from these islands is composed of four different terms, as described by the distorted-wave Born approximation and can be used to determine their shape, size and full triangular symmetry.

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References

Cowley, J.M. (1975), *Diffraction Physics*, North-Holland, Amsterdam.
Kegel, I., Metzger, T.H., Fratzl, P., Peisl, J., Lorke, A., Garcia, J.M. & Petroff, P.M. (1998). *Europhys. Lett.* **45**, 222-227.
Köhler, U., Jusko, O., Pietsch, G., Müller, B. & Henzler, M. (1991). *Surf. Sci.* **248,** 1321-1329.
Kovats, Z. (1999), private communication.
Legoues, F.K., Reuter, M.C., Hammar, M., Tersoff. J. & Tromp, R.M. (1994). *Phys. Rev. Lett*. **73**, 300-303.
Leonard, D., Krishnamurthy, M., Reaves, C.M., Denbaars, S. & Petroff, *P.M. (1993) Appl. Phys. Lett*. **63**, 3203-3205.
Mo, Y-W., Savage, D.E., Swartzentruber, B.S., & Lagally, M.G. (1990) *Phys. Rev. Lett.* **65**, 1020-1023.
Rasmussen, F. B., Baker, J., Nielsen, M., Feidenhans'l, R. & Johnson, R.L. (1997). *Phys. Rev. Lett.* **79**, 4413-4416.
Rauscher, M., Paniago, R., Metzger, T.H., Kovats, K., Peisl, J., Schulze, J., & Eisele, I. (1999). *Journal of Applied Physics (in press).*
Rupp, T., Messarosch, J., Eisele, I. (1998). *J. Crystal Growth* **183** 99-103.
Steinfort, A. J., Scholte, P. M. L. O., Ettema, A., Tuinstra F., Nielsen, M., Landemark, E., Smilgies, D.-M., Feidenhans'l, R., Falkenberg, G., Seehofer, L. & R. L. Johnson (1996). *Phys. Rev. Lett.* **77**, 2009-2012.
Tersoff, J., Teichert, C. and Lagally, M.G. (1996). *Phys. Rev. Lett*. **76**, 1675.
Voigtländer, B. & Zinner, A. (1993). *Appl. Phys. Lett.* **63**, 3055-3057.
Zotov, A.V., Kulakov, M.A., Ryzhkov, S.V., Lifshits, V.G., Bullemer, B. & Eisele, I. (1996) *Surf. Sci.* **352-354,** 358-363.
Suzuki, M., Kudoh, Y., Homma, Y., & Kaneko, R. (1991). *Appl. Phys. Lett*. **58**, 2225-2228 and references therein.

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