Aspect ratio of liquid phase epitaxial SiGe/Si(001) islands as probed by high resolution x-ray diffraction

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X-ray diffuse scattering is used to probe size, shape, and strain distribution of self-organized SiGe/Si(001) islands, which were grown by liquid phase epitaxy. The SiGe islands show a truncated pyramidal shape with {111} side facets and a (001) top facet and they are highly uniform in size. With an averaged island base width of 130 nm and a corresponding height of 65 nm all the islands have a characteristic geometrical *base-to-height* aspect ratio of about 2. X-ray diffuse scattering is used to locally probe the elastically relaxed regions inside the island apex and the strongly strained regions near the substrate-island interface. It is found that the geometrical aspect ratio has a large impact on the x-ray diffuse intensity pattern in reciprocal space. By performing corresponding kinematical x-ray simulations this fact can be utilized to determine the aspect ratio with high sensitivity. © 2004 American Institute of Physics. [DOI: 10.1063/1.1763994]

I. INTRODUCTION

Owing to their exceptional physical properties low-dimensional semiconductor structures (e.g., quantum dots) have attracted considerable interest. While *top-bottom* approaches to generate quantum dots mostly rely on highly sophisticated masking and etching techniques, both self-formation and spatial orientation during heteroepitaxy can provide an interesting alternative. On the other hand, heteroepitaxial growth exhibits a widespread complexity, whose details are not completely understood yet. Therefore, model systems as, e.g., SiGe/Si(001) have been extensively investigated to attain a more comprehensive knowledge of crystal growth (see, e.g., Refs. 1–5).

Heteroepitaxial growth inherently implies the formation of elastic strain owing to the different lattice parameters of the epitaxial layer and the substrate. In the Stranski-Krastanow mode⁶ the growth initially starts by wetting the substrate with a thin planar layer. This process is accompanied by the formation of strain energy, which increases with enhancing layer thickness. When approaching a critical thickness, which amounts to typically a few monolayers, the formation of three-dimensional island growth becomes energetically favorable since the gain in energy due to elastic strain relaxation is larger than the total amount of surface free energy.

For liquid phase epitaxial (LPE) grown SiGe islands the scaling law $w \propto f^{-2.03}$ has been found⁷ which interrelates the island base width w and the lattice mismatch f between island and the underlying substrate. This scaling behavior addresses strain as the key property which restricts the final

island size, and it represents a very useful tool to adjust the island size by choosing an appropriate atomic composition in layer.

During evolution the island shape and strain distribution inside the islands may alter. This is also significantly affected by the specific growth conditions. Pure germanium islands grown by molecular beam epitaxy (MBE) evolve by hut clusters⁸ via pyramids resulting finally in domes.⁹ While MBE grown SiGe islands exhibit {10 $\bar{m}$ }-type facets,^{8,10,11} SiGe islands generated by LPE consist of truncated pyramids with {11 $\bar{m}$ }-type facets.^{12–14} Thus, they are azimuthally 45° off their MBE counterparts. It is interesting to note that for both individual methods the respective growth sequences do not qualitatively depend on the lattice mismatch at a certain concentration window.^{15,16}

Although providing high resolution in real space direct imaging techniques give information on a restricted number of objects only and thus suffer from low statistics. On the other hand, scattering techniques yield information on the basis of large ensembles and thus represent a statistically relevant measure. Among these techniques x-ray diffuse scattering has turned out to be a very powerful tool to investigate mesoscopic and nanoscale semiconductor structures. This includes the determination of shape, size, chemical composition, strain profile, and positional correlation (e.g., Refs. 16–22). In this work we apply high resolution x-ray diffraction combined with corresponding scattering simulations. These are based on kinematical scattering theory and use the strain field within SiGe/Si(001) nanoscale islands as determined by finite element calculations based on linear elasticity theory.¹⁶

II. SAMPLE

Respective samples were grown by liquid phase epitaxy which operates rather close to thermodynamical equilibrium.

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FIG. 1. Atomic force (a) and a scanning electron micrograph (b) of the sample surface. Typically, the islands consist of truncated pyramids with $\{111\}$ -type side facets and a (001) top facet.

As compared to germanium, silicon is at least two orders of magnitude less soluble within bismuth. Consequently, the bismuth solvent can be saturated with silicon, whereas a germanium mol fraction of 0.0103 within the solution has to be carefully adjusted in order to give 30% germanium in the solid state. To ensure that growth takes place at conditions close to thermodynamical equilibrium a homogenization procedure of several hours at the growth temperature (600 °C) was performed. After an *in situ* desorption of the natural silicon oxide at 930 °C the actual growth temperature was adjusted 2 K below the saturation temperature to initiate the growth process.

Figure 1 represents an atomic force (a) and scanning electron micrograph (b) of the resulting sample surface. Nearly all islands are of uniform shape and size and they consist of truncated pyramids defined by four $\{111\}$ side facets and a (001) top facet. A geometrical aspect ratio $Q = w/h$ of ≈ 2 is found, where $w = 130$ nm denotes the island base width along the $[110]$ direction and $h = 65$ nm is the final island height. This result can be explained by equilibrium shape calculations. Here different ratios Q at fixed $\{111\}$ side and a (001) top facet have been considered and a local minimum in total energy has been found for $Q = 2$ (Ref. 23).

III. EXPERIMENT

In order to determine the island shape and residual strain distribution we have evaluated the diffusely scattered intensity around various reciprocal lattice points. However, we will take particular advantage of asymmetrical reflections, which probe both the horizontal and vertical lattice spacings independently, and thus enable a clear distinction between a

tetragonally distorted lattice and a cubic lattice referring to different areas inside the islands. The scattering experiments were carried out at BW2 station at HASYLAB/DESY applying highly collimated x-rays with an energy of 8 keV and an energy band width of $\Delta E/E = 10^{-4}$. A position sensitive detector (PSD) with an inherent resolution of $80 \mu\text{m}$ was placed 750 mm behind the sample. By utilizing a small x-ray spot at the sample, each channel of the PSD corresponds to a certain scattering angle. Thus, a PSD simultaneously records the diffuse intensity distribution along a (curved) line in reciprocal space, and a 2D reciprocal space map can be recorded by a single rocking scan of the sample. Although the resolution is definitely worse than with a triple crystal arrangement this multidetection setup ensures fast data acquisition at medium resolution of typically $\Delta q = 5 \times 10^{-4} \text{ \AA}^{-1}$ which is sufficient to detect the rather broad diffuse intensity. Perpendicular to the scattering plane a pair of 1 mm slits is used thus integrating the x-ray diffuse intensity in reciprocal space within $\Delta q = 8 \times 10^{-3} \text{ \AA}^{-1}$.

IV. RESULTS AND DISCUSSION

The chemical composition inside the islands, the resulting strain distribution, positional correlation, and the island shape itself contribute in a very complex way to the diffuse scattering, generally preventing a direct access to those parameters. Hence, scattering simulations are an essential part of data evaluation. We have treated the scattering process within the kinematical approximation where the scattered amplitudes sum up coherently without taking into account absorption, extinction, and multiple scattering processes. Consequently, our approach is less useful for treating the scattering from the Si substrate, where multiple scattering processes cannot be neglected. However, kinematical theory is a rather good approximation as long as the structures are comparatively strongly distorted and restricted in size. This holds for the small and strongly strained islands as discussed in this work. For more details concerning the simulations see Ref. 14.

Despite the limits of kinematical theory, one has to take into account the substrate underneath the island and wetting layer in order to correctly calculate the extended strain field in the surrounding areas of the island. The calculation of the three-dimensional strain field was numerically performed in the framework of the finite element method (FEM), which is based on linear elasticity theory and which takes into account both the three dimensional Ge composition profile and the elastic anisotropy. FEM requires some initial information concerning the island shape and size, which can be provided by scanning probe microscopy, see Fig. 1. Then, throughout subsequent iterations of the simulation procedure the impact of various parameters as the Ge composition profile and the island shape/size to the diffuse scattering can be tested and, if necessary, further improved until satisfactory agreement with experiment is achieved.

Figure 2(a) depicts the diffusely scattered intensity around the asymmetrical 404 reciprocal lattice point. The diffuse intensity from the Si substrate is mainly concentrated in a cloud around the coherent substrate peak (marked as

FIG. 2. Diffusely scattered intensity in the proximity of the asymmetric 404 silicon reciprocal lattice point S. (a) experimental data, (b)–(e) corresponding simulations. The intensity cloud I at $-q_{100}=q_{001}=4.57 \text{ \AA}^{-1}$ is related to the scattering at elastically relaxed parts in the island top, whereas the feature T corresponds to strained parts at the island bottom. A set of parallel fringes F appears tilted 45° off the $[001]$ direction. The simulations assume islands of 130 nm base width but with different heights: (b) 39 nm, (c) 52 nm, (d) 65 nm, and (e) 78 nm.

“S”). It can be well distinguished from the diffuse scattering from the SiGe islands which is distributed around a bright maximum (marked as “T”). This feature can be attributed to the island apex consisting of an elastically relaxed lattice of cubic symmetry.²⁰ Additionally, the comparatively broad and weak tail (T) originates from the strongly strained regions close to the island-substrate interface. Here, the horizontal lattice spacings are close to that of the substrate; however, the lattice is vertically elongated.

While there are $\{111\}$ -type facets present within the $[110]$ zone there are no facets existing in the $[100]$ zone [see Fig. 1(b)]. Nevertheless we find a bundle of parallel fringes (F) in close vicinity of the island peak (I) in Fig. 2(a). They are tilted 45° off the $[001]$ direction. This might be indicative of a mean surface (i.e., facet) of the type $\{101\}$. A detailed inspection of the scanning electron micrograph in Fig. 1(b) supposes somewhat coarsened $\langle 101 \rangle$ island edges. At comparable islands containing 10% germanium and with a base width of around $w=1500 \text{ nm}$ the $\langle 101 \rangle$ edges are found to consist of small $\{111\}$ facets locally forming a $\{101\}$ mean surface, see Fig. 4 in Ref. 16. However, as we will demonstrate in this work, the observed fringes are *not created* by $\{101\}$ surfaces. They are, instead, basically caused by the strain distribution inside the island, which is, nevertheless, sensitive to the island shape.

Figures 2(b)–2(e) show results of simulations of x-ray diffuse intensity patterns that can be compared to the experimental results displayed in Fig. 2(a). The FEM strain calculations were used as input for respective x-ray simulations. In all simulations the islands are composed of four $\{111\}$ sides and a (001) top facet. The islands have been placed on a silicon substrate with lateral dimensions of twice the island base. While the island height changes from $h=39 \text{ nm}$ (b) via $h=52 \text{ nm}$ (c) and $h=65 \text{ nm}$ (d) finally up to $h=78 \text{ nm}$ (e) the corresponding lateral width of $w=130 \text{ nm}$ at the island bottom was kept fixed, thus resulting in different aspect ratios Q (see the schematic insets). According to previous investigations a steplike vertical germanium profile inside the islands has been assumed.¹⁶ In this case, the composition changes from $\text{Si}_{0.75}\text{Ge}_{0.25}$ at the island bottom towards $\text{Si}_{0.7}\text{Ge}_{0.3}$ at $h=22 \text{ nm}$ (b-d) and 26 nm (e). However, the interface width is defined within an accuracy of about 4 nm accounting for a

calculation grid with a lattice parameter four times larger than that of silicon and considering the interface by at least one node on both sides.

Since scattering from the silicon substrate has been completely omitted in the simulations there is no contribution of scattering from a pure silicon lattice. However, the simulations prove a significant impact of the island aspect ratio Q on the diffuse intensity distribution: Most obviously, the inclination angle of the fringes with respect to the $[001]$ direction monotonically decreases from about 60° (b) via 54° (c) to 45° (d) meeting the experimental value. When the island height approaches 78 nm (e) the fringe pattern is even partly washed out. Although all angles defined by adjacent facets are kept fixed for different aspect ratios Q , it is generally not possible to address this observation exclusively to a shape phenomenon since an altered aspect ratio correspondingly modifies the strain field inside the island. In fact, the intensity distribution as shown in Fig. 2 is determined by a complex interplay of both the island shape and size and the strain distribution inside the island. On the other hand feature (I) appears more and more pronounced on decreasing the aspect ratio Q . This clearly proves that islands with small aspect ratio Q —and therein regions close to the apex—can relax more efficiently than shallow islands. For the simulations (d, e) using $h=65 \text{ nm}$ and 78 nm the intense spot I is in good agreement with experiment indicating a cubic, nearly perfectly relaxed lattice at the island top, which is also evident in the FEM strain calculations (not shown here). Consequently, an increasing island height does not further modify feature I. Finally, the tail-like diffuse intensity pattern (T), which horizontally, appears close to the substrate position, indicates a tetragonally distorted lattice close to the substrate-island interface. Thus, its particular shape and position in reciprocal space does not depend on the island aspect ratio Q . The simulation (d)—referring to an island height of 65 nm and corresponding aspect ratio of $Q=2$ —clearly gives the best agreement with the experimental data. This result is confirmed by scanning electron micrographs, see Fig. 1(b). However, it provides a statistically relevant quantity since the x-ray scattering experiment averages over an ensemble of $\approx 3 \times 10^7$ islands taking an island density of $3 \times 10^9 \text{ cm}^{-2}$ and an illuminated sample area of 1 mm^2 into account.

V. CONCLUSION

In conclusion, we investigated LPE grown $\text{Si}_{0.7}\text{Ge}_{0.3}/\text{Si}(001)$ nanoscale islands. Scanning probe micrographs indicate islands of uniform shape forming truncated pyramids with $\{111\}$ side facets and a (001) top facet. The island ensemble exhibits a very narrow size distribution with an averaged island base width of $w=130$ nm and an island height of $h=65$ nm, resulting in a geometrical aspect ratio base vs height of 2. The SiGe islands were investigated by x-ray diffuse scattering. Experimental diffuse intensity distributions were compared to corresponding kinematical scattering simulations. These simulations use the strain field as calculated by the finite element method. The diffuse scattered intensity in the vicinity of the asymmetrical Si 404 reflection indicates a fully relaxed lattice in the island top. The scattering simulations show that the island aspect ratio significantly influences the scattering pattern. This, finally, enabled us to determine very accurately the island shape.

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¹P. Sutter and M. G. Lagally, Phys. Rev. Lett. **84**, 4637 (2000).

²V. B. Shenoy, C. V. Ciobanu, and L. B. Freund, Appl. Phys. Lett. **81**, 304

(2002).

³P. Sutter, P. Zahl, and E. Sutter, Appl. Phys. Lett. **82**, 3454 (2003).

⁴J. Tersoff, B. Spencer A. Rastelli, and H. von Känel, Phys. Rev. Lett. **89**, 196104 (2002).

⁵R. Tromp, F. M. Ross, and M. Reuter Phys. Rev. Lett. **84**, 4641 (2000).

⁶I. N. Stranski and L. Krastanow, Sitzungsberichte d. Akademie d. Wissenschaften in Wien, Abt. IIb 146, 797 (1937).

⁷W. Dorsch, H. P. Strunk, H. Wawra, G. Wagner, J. Groenen, and R. Carles, Appl. Phys. Lett. **72**, 179 (1998).

⁸Y.-W. Mo., D. E. Savage, B. S. Swartzentruber, and M. G. Lagally, Phys. Rev. Lett. **65**, 1020 (1990).

⁹F. M. Ross, M. C. Reuter, and R. M. Tromp, Science **286**, 193 (1999).

¹⁰M. Kästner and B. Voigtländer, Phys. Rev. Lett. **82**, 2745 (1999).

¹¹A. Rastelli, H. von Känel, B. J. Spencer, and J. Tersoff, Phys. Rev. B **68**, 115301 (2003).

¹²J. Groenen, R. Carles, S. Christiansen, M. Albrecht, W. Dorsch, H. P. Strunk, H. Wawra, and G. Wagner, Appl. Phys. Lett. **71**, 3856 (1997).

¹³M. Schmidbauer, T. Wiebach, H. Raidt, M. Hanke, R. Köhler, and H. Wawra, Phys. Rev. B **58**, 10523 (1998).

¹⁴M. Schmidbauer, M. Hanke, and R. Köhler, Cryst. Res. Technol. **37**, 3 (2002).

¹⁵J. Florio, G. Lucadamo, E. Chason, L. Freund, M. Sinclair, R. Twisten, and R. Hwang, Phys. Rev. Lett. **80**, 4717 (1998).

¹⁶M. Hanke, M. Schmidbauer, D. Grigoriev, P. Schäfer, R. Köhler, A.-K. Gerlitzke, and H. Wawra, Phys. Rev. B **69**, 075317 (2004).

¹⁷R. Lechner, T. Schüllli, V. Holý, G. Springholz, J. Stangl, A. Raab, G. Bauer, and T. Metzger, Appl. Phys. Lett. **84**, 885 (2004).

¹⁸T. Schüllli, J. Stangl, Z. Zhong, R. Lechner, M. Sztucki, T. Metzger, and G. Bauer, Phys. Rev. Lett. **90**, 066105 (2003).

¹⁹I. Kegel, T. H. Metzger, A. Lorke, J. Peisl, J. Stangl, G. Bauer, J. M. Garcia, and P. M. Petroff, Phys. Rev. Lett. **85**, 1694 (2000).

²⁰T. Wiebach, M. Schmidbauer, M. Hanke, H. Raidt, R. Köhler, and H. Wawra, Phys. Rev. B **61**, 5571 (2000).

²¹J. Stangl *et al.*, Appl. Phys. Lett. **79**, 1474 (2001).

²²M. Schmidbauer, *X-Ray Diffuse Scattering From Self-Organized Mesoscopic Semiconductor Systems, Springer Tracts in Modern Physics*, Vol. 199 (Springer, Berlin, 2004).

²³M. Hanke, M. Schmidbauer, R. Köhler, F. Syrowatka, A. K. Gerlitzke, and T. Boeck, Appl. Phys. Lett. **84**, 5228 (2004).