Direct Determination of Strain and Composition Profiles in SiGe Islands by Anomalous X-Ray Diffraction at High Momentum Transfer

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Anomalous x-ray scattering is employed for quantitative measurements of the Ge composition profile in islands on Si(001). The anomalous effect in SiGe is enhanced exploiting the dependence of the complex atomic form factors on the momentum transfer. Comparing the intensity ratios for x-ray energies below and close to the K edge of Ge at various Bragg reflections in the grazing incidence diffraction setup, the sensitivity for the Ge profile is considerably enhanced. The method is demonstrated for SiGe dome-shaped islands grown on Si(001). It is found that the composition inside the island changes rather abruptly, whereas the lattice parameter relaxes continuously.

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The self-organized growth of semiconductor quantum dots has attracted considerable interest, aiming both on fundamental investigations of their structural, electronic, and optical properties, as well as on their potential for electronic and optoelectronic devices. Quite recently, quantum dots have also been suggested for the implementation of solid state quantum computing and information storage [1]. In the Stranski-Krastanow growth of such quantum dots a fundamental morphological instability in the heteroepitaxy of semiconductors with a certain lattice mismatch is exploited, which is driven by the elastic relaxation of strain energy at the expense of an increase of surface energy. Apart from the growth of InAs islands on GaAs, Ge islands on Si have been the subject of numerous studies. Such islands show interesting morphological shape transitions as a function of growth parameters [2]. Despite the fact that they consist of semiconductors with an indirect energy gap, electroluminescence even at room temperature has been achieved from buried Ge rich islands in Si at about a wavelength of 1.3 μm [3]. The recombination energy of the electron-hole pairs depends crucially on the size, the shape, the strain status, and the chemical composition of the islands.

From a series of studies, including, e.g., transmission electron microscopy (TEM) investigations, using energy selective imaging or digital image analysis, it is known that intermixing in such islands exists, but difficulties in the quantification of the composition profile along the growth direction remain [4–6]. This profile, together with the strain distribution is crucial for the electronic band structure of the embedded islands and thus for any applications. In addition to TEM, x-ray studies on strain and composition profiles have been performed. For uncapped InAs islands the desired chemical information was obtained by a direct method based on grazing incidence diffraction, making use of the difference between strong [like the (400)] and weak [like the (200)] superstructure reflections in these materials, which crystallize in the zinc-blende structure [7–9]. For Ge islands on Si, which both crystallize in the diamond lattice, this method fails. Thus other approaches for Ge islands in a Si matrix were used. These are based on the measurement and simulation of reciprocal space maps, using a fitting procedure and model assumptions for chemical composition profiles in the islands [10,11] and corresponding finite element or analytical calculations for the strains. However, as shown recently, for capped islands the scattered intensity distribution is mainly sensitive to the average Ge content, and thus quantitative information on chemical composition profiles for buried islands is rather difficult to obtain [12].

In this Letter we show that anomalous x-ray diffraction can indeed be used to obtain the necessary chemical sensitivity for a *direct* determination of composition profiles in SiGe islands. It is a well established technique to tune the x-ray energy close to an absorption edge to enhance the sensitivity for a particular element [13]. In the SiGe system, the only edge which allows for an x-ray wavelength that can be used for diffraction is the Ge K edge. In general, the energy and momentum dependency of the atomic scattering factor is expressed by

$$f(Q, E) = f_0(Q) + f'(E) + if''(E) \qquad (1)$$

with $f'(E)$ and $f''(E)$ being the real and imaginary parts of the energy dependent correction that become important close to an absorption edge. The Q dependency of the form factor originates from the non-negligible spatial distribution of the electrons of an atom, as it represents the Fourier transform of the electron distribution. Therefore f_0 equals the total number of electrons Z for forward scattering, but decreases for higher momentum transfer Q. This decrease is generally neglected in the correction terms f' and f'', based on the argument that the inner electron shells that are responsible for these

terms are very localized in space. Theoretical and experimental studies on the validity of this approximation justified its use for all momentum transfers accessible at energies in the vicinity of the K-absorption edge [14,15]. Thus the anomalous scattering effects can be enhanced considerably by measuring Bragg reflections for high momentum transfers and hence improve the sensitivity for the composition in the SiGe system. $f'$and f'' are presented in Fig. 1 as a function of energy. The two probed energies $E_1 = 11\,043$ eV and $E_2 = 11\,103$ eV are indicated. For a precise determination of the atomic scattering factors, f''_{Ge} was measured via fluorescence and f'_{Ge} was calculated using the Kramers-Kronig dispersion relation for the measured f''_{Ge} values in the vicinity of the edge and tabulated values from [16] for the remaining part of the spectrum. The momentum dependent behavior of $f_0(Q)$ of Ge and Si was calculated with a code based on the parameters derived in [17]. Figure 2(a) shows the momentum dependence of the real parts of the scattering factors for the two energies for Ge and Si. At these x-ray energies, the corrections f' and f'' can be neglected for Si. It is clear that for high Q the Si scattering becomes more pronounced with respect to the Ge scattering.

For low momentum transfers at the (220) reflection, where the Q correction in $f_0(Q)$ is small, the anomalous effect changes the scattering factor of pure Ge by a factor of 0.77, causing a ratio between the diffracted intensities at both energies of 1.7. A compositional change from pure Ge to Si_xGe_{1-x} would result in a deviation from this ratio. As the scattering of Ge is still fairly large compared to that of Si at the (220) Q value, this deviation is very small and allows only a very crude determination of the composition. As $f_0(Q)$ decays for higher Q values, the ratio in the scattered intensity from pure Ge between E_1 and E_2 increases considerably to, e.g., 4.7, at the (800) reflection as illustrated in Fig. 2(b). The deviation from this ratio for a $Si_{0.3}Ge_{0.7}$ alloy amounts to 22% at the (800) reflection, whereas it causes a change of only 9% at the (220) reflection. Therefore the method of measuring reflections of large Q values yields a higher compositional sensitivity.

In addition, the high indexed reflections allow for a better discrimination of strain and shape. The broadening of a Bragg reflection due to the finite size R amounts to $\frac{2\pi}{R}$ and remains constant in reciprocal space, whereas the strain-induced broadening $\frac{\Delta a}{a}$ increases with Q. Therefore, if the condition $\frac{2\pi}{R} < \frac{\Delta a}{a} Q$ is fulfilled we can extract directly the composition for different lattice parameters with the resolution Δa [7]. To discriminate between two regions with a difference of, e.g., 0.2% in a lattice parameter would require a size R of about 300 Å of the diffracting object when probing the (800) reflection. Evaluating the intensity ratio for the two energies E_1 and E_2 at a given momentum transfer Q_{rad}, one can now use this direct method on a high indexed reflection to link composition and lattice parameter without any model assumptions. As the scattering amplitude in Si is virtually the same for both energies, no elaborate normalization of the intensities is necessary. Subsequently, this compositional profile can be used to simulate the diffracted intensity in the vicinity of Bragg reflections. This allows a discrimination of strain, shape, and composition as the most important parameters influencing the band structure of nanostructures. As a first application of the method, a SiGe island sample grown by molecular beam epitaxy on Si(001) has been investigated. At a substrate temperature of 600 °C seven monolayers of Ge have been deposited, leading to islands with a height of

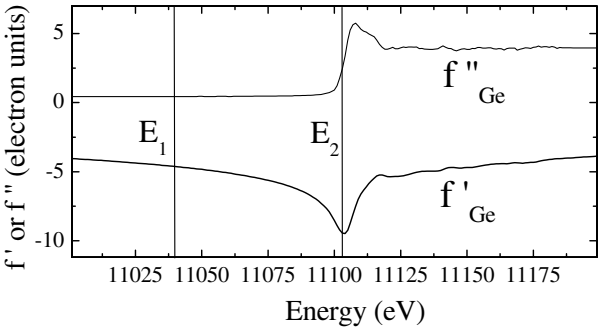

FIG. 1. Anomalous corrections f' and f'' for Ge in the vicinity of the K-absorption edge. The two perpendicular lines mark the energies $E_1 = 11\,043$ eV and $E_2 = 11\,103$ eV.

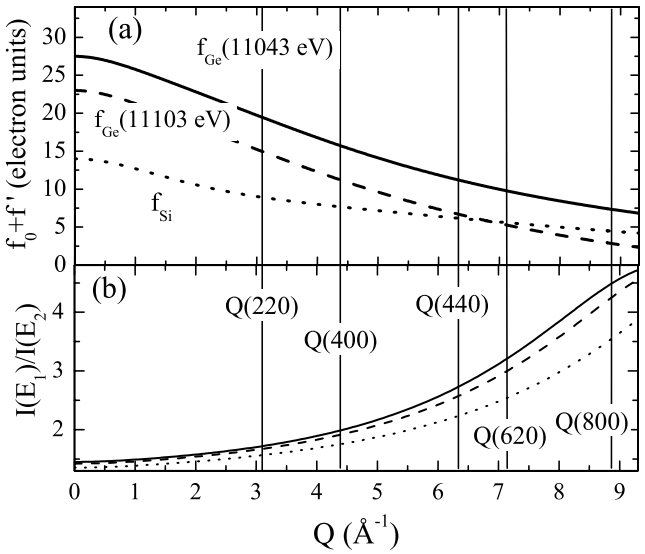

FIG. 2. (a) Momentum dependent real part of the scattering factor for Ge at $E_1 = 11\,043$ eV (solid line), Ge at $E_2 = 11\,103$ eV (dashed line), and Si (dotted line). (b) Theoretical momentum dependence of the ratio of the diffracted intensities at $E_1 = 11\,043$ eV and $E_2 = 11\,103$ eV for pure Ge (solid line), $Si_{0.1}Ge_{0.9}$ (dashed line), and $Si_{0.3}Ge_{0.7}$ (dotted line). Perpendicular lines mark the positions of the probed in-plane reflections.

about 140 Å and a width at the base of about 700 Å. Atomic force microscopy (AFM) reveals a round shape of the islands and a very narrow size distribution of less than 10% half-width in the island heights $\Delta h/h$. The anomalous diffraction experiment was performed at the beamline ID01 at the European Synchrotron Radiation Facility in Grenoble, France. To suppress the thermal diffuse scattering from the substrate, the grazing incidence diffraction method was chosen. For both energies, the incident and exit angles of the beam were kept constant at 0.1°, well below the critical angle α_c so that the change in the transmission function of Si for the small energy change could be neglected. Figures 3(b)–3(f) shows radial scans for the five in-plane reflections (220), (400), (440), (620), and (800), as illustrated in Fig. 3(a). Q_{rad} describes the momentum transfer along the reciprocal lattice vector. The strong enhancement of the anomalous effect is visible. In particular, in Fig. 3(f), the steep decay of the intensity for 11 103 eV at about 9.2 $\mathrm{Å}^{-1}$ is a clear sign of a rather abrupt change of the Ge content in the dots which could not be resolved at low $(hk0)$ reflections. From the intensity ratios $\frac{I(E_1)}{I(E_2)}$ of the most sensitive reflections (620) and (800) [full lines in Figs. 3(e) and 3(f)] the composition for a certain lattice parameter is directly extracted. The evaluated composition is plotted as a function of the local in-plane lattice parameter in Fig. 3(g). A region with vanishing Ge content exhibiting tensile strain with respect to the substrate is clearly visible. This can be attributed to the strained region in the Si substrate below the Ge island, as sketched in Fig. 5 below. With the composition values as extracted from the high indexed (800) and (620) reflections we obtain a direct link between lattice parameter and composition. In a second step, we recorded line scans at a fixed energy of 11 043 eV perpendicular to the radial scan direction. These angular scans are sensitive to the size of a region with fixed lattice parameter, chosen by the position Q_{rad} [7]. For the (400) reflection, the angular scans are shown in Fig. 4(b), together with their radial positions as indicated in Fig. 4(a). From the form factor oscillations we

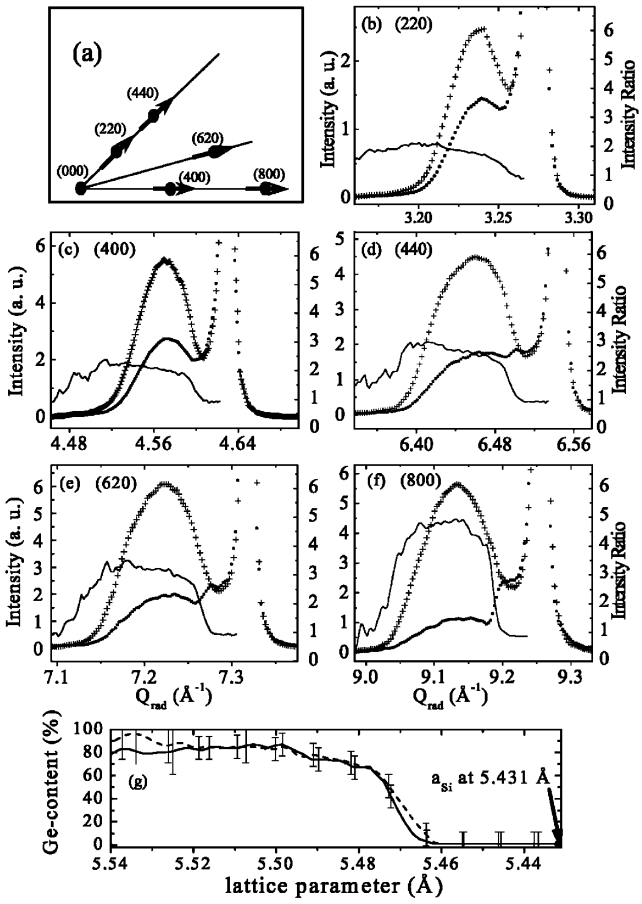

FIG. 3. (a) Sketch of the (001) plane with the probed in-plane reflections. The arrows indicate that all scans were performed in a radial direction. (b)–(f) Reflections (220), (400), (440), (620), and (800) for the Ge islands recorded at 11 043 eV (crosses) and 11 103 eV (full dots). In addition, the ratio between the intensities for the two energies is plotted (full line). The Bragg peak from the Si substrate to the right of the island peak is not shown to the full intensity. (g) Evaluation of the Ge content from the reflections (800) (full line) and the (620) (dashed line).

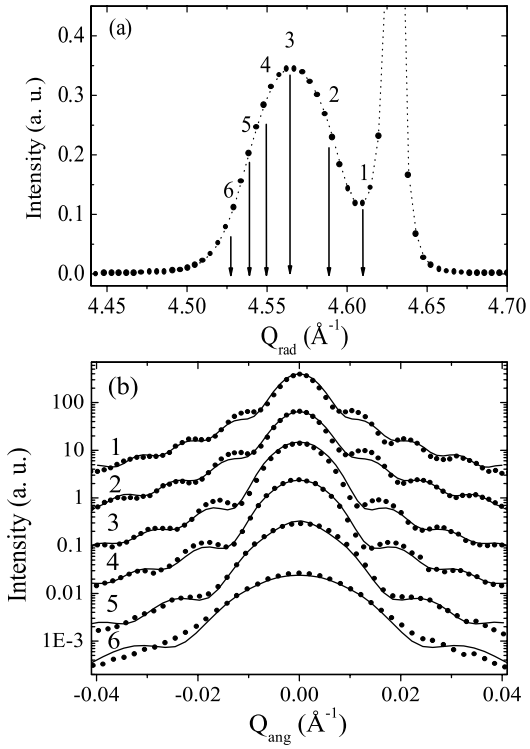

FIG. 4. (a) Radial scan close to the (400) reflection at 11 043 eV. At the numbered positions, scans in the perpendicular ⟨010⟩ direction were performed as plotted below. (b) Angular scans and fit curves for six positions as indicated above. The scans are shifted with respect to each other for clarity.

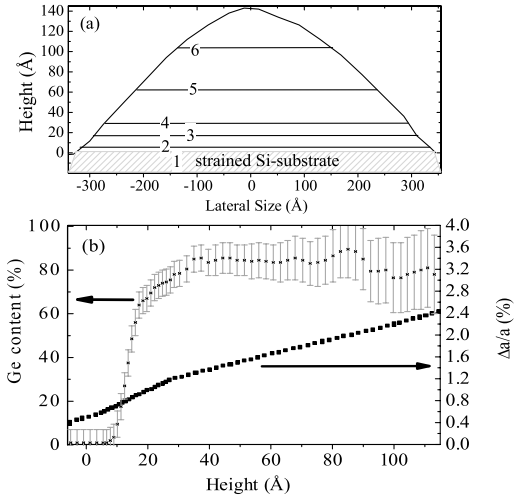

FIG. 5. (a) Cross section profile of one dot extracted from the AFM data. The lateral size as determined from the scans in Fig. 4(b) can be attributed to a certain height (see numbers). (b) The relaxation (black squares) $\frac{\Delta a}{a}$ and the Ge content (plot with error bars) are plotted over the height z.

determine the lateral size of a region with a common lattice parameter. Using the cross-section profile of the islands from AFM as shown in Fig. 5(a), the lateral size as derived from the angular scans can be associated with a certain height within the islands, linking the height h with the in-plane momentum transfer Q_{rad}. Combining all results we can finally reconstruct the Ge concentration as a function of the height in the island. The result is shown in Fig. 5(b). In addition, the evolution of the lattice parameter relaxation is plotted. The figure clearly shows a rather abrupt interface between Si and the SiGe island, with an almost constant Ge content of roughly 80% above a height of 20 Å. With respect to the Si, the in-plane lattice parameter relaxes from 0.4% at the bottom of the island to about 2.4% at the top (note that strain free Ge would require a relaxation of 4%). The error bar increases with height, as the amount of material and hence the scattered intensity decreases. Additionally, the form factor is spread out more and more in reciprocal space, further decreasing the resolution. Because of the low intensity from the islands, Laue and thermal diffuse scattering contributions to the diffuse intensity cannot be neglected any more. In conclusion, we have shown that exploiting the dependence of the scattering factor on the momentum transfer together with anomalous scattering allows for the determination and discrimination of composition and strain profiles in semiconductor nanostructures. Strain gradients are directly obtained from the measurements without the necessity of model calculations. It thus allows the determination of the residual elastic energy and opens up the opportunity to determine the interdiffusion behavior for different growth conditions which sensitively influence the island formation.

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Note added.—After submission of our manuscript a paper by R. Magalhaes-Paniago *et al.* was published [18], in which anomalous x-ray scattering was used in order to determine composition profiles in Ge islands, too. However, as we show in our work, it is essential for the required sensitivity of Ge content profiling to exploit the strong dependence of the atomic form factor f_0 on the magnitude of the momentum transfer in the diffraction experiment, as shown in Fig. 2.

[1] P. M. Petroff, A. Lorke, and A. Imamoglu, Phys. Today **54**, No. 5, 46 (2001).
[2] G. Medeiros-Ribero *et al.*, Science **279**, 353 (1998).
[3] L. Vescan *et al.*, J. Appl. Phys. **87**, 7275 (2000).
[4] S. A. Chaparro *et al.*, Phys. Rev. Lett. **83**, 1199 (1999).
[5] Chuan-Pu Liu, J. M. Gibson, D. G. Cahill, T. I. Kamins, D. P. Basile, and R. S. Williams, Phys. Rev. Lett. **84**, 1958 (2000).
[6] T. Walther, A. G. Cullis, D. J. Norris, and M. Hopkinson, Phys. Rev. Lett. **86**, 2381 (2001).
[7] I. Kegel *et al.*, Phys. Rev. Lett. **85**, 1694 (2000).
[8] T. Schülli *et al.*, Appl. Phys. Lett. **81**, 448 (2002).
[9] In principle, coherent x-ray diffraction may be used to map strain fields in nanocrystals: I. K. Robinson *et al.*, Phys. Rev. Lett. **87**, 195505 (2001).
[10] A. Rastelli, M. Kummer, and H. von Kanel, Phys. Rev. Lett. **87**, 256101 (2001).
[11] Th. Wiebach *et al.*, Phys. Rev. B **61**, 5571 (2000).
[12] A. Hesse *et al.*, Phys. Rev. B **66**, 085321 (2002).
[13] G. Materlik, C. J. Sparks, and K. Fisher, *Resonant Anomalous X-Ray Scattering: Theory and Applications* (North-Holland, Amsterdam, 1994).
[14] P. M. Bergstrom *et al.*, Acta Crystallogr. Sect. A **53**, 7 (1997).
[15] T. Fukamachi *et al.*, Acta Crystallogr. Sect. A **49**, 573 (1993).
[16] L. Kissel *et al.*, Acta Crystallogr. Sect. A **51**, 271 (1995).
[17] J. Baró, M. Roteta, J. M. Fernández-Varea, and F. Salvat, Radiat. Phys. Chem. **44**, 531 (1994).
[18] R. Magalhaes-Paniago, G. Medeiros-Ribeiro, A. Malachias, S. Kycia, T. I. Kamins, and R. Stan Williams, Phys. Rev. B **66**, 245312 (2002).