Strain status of self-assembled InAs quantum dots

K. Zhang,^{a)} Ch. Heyn, and W. Hansen

Institute for Applied Physics, University of Hamburg, Jungiusstraße 11, D-20355 Hamburg, Germany

Th. Schmidt and J. Falta

Institute for Solid State Physics, University of Bremen, Postfach 330440, D-28334 Bremen, Germany

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Grazing incidence x-ray diffraction experiments employing the asymmetric (202) Bragg diffraction have been performed to characterize self-assembled InAs quantum dots grown by molecular-beam epitaxy. We find that the strain is elastically relaxed with different components. The volume distribution of partially strained InAs inside islands is peaked at intermediate strain values. The fraction of both almost fully strained and totally relaxed InAs is found to be small. In addition, a small volume fraction of relaxed $\text{In}_x\text{Ga}_{1-x}\text{As}$ is found. © 2000 American Institute of Physics. [S0003-6951(00)02335-4]

Heteroepitaxy of materials with a large lattice mismatch to the substrate as, e.g., InAs on GaAs(001) surfaces, leads to islanding of the film. Structural studies report that InAs islands grown by molecular-beam epitaxy (MBE) in the so-called Stranski–Krastanov growth mode are coherently strained, dislocation free, and have diameters of a few 10 nm. In particular, the surprisingly narrow distribution that was found for the island diameter made the Stranski–Krastanov InAs islands a very intriguing system with possible applications for optoelectronic devices as well as for fundamental studies of the carrier systems confined in the islands.¹ The small size and strong structural confinement leads to atomic-like, zero-dimensional electronic states in the InAs, and hence, the InAs islands are often called quantum dots (QDs). A few years ago, experimental evidence for the existence of zero-dimensional electron states in self-assembled InAs QDs was obtained with capacitance^{2,3} and Fourier infrared spectroscopy.^{3,4} Since then, InAs QDs have been studied very intensively by many different techniques.^{5–11} It is presently thought that the island formation is entirely driven by the strain originating from the lattice mismatch between InAs and GaAs.¹² Moreover, the electronic states are strongly dependent on the strain status of the dots.⁵ On the other hand, so far only very few experiments exist that yield quantitative information on the strain of the dots.^{5,8,12} From conventional high-resolution x-ray diffraction of single or multiple InAs QD layers, an average strain value has been deduced.^{5,12} However, from the conventional diffraction experiments, it is very difficult to obtain detailed information on the strain distribution inside the InAs QDs due to intensity problems.

In this letter, we address the strain status of the material inside the InAs QDs. We report grazing-incidence x-ray diffraction (GIXRD) experiments on uncapped InAs QD single-layer samples. The grazing-incidence technique strongly enhances the intensity of the InAs QDs in comparison to conventional x-ray techniques. In combination with high-flux synchrotron radiation, this allows us to map out the diffraction intensity in reciprocal space in the vicinity of the

GaAs(202) Bragg diffraction spot. Previously, Kegel *et al.*⁸ used this technique at the in-plane (220) Bragg diffraction spot and analyzed their results using a so-called isostrain model in order to determine a strain distribution within the dots. In comparison, our scattering geometry has several advantages. In our case, the diffraction intensity of the InAs islands is well separated from the diffuse intensity of the GaAs(202) substrate Bragg peak. Furthermore, we are able to distinguish the diffraction intensity between the strained pure InAs and intermixed $\text{In}_x\text{Ga}_{1-x}\text{As}$, which cannot be resolved by GIXRD in the in-plane diffraction geometry. Thus, we can determine the role of intermixed $\text{In}_x\text{Ga}_{1-x}\text{As}$. Recently, it was reported from a scanning tunneling microscopy study that the intermixing inside the InAs QDs at common growth temperatures, i.e., $420^\circ\text{C} \leq T \leq 500^\circ\text{C}$, is strong.¹³

Our samples have been prepared by MBE in an UHV system. The InAs QD growth procedure is the same as published previously.¹¹ Here, we chose the growth temperature of 450°C within the temperature range mentioned above.¹³ The InAs deposit corresponds to a coverage of 2.3 ML. The critical thickness for dot formation is found to occur at about 1.5 ML, as monitored by reflection high-energy electron diffraction.

GIXRD measurements have been performed at the undulator beamline BW1 of the Hamburger synchrotron radiation laboratory (HASYLAB). The wavelength was chosen to be $1.17 \text{ \AA}$. The incidence angle was set to 0.15° , which is smaller than the total external reflection angle of 0.23° . The sample area illuminated by the incoming beam was $16 \times 2 \text{ mm}^2$, providing sufficient statistics on a large number of dots. A position-sensitive detector was mounted parallel to the sample surface with a lateral resolution in reciprocal space of $2.1 \times 10^{-3} \text{ \AA}^{-1}$. Scans of the vertical scattering vector q_z near the GaAs(202) Bragg reflection have been performed in a range between $q_z = 1.70$ and 2.0 reciprocal lattice units (rlu). From these data a reciprocal space map was composed showing the scattering intensity as a function of q_x and q_z . A zoom of the map is presented in Fig. 1, where the scattering intensity is displayed in gray scale. Here, $(\Delta q_x, \Delta q_z) = (0, 0)$ rlu corresponds to the GaAs(202) Bragg spot. A broad diffraction spot in the center of Fig. 1 is

^{a)}Electronic mail: kzhang@physnet.uni-hamburg.de

FIG. 1. Reciprocal space Δq_x - Δq_z map of GIXRD (202) Bragg diffraction for InAs QDs on GaAs substrate (001). The black dot is the expected diffraction position for bulk-like InAs (totally relaxed InAs). $G(100) = 2\pi/a_o^{\text{GaAs}}$, where $a_o = 5.65 \text{ \AA}$ is the GaAs lattice constant. Data are cut at $\Delta q_x = 0$ due to high diffraction intensity from the (202) crystal truncation rod.

observed. The black dot in Fig. 1 indicates the diffraction point expected for totally relaxed InAs, i.e., $(\Delta q_x, \Delta q_z) = (-0.135, -0.135) \text{ rlu}$. One can clearly see that most of the diffraction intensity is continuously distributed between this point and the position of $(0, -0.263) \text{ rlu}$ at which the diffraction intensity of the fully strained InAs matching to the GaAs substrate would be expected. The maximum intensity is located half way in between these two points. The observation of the broad scattering peak in reciprocal space clearly points to the presence of InAs QDs with a partial strain relaxation inside. The broadening of the spot in q_x and q_z orientation can be explained by the finite size of the volume elements at given strain states.

From the position distribution of intensity maxima in reciprocal space we are able to deduce the strain state and composition inside the dots. The wetting layer is not accessible to x-ray diffraction. First, it expands only over a few atomic layers leading to very broad features in reciprocal space, i.e., in q_z . Second, the wetting layer distributed in the regime between two QDs is likely fully oxidized as it is uncapped. In Fig. 2 we plot the intensity maxima positions (full circles) as determined from Δq_z cuts of the data in Fig. 1. The cuts were taken along Δq_z direction in order to account for the aspect ratio of the InAs QDs and avoid strong finite-size effects. The distribution of the peak positions (full circles) has a linear relation in Δq_x - Δq_z space, which is in good correspondence to the elastic strain function represented by the solid line, i.e., $\Delta q_z = \beta - \alpha \cdot \Delta q_x$, where $\beta = -0.263$ and $\alpha = 0.945$ as calculated by elastic theory for the strained bulk-like InAs.¹⁴ The present position distribution confirms that this broad scattering peak in Fig. 1 is contributed entirely by the strained InAs components inside QDs.

The scattering intensities at both extreme positions of totally relaxed InAs as well as InAs almost fully matched to GaAs are very small. In particular, the almost complete absence of fully strained InAs [i.e., at the position of $(\Delta q_x, \Delta q_z) = (0, -0.263)$ in the reciprocal space] is surpris-

FIG. 2. Position distribution of GIXRD intensity maxima in Δq_x - Δq_z space. Full circles denote the strained InAs and open circles represent the intermixed $\text{In}_x\text{Ga}_{1-x}\text{As}$ alloy. The inset is an example of the distinction of GIXRD intensities between the strained InAs (a) and intermixed $\text{In}_x\text{Ga}_{1-x}\text{As}$ (b) with the same Δq_x range, (a) $\Delta q_z = -0.21 \text{ rlu}$ and (b) $\Delta q_z = -0.06 \text{ rlu}$. $G(100) = 2\pi/a_o^{\text{GaAs}}$.

ing in view of the matching between the InAs dots and the GaAs substrate. This puzzle may possibly be resolved by the existence of intermixed $\text{In}_x\text{Ga}_{1-x}\text{As}$ at the interface. This, however, is not directly accessible in the experiment due to the extremely thin wetting layer.

Beside the intense peak, a small additional one is found close to the GaAs substrate peak, as displayed in the inset of Fig. 2. These scans show the same Δq_x range at different Δq_z values with peaks located at $\Delta q_x = -0.06 \text{ rlu}$. The separation in Δq_z allows us to distinguish between strain and composition effects. The open circles in Fig. 2 denote the peak positions of the diffraction intensity for the intermixed $\text{In}_x\text{Ga}_{1-x}\text{As}$ in the Δq_x - Δq_z plane. The dashed line indicates the positions of $\Delta q_z = \Delta q_x$ as expected for unstrained $\text{In}_x\text{Ga}_{1-x}\text{As}$. The positions of the intensity maxima can be resolved in the range of $(-0.06, -0.06) \text{ rlu} \leq (\Delta q_x, \Delta q_z) \leq (-0.03, -0.03) \text{ rlu}$, corresponding to an In content x in the range of $0.21 \leq x \leq 0.42$. Outside this range the scattering intensity can hardly be measured as the peak merges either into the background or into the diffuse intensity of the GaAs(202) peak.

Hence, the data show a large contribution from partially strained InAs and a small one from unstrained $\text{In}_{1-x}\text{Ga}_x\text{As}$. However, in samples grown at 450°C substrate temperature, no evidence is found for the presence of partially strained $\text{In}_x\text{Ga}_{1-x}\text{As}$, which would result in intensity in between the dashed and the solid line of Fig. 2.

The almost complete absence of fully strained InAs is in contrast to the interpretation inferred from GIXRD data taken in the vicinity of the in-plane GaAs(220) Bragg spot.⁸ However, in that experiment overlap of the InAs diffraction intensity with the strong (220) GaAs Bragg spot renders a determination of the intensity behavior for $\Delta q_x, \Delta q_y \rightarrow 0$ impossible.

In order to derive the volume fraction between the $\text{In}_x\text{Ga}_{1-x}\text{As}$ alloy and the strained InAs structure inside dots,

FIG. 3. Normalized volume fraction $f_x \cdot \Delta \epsilon_x$ for the different strain components inside the InAs QDs as determined from the GIXRD peak intensity maxima in Fig. 1. $\epsilon_x = (a^{\text{InAs}} - a_o^{\text{GaAs}}) / a_o^{\text{InAs}}$, where $a_o^{\text{InAs}} = 6.06 \text{ \AA}$ for InAs bulk crystal. The interval value of ϵ_x is $\Delta \epsilon_x = 0.01$.

we integrate the respective diffraction intensities over the measured area in reciprocal space. The volume fraction of relaxed $\text{In}_x\text{Ga}_{1-x}\text{As}$ versus partially strained InAs is directly proportional to the ratio of the corresponding integrated intensities of the diffraction peaks. Using this relation, we obtain a value of 3% for the relative volume fraction of the intermixed $\text{In}_x\text{Ga}_{1-x}\text{As}$. Here, the wetting layer is excluded for reasons explained above. The volume fraction of $\text{In}_x\text{Ga}_{1-x}\text{As}$ is small in comparison to the findings reported previously, however, this discrepancy may be explained by differences in temperature measurements, placing our growth conditions at the low-temperature side in the range of Joyce *et al.*,¹³ who reported substantial Ga intermixing into InAs QDs at higher growth temperatures.

From Fig. 1 we can determine the maximum peak intensities for the strained InAs (full circles in Fig. 2) as a function of the lateral strain component ϵ_x . As the measured peak intensity is proportional to the square of the number of crystal unit cells, we calculate the volume fraction $f_x \cdot \Delta \epsilon_x$ of the different strain components normalized to the InAs QD volume, as shown in Fig. 3. The distribution of these intensity maxima can be described by a Lorentzian function. The maximum value and, hence, the dominant volume fraction of strained InAs, is found at strain values around $\epsilon_x = 3.5\%$ and the volume fraction of both totally relaxed and almost fully strained InAs is quite small. This is in contrast to the assumption of the isostrain model used by Kegel *et al.*⁸ for the interpretation of their data obtained by in-plane x-ray diffraction.

From transmission electron microscopy it is known that QDs with small size, as investigated here, are free of dislocations.¹ Moreover, our data indicate elastic strain relaxation inside the InAs QDs. On the other hand, we only find an almost vanishing fraction of the InAs to show the in-plane GaAs lattice constant, i.e., fully strained InAs. Hence, the

adjustment of lateral lattice parameters between the substrate and the InAs QDs must be accomplished by an interlayer at the interface. This points to an enhanced intermixing of Ga and In near the interface, which also might affect and be triggered by the wetting layer.

In conclusion, we present results of GIXRD experiments in a diffraction geometry that allows the determination of strain and composition inside the InAs QDs. The GIXRD data demonstrate that InAs QDs are elastically relaxed with different strain components. For the InAs QDs, the volume distribution of different strain components can be described by a Lorentzian-like function that is peaked at intermediate strain values around 3.5% and shows that only a small volume fraction of the InAs is fully strained or totally relaxed. We also find intermixing of Ga and In and the formation of $\text{In}_x\text{Ga}_{1-x}\text{As}$ with $0.21 \leq x \leq 0.42$. However, the diffraction intensity originating from intermixed materials is quite weak. We reveal for the above composition range a volume fraction of intermixed materials within the InAs QDs, of 3% only. We suggest that this material is located in relaxed $\text{In}_x\text{Ga}_{1-x}\text{As}$ islands, which may be related to the findings of Joyce *et al.*¹³ Due to a slightly lower growth temperature in our case, these islands may be in competition to growth of strained InAs islands. The almost complete absence of fully strained InAs can be explained by additional intermixing at the dot base, however, this is not directly accessible to x-ray diffraction.

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