High-resolution x-ray diffraction of self-organized InGaAs/GaAs quantum dot structures

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The structural properties of highly strained buried In$_x$Ga$_{1-x}$As layers on GaAs substrates are investigated by high-resolution x-ray diffraction. Such layers of a few monolayers in thickness serve for the formation of self-organized quantum dots by the Stranski–Krastanow growth mode. Exceeding a critical layer thickness the growth mode changes from two-dimensional Frank–van der Merwe to the three-dimensional Stranski–Krastanow mode resulting in the formation of coherently strained In$_x$Ga$_{1-x}$As islands. X-ray spectra of such structures below the growth mode transition can be perfectly simulated using dynamical theory allowing for determination of layer thickness with submonolayer sensitivity and composition within 5%. Dot formation manifests itself in a decrease of the effective In content of the wetting layer. © 1996 American Institute of Physics.

Heteroepitaxial growth of highly strained InGaAs/GaAs has gained increasing interest as it offers the possibility to produce nanoscale structures like quantum dots (QDs) and quantum wires in situ without any substrate patterning process. These low dimensional structures show unique physical properties, particularly interesting for novel optoelectronic devices like QD lasers with low threshold current density and high $T_0$. For periodic quantum dots high-resolution x-ray diffraction (HRXRD) was shown to provide structural information on nonrandom strain.

Self-organized quantum dot structures were mainly investigated until now by means of optical methods and microscopic techniques such as atomic force and electron microscopy. Due to their low degree of periodicity little is known about their structural properties. In this letter, we report on HRXRD investigation of such quantum dot structures.

A series of five samples of constant composition In$_{0.55}$Ga$_{0.45}$As but of different nominal layer thickness was grown on exactly (001) oriented undoped GaAs substrates by metalorganic vapor phase deposition (MOCVD). The growth rate and composition were controlled via the growth of thick reference layers.

The MOCVD growth was carried out in a horizontal, lamp-heated reactor equipped with a rotating susceptor at a pressure of 20 mbar. As source materials, trimethylindium, trimethylgallium, and pure arsine were used. After deposition of a 200 nm thick GaAs buffer layer at 670 °C the growth was interrupted and the temperature was reduced to 500 °C for growing the InGaAs layer at a growth rate of 0.5 monolayers/s with a V/III ratio of 400. After a growth interruption of 5 s the structure was nominally capped with 30 nm GaAs grown at 500 °C.

In order to determine layer thickness and composition independently of optical methods, x-ray diffraction and grazing incidence x-ray reflection spectra are taken using a Philips MRD materials research diffractometer employing Cu $K_{α1}$ radiation with a four-crystal monochromator set for the (220) Ge reflection mode in the primary beam. Rocking curves were simulated using a computer program which is based on the solution of the Takagi–Taupin equations of dynamical diffraction theory.

The photoluminescence (PL) spectra of the 2, 3, 5, and 8 ML samples are shown in Fig. 1. The emission lines at $> 1.5$ eV are due to the GaAs substrate. The quantum well luminescence from the InGaAs layers is denoted by QW. The beginning of dot formation at $d > 3$ ML manifests itself in an additional PL line at $> 1.15$ eV. When $d$ is increased to 5 ML the dot luminescence increases and clearly dominates the spectrum. The line position remains nearly constant in contrast to the QW line which continuously shifts to lower energy with growing thickness. This behavior can be explained...
by a higher density of dots of the same size in the 5 ML sample as compared to the 3 ML sample. When \(d\) is further increased to 8 ML the dot luminescence intensity remains constant but its energetic position is shifted 90 meV towards lower energy indicating enlargement of already existing dots without significant change in dot density, consistent with transmission electron microscope (TEM) images.

Figure 2 shows a (004) \(\omega/2\Theta\) scan of a 3 monolayer (ML) GaAs/In\(_{0.55}\)Ga\(_{0.45}\)As/GaAs sample together with three simulated curves where the composition was varied in steps of 5%. Note the logarithmic scale and the dynamic range of 6 orders of magnitude. The interference pattern (Pendellösung fringes) is caused by the phase shift between the diffracted waves from the upper GaAs cap layer and the lower GaAs substrate which are separated by 3 ML In\(_{0.55}\)Ga\(_{0.45}\)As having a lattice constant in growth direction which is different from the GaAs one by \((\Delta a/a)_z = 7.89\%\). The GaAs cap layer has the same lattice constant \(a_z\) in comparison to the substrate, but its relative atomic positions in growth direction are shifted by a constant amount. The effective phase shift depends on the strain-thickness product.\(^{12,13}\) The GaAs cap layer thickness is 29 nm as determined from the Pendellösung fringe spacing. This result is confirmed by grazing incidence reflection measurements which serve as an independent measurement of the layer thickness. The high sensitivity of the details of the interference fringes around the GaAs(004) Bragg peak allows for determination of the wetting layer thickness with submonolayer resolution once the strain, i.e., the composition, is known. In turn, from the variation of the shape around the substrate peak the composition could be determined within 5% at a given thickness. In Fig. 3 the experimental and calculated spectra for 1, 2, and 3 ML samples of the same composition \(x_{ln} = 55\%\) and same cap layer thickness \(d_c = 29\) nm are shown. All curves are normalized to the same intensity. There is nearly perfect agreement between the simulated curves and the experimental ones for all three samples indicating completely coherent scattering as described by the dynamical theory. Comparing the 1 and 3 ML spectra, we observe that the shape next to the substrate peak has drastically changed from an asymmetric to a nearly symmetric one. Moreover the intensity of all fringes has increased by a factor of \(\approx 2\) as compared to the 1 and 2 ML spectra.

From Holloway’s analysis\(^{14}\) it is seen that in the (004) reflection the interference system cycles around the substrate reflex with period \(2\pi\) for an InGaAs thickness change \(d = a_0/4\varepsilon_z\), where \(a_0 = 0.5635\) nm is the lattice constant of the GaAs and \(\varepsilon_z = 0.0789\) is the effective mismatch of the strained In\(_{0.55}\)Ga\(_{0.45}\)As layer in the growth direction, i.e., \(d = 1.785\) nm. A symmetric shape of the interference pattern is only possible if the in-phase conditions are fulfilled, i.e., when the phase shift amounts to multiples of \(\pi\). Thus, a symmetric spectrum is expected for the first time with \(1\times\pi\) corresponding to \(d/2 = 0.893\) nm which is just 3 ML.

Applying the same parameter set now to the 5 and 8 ML samples yields rather poor agreement with the experimental spectra as shown in Fig. 4 by the dotted lines (for clarity the top of the substrate peak is now shown). First of all, the periods of the simulated Pendellösung oscillation fringes do not fit the experimental ones. In addition, there are significant differences between the experimental and theoretical envelope functions and intensities of the fringe pattern. For instance, in the simulations there are nodes in the fringe pattern at \(+2800\) arcsec for the 5 ML sample and at...
Krastanow growth mode. Using plan-view transmission electron microscopy the island density was determined to be approximately 5% of the whole substrate surface. In addition, the occurrence of dots and clusters will reduce the nominal layer thickness in these samples. Assuming the dots have lateral dimensions up to 100 nm and a height of about 50 nm and a density of 1.3 × 10^10 cm^{-2}, the dots have lateral dimensions ≤20 nm×20 nm and a height of ~6−7 nm filling about 5% of the whole substrate surface. In addition there are large clusters with a diameter up to 100 nm at a height of 50 nm and a density of 1.3×10^8 cm^{-2} and 1.8×10^9 cm^{-2} for the 5 and 8 ML samples, respectively. Of course, the occurrence of dots and clusters will reduce the nominal layer thickness in these samples. Assuming the dots and clusters being of pyramidal shape and all incoming group III atoms being incorporated into the layer, the volume contribution of the dots and clusters will reduce the effective wetting layer thickness by about 1 ML. However, we obtained no better agreement between experiment and theory using reduced thicknesses at the same composition.

Satisfactory agreement is only obtained when reducing the In content of the wetting layer in both samples from 55% to ~45%. The corresponding simulations are shown as solid lines in Fig. 4. It is important to note that the calculated positions of the QW luminescence of the 5 and 8 ML samples are also in much better agreement with the reduced In concentration of 45% than with the 55% value. Thus, we conclude that the formation of the dots is associated with a decrease of the effective In content of the wetting layer. The effect is attributed to an enhanced strain-driven In-interlayer migration at that thickness which prevents the In from being incorporated into the layer. Indium surface segregation has already been observed some time ago with MBE grown InGaAs/GaAs layers.\(^{16,17}\) Because of segregation and re-evaporation the true In mole fraction in InGaAs quantum wells was less than intended. Since a reevaporation of In can be excluded at our growth temperature of 500 °C we speculate that the In might be accumulated in the large clusters observed by TEM in these samples.

In summary, high-resolution x-ray diffraction is suitable for the characterization of buried quantum dot structures. From a comparison of the x-ray spectra of dotted and undotted InGaAs layers a reduction of the effective In content of the wetting layer is found in the dotted layers. The effect is attributed to an enhanced strain-driven In migration at the Frank–van der Merwe to the Stranski–Krastanow growth mode transition.

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\(^{10}\)S. Takagi, Acta Crystallogr. 15, 1311 (1962).


