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X-ray standing wave and high-resolution x-ray diffraction study of the GaAs/InAs/GaAs(100) heterointerface

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The composition profile of a InAs monolayer buried in a GaAs matrix is studied by combining high-resolution x-ray diffraction and x-ray standing-wave experiments. This combination provides a comprehensive structural analysis in terms of strain status, layer thickness, and interfacial atomic configuration of this extremely thin layer. We found the InAs layer to be pseudomorphically matched to the GaAs host crystal. Moreover, we measure a total amount of In (6.739×10^{14} atoms/cm^2) distributed, at the heterointerface, within 3 monolayers in the following percentages: 75% in the first monolayer and 20% and 5% in the second and third, respectively. The In atoms are not randomly distributed as they would be in $Ga_xIn_{1-x}As$ alloy, but form InAs terraces. Finally, these results demonstrate the growth procedure employed to be very efficient in minimizing the impact of In segregation.

Segregation phenomena at III-V semiconductor heterointerfaces have been the key focus of numerous investigations throughout the past few years.[1–7] Since segregation effects can alter the originally intended composition profile of the heterostructure as well as its electronic properties beyond recognition, it is of vital importance to determine the actual composition profile in the final structure. Gerard and Marzin have suggested an elegant indirect approach for the InAs/GaAs system which, however, is based on unreliable band-structure data.[8] High-resolution transmission-electron microscopy (HRTEM) also has shown the capability to image such heterostructures; however, the translation of a TEM image into a concentration profile is accompanied by ambiguities.[9] Furthermore, HRTEM is a local probe of the heterostructure.

In this paper we present a versatile method to determine the composition profiles in III-V heterostructures on the monolayer scale which is based on a combination of high-resolution x-ray diffraction (HRXRD) and x-ray standing wave (XSW). Our study of an InAs/GaAs heterostructure shows that this method also provides very detailed information about structural parameters such as strain status, layer thickness, atomic distribution of the In atoms, and interface quality.

The structure investigated was grown by elemental source molecular-beam epitaxy. During the growth, the surface reconstruction was checked by means of *in situ* reflection high-energy electron diffraction. A GaAs buffer layer, 1 μm thick, was grown on the (100)-GaAs substrate at a temperature of 580 °C with a growth rate of 2 Å/s. Then, the temperature was lowered down to 420 °C and 1.2 monolayers (ML) (Ref. 10) of InAs was deposited in steps of 0.6-ML increments, annealing the sample at 420 °C for 120 s after each InAs deposition step. Then, 5 ML of GaAs were grown at 420 °C on the InAs layer, the growth was interrupted and the temperature was raised up to 540 °C. This higher temperature causes desorption of the In atoms which have eventually segregated at the growth surface (flash-off step). A cap layer of GaAs, 300 Å thick, was grown at 540 °C on the 5-ML GaAs layer grown at lower temperature. The as-grown structure was investigated by HRXRD and XSW.

The HRXRD study was performed by means of a computer-controlled double-crystal diffractometer in the nondispersive $(n,-n)$ geometry.[11] A rotating anode (Cu target) was used as x-ray source ($\lambda=1.54$ Å) and an asymmetrically cut Ge (100) monochromator was used to collimate the incident beam. The angular resolution of this instrument was calculated to be 11 μrad. The experimental x-ray patterns were analyzed by the x-ray diffraction dynamical theory for distorted crystals in order to determine strain (chemical composition) and thickness of the layer structure.[11]

The x-ray standing-wave experiments were performed on the setup of the Laboratoire de Minéralogie-Cristallographie (Paris, France), which is installed on the beam line D15B of the DCI storage ring of the Labora-

toire pour l'Utilisation du Rayonnement Electromagnétqiue (LURE, Orsay, France). The equipment, described elsewhere,[12] was modified by putting a Si(100) channel-cut monochromator[13] in a vacuum chamber (10^{-5} torr). The sample was measured in N_2 gas atmosphere in order to avoid any effect of contamination on the fluorescence yield of the $L\alpha_1$ emission line of In from the $K\alpha_1$ line of Ar.

It should be noted that much attention was paid to the structural quality of the substrate crystal. Structural defects in the substrate may produce diffuse scattering which destroys coherent phenomena such as interference processes. Therefore, the samples selected for our experiments showed a shape and a full width at half-maximum of the Darwin curve in excellent agreement with the dynamical x-ray diffraction theory.

The XSW results were interpreted by means of the dynamical theory of x-ray diffraction.[14,15] According to this theory, standing waves occur in the crystal as a result of the interference process between incident and diffracted wave fields. The standing-wave pattern has the periodicity of the reflecting planes. By rocking the crystal within the angular range of the Darwin curve, the standing-wave pattern shifts by half a period. Therefore, a layer of atoms buried in the host lattice will experience the highest absorption when the antinodal planes of the electric field pass through them. Consequently, if any phenomena related to the x-ray field intensity at the investigated atom, for example, x-ray fluorescence or photoelectron emission, is monitored simultaneously with the rocking curve, the shape of this signal will change according to the position of the emitting atoms[16] with respect to the reflecting planes.

The normalized x-ray fluorescence intensity $Y(\theta)$ is given by

$$Y(\theta)\sim 1+R(\theta)+2\sqrt{R(\theta)}F\cos[\nu(\theta)-2\pi P]\ , \qquad (1)$$

where $R(\theta)$ is the reflectivity and $\nu(\theta)$ is the phase between the incident and diffracted fields. The parameters F and P will be called *coherent fraction* and *position,* respectively. P is normalized to the interplanar distance of the substrate diffracting planes. In the case of occupancy of N different lattice planes, the resulting F and P are related to the percentages f_i of impurity atoms distributed on different positions P_i by the following expressions:[17]

$$P=\frac{1}{2\pi}\arctan\left[\frac{\sum_{i=1}^{N} f_i\sin(2\pi P_i)}{\sum_{i=1}^{N} f_i\cos(2\pi P_i)}\right] \qquad (2)$$

and

$$F=D_{\mathrm{DW}}(1-D_S)A_G \qquad (3)$$

with

$$A_G=\left[\left[\sum_{i=1}^{N} f_i\sin(2\pi P_i)\right]^2+\left[\sum_{i=1}^{N} f_i\cos(2\pi P_i)\right]^2\right]^{1/2}, \qquad (4)$$

where A_G is the multiple positions factor, D_{DW} is the Debye-Waller factor which represents the thermal vibrations, and D_S represents the random static disorder.[18] By fitting the rocking curve and the fluorescence yield it is possible to evaluate P and F and hence to obtain information about the positions of the impurity atoms and the atomic disorder.

Figure 1 shows the experimental (a) and simulated (b) rocking curve recorded in the vicinity of the (422) Bragg reflection, in the grazing incident angle geometry, by HRXRD. The structure investigated here is, besides the substrate, a three-layer system: a GaAs buffer layer, an InAs interlayer, and a GaAs cap layer. The intensity modulation (*Pendellösung*) of the interference pattern is caused by two-field beating phenomena between the beams diffracted by the GaAs bulk crystal (buffer layer and substrate) and by the thin GaAs cap layer, which are dephased due to the presence of the InAs interlayer. The theoretical model used in the analysis of the experimental patterns correlates the intensity modulation (ΔR) of the interference pattern directly with the total amount of In atoms per cm^2 at the heterointerface (N) by means of the following expression:

$$\Delta R\approx\sin[-8\pi\sin(2\theta_B)N(\omega+s)/\lambda|\gamma_h|a^2]\ , \qquad (5)$$

where θ_B is the Bragg angle corresponding to the GaAs diffracting planes, $\gamma_h=\sin(\theta_B+\alpha)$ with α being the angle between the diffracting planes and the surface, a is the in-plane lattice constant, ω is the deviation from the Bragg angle, and s is the strain function. The strain function depends on the strain in the growth plane (in-plane) and on the strain perpendicular to the growth plane.

The in-plane strain of the InAs layer determined from the experimental pattern was zero, i.e., the InAs layer is pseudomorphically grown on the GaAs substrate. In fact, the total amount of InAs deposited during the growth (1.2 ML) does not exceed the critical thickness (2–3 ML),[19] and explains in this way the pseudomorphism of the InAs layer. The best fit resulting from the theoretical model used simulates a GaAs bulk crystal covered by an InAs layer, containing a total amount of In atoms of 6.739×10^{14} atoms/cm^2,[10] and by a GaAs cap

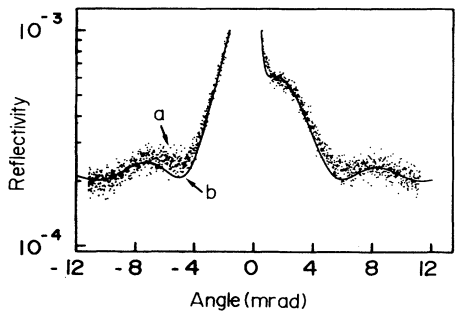

FIG. 1. The dotted curve (*a*) represents the experimental x-ray diffraction pattern recorded in the vicinity of the GaAs-(422) Bragg angle for incident glancing angle geometry; the solid line (*b*) is simulated for a structure consisting of 1-μm-thick GaAs buffer layer, a 1.08-ML InAs interlayer, and a 27.5-nm GaAs cap layer.

layer, 27.5 nm thick. In order to demonstrate the accuracy of the In determination, we report in Fig. 2 three simulations differing in the In content, namely, 6.489×10^{14} atoms/cm^2 (curve *a*), 6.864×10^{14} atoms/cm^2 (curve *b*), and 7.238×10^{14} atoms/cm^2 (curve *c*). Differences of 2% in the total amount of In atoms, i.e., $\Delta N=\pm1.2\times10^{13}$ atoms/cm^2, are still detectable.

HRXRD gives only qualitative information about the atomic interface configuration, i.e., about the spatial distribution of In atoms at the interface, whereas XSW allows the accurate quantitative evaluation of the atomic positions at the interface.

Figure 3 shows the experimental rocking curve (open circles) and the $L\alpha$ fluorescence intensity from the In atoms (solid circles) as a function of the Bragg angle corresponding to the (400)-GaAs reflection. The two solid lines (*a* and *b*) correspond to the best fits of the experimental patterns obtained by convoluting the theoretical curves with a Gaussian profile of 2.04 arcsec full width at half-maximum. The reason of the convolution is related to the wavelength angular dispersion due to the mismatch between the monochromator (Si channel cut) and the GaAs sample. By fitting our fluorescence data we are able to evaluate the two free parameters introduced in Eq. (1), i.e., the resulting coherent fraction F and the atomic position P. We found from the best fit the following values: $F=0.58-0.07$ and $P=1.17-0.02$. The coherent fraction F is the product of D_{DW}, the Debye-Waller (DW) factor, $1-D_S$, where D_S is the random static disorder and the occupation factor for multiple positions A_G [Eq. (4)]. The lower F is, the higher the interface static disorder. The atomic position P is the ratio between the distance of the indium atoms from the diffracting planes d_{In} and the periodicity of the standing-wave pattern which coincides with the GaAs-(400) plane distance, d_{GaAs}.

The XSW data cannot be interpreted by assuming that the In atoms are incorporated on one single position ($A_G=1$), i.e., only in one (100) monolayer. The position $P=1.17\pm0.02$ is in principle consistent with a single InAs ML having a strain between 15% and 19%. However, in this case, the low coherent fraction of 0.58 has to be exclusively explained by thermal and static disorder. Different D_{DW} factors between a minimum value of 0.89 and a maximum of 0.95 can be calculated for the (400)-GaAs reflection.[20] Therefore, $F=0.58\pm0.07$ should be mainly due to a very high static disorder, namely, 35–40 % of the In atoms occupying random positions. Such a high disorder is, however, very unlikely because high-resolution electron microscopy always revealed almost perfect lattice images recorded on other GaAs/InAs/GaAs heterostructures grown by means of the same procedure and under the same conditions.[21,22] Therefore, it is more reasonable to assume an incorporation on multiple positions (terrace nucleation). Considering a Debye-Waller factor of ~0.9, we found that our experimental data are entirely consistent with the latter possibility by assuming that 75% of the total amount of In atoms (f_1) are incorporated on the first position ($P_1=1.15$), 20% (f_2) on the second ($P_2=3.45$), and 5% (f_3) on the third ($P_3=5.75$). The error in the occupation probabilities is about 5%, and that in the position about 1%. Each position is displaced with respect to the GaAs planes by 15% as we would expect in the case of terrace nucleation of the deposited InAs layer. On the contrary, a $Ga_xIn_{1-x}As$ alloy formation would result in a displacement of the In position with respect to the GaAs diffraction planes varying with the InAs mole fraction.

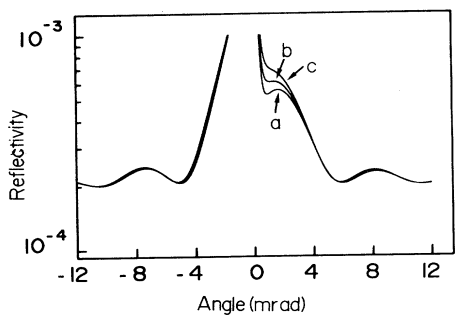

FIG. 2. Simulations of x-ray diffraction curves done for a pseudomorphic InAs interlayer of 1.04 ML (*a*), 1.1 ML (*b*), and 1.16 ML (*c*) thickness. The interlayer is inserted between a 1-μm GaAs buffer layer and a 27.5-nm GaAs layer.

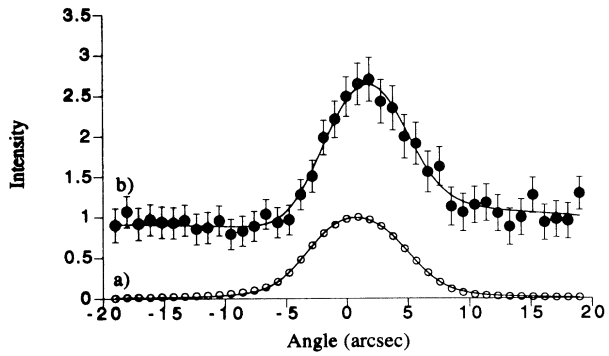

FIG. 3. The dotted curves are the rocking curve (bottom) and the In $L\alpha$ fluorescence peak (top) recorded in the vicinity of the (400) reflection of the GaAs substrate. The solid lines are the best fits of the experimental curves.

Furthermore, the In distribution detected from the XSW data clearly shows that the employed growth procedure is very efficient in minimizing the effect of In segregation. The In concentration drops to zero within three monolayers. Moreover, multiplying f_1 by the total amount of In atoms measured by HRXRD (1.08 ML), it results that 0.82 ML is the actual amount of InAs in the first monolayer. The 0.18 ML of InAs missing the first monolayer could be explained as a segregation process or as nucleation in form of terraces directly in the first ML. Even in the segregation hypothesis, it should be noted that the employed flash-off step is very efficient in depleting also the topmost layers of the low-temperature GaAs cap from segregated indium.

In conclusion, we have demonstrated that concentration profiles in semiconductor heterostructures can be determined with monolayer resolution by combining XSW and HRXRD measurements. This combination provides a complete description of the strain and of the interfacial atomic configuration of the investigated

InAs/GaAs buried interface. In particular, the strain status at the heterointerface showed a pseudomorphic InAs layer and an In content of 6.739×10^{14} atoms/cm^2. The distribution of In atoms per monolayer is as follows: 75% are located in the first (100) monolayer, 20% in the second, and 5% in the third. The positions of the In atoms (strain status) clearly indicates InAs terrace formation rather than a $Ga_xIn_{1-x}As$ alloy, where the In atoms are randomly distributed.

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