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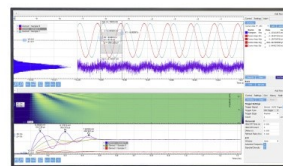
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Ex-situ investigation of indium segregation in InGaAs/GaAs quantum wells using high-resolution x-ray diffraction

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Calculations using the dynamical theory of diffraction together with a sample model which considers the segregation of indium atoms were employed to fit the high-resolution x-ray spectra of strained InGaAs/GaAs quantum wells grown by molecular-beam epitaxy. The segregation coefficients obtained from the best fits to the experimental data of samples grown at different temperatures are in excellent agreement with the expected values and confirm that x-ray diffraction is a valuable tool for the investigation of the segregation phenomenon. © 2003 American Institute of Physics. [DOI: 10.1063/1.1621738]

INTRODUCTION

The heteroepitaxy of InGaAs layers on GaAs substrates is characterized¹ by a strong segregation of indium (In) atoms that accumulate at the growth front and strongly modify the In-composition profile, resulting in different electronic and optical properties of the devices based on that material. The origin of segregation is still under debate^{2,3} and, therefore, it is very important to have as many tools as possible with which to investigate its underlying physical mechanisms. X-ray diffraction has already been used to probe In segregation but, depending upon the structure of the samples, it was found to be useless⁴ or to provide only limited information.⁵

We propose a versatile method by which to determine the composition profile of InGaAs/GaAs quantum wells (QWs) that is based on a combination of high-resolution x-ray diffraction (HRXRD) measurements and a best-fit simulation procedure. The segregation equations proposed by Muraki and co-workers⁶ are introduced directly into the calculations of HRXRD spectra to determine the segregation coefficient R which was found to be in excellent agreement with the value obtained by reflection high-energy electron diffraction (RHEED) measurements performed *in situ* during growth of the samples.

EXPERIMENTAL DETAILS

The epitaxial layers were grown in a GEN II molecular-beam epitaxy (MBE) system on top of GaAs(001) substrates. After oxide desorption and a 2500 Å thick GaAs buffer grown at 570 °C, the substrate temperature was lowered to deposit 100 Å of In_{0.15}Ga_{0.85}As material followed by a 100 Å thick GaAs layer. The rest of the GaAs cap (400 Å) was grown at 570 °C. The In_{0.15}Ga_{0.85}As well and the first part of the GaAs cap of each sample were grown at different substrate temperatures (520, 460 and 370 °C) in order to investigate its influence on the In-segregation process. HRXRD

measurements were performed with Cu K α_1 radiation in a Philips X'Pert high-resolution diffractometer in a triple-axis configuration with a four-crystal Ge(220) monochromator in the primary optics and a Ge(220) channel-cut analyzer in front of the detector.

RESULTS AND DISCUSSION

Figure 1 shows a $\omega/2\theta$ scan measured around the (004) Bragg diffraction peak for the three samples. All the spectra exhibit similar features: the most intense narrow peak belongs to the GaAs substrate and is used as a reference, while the weaker broader band centered around 32.4° is due to the InGaAs layer that is inhomogeneous (due to alloy fluctuations and segregation) and therefore exhibits a wider range of (vertical) lattice parameters resulting from the different lattice constants of InAs and GaAs. Fringes modulate the spectra and originate from interference of the electromagnetic radiation within the layered structure of the samples, thus confirming their excellent structural quality.⁷

In general, simulation using diffraction theory together with a sample model is performed in order to best fit the experimental x-ray data and provide the exact thickness and composition of each layer that constitutes the sample. Obviously, a sample model which only considers layers with uniform composition cannot be used to simulate the HRXRD pattern of our samples, because segregation strongly modifies the In concentration at both QW interfaces. Since all information about the structural parameters of the layers (thickness, composition and strain) is embodied in the HRXRD spectra, the interference fringes might provide an excellent probe of the In composition profile in the samples, in contrast to what was suggested by Fujimoto *et al.*⁸ who neglected them.

In order to simulate the measured $\omega/2\theta$ scans displayed in Fig. 1, the spectra were calculated within the framework of the dynamical theory of x-ray diffraction using Takagi-Taupin equations⁹ and a sample model which took into account the segregation equations proposed by Muraki and

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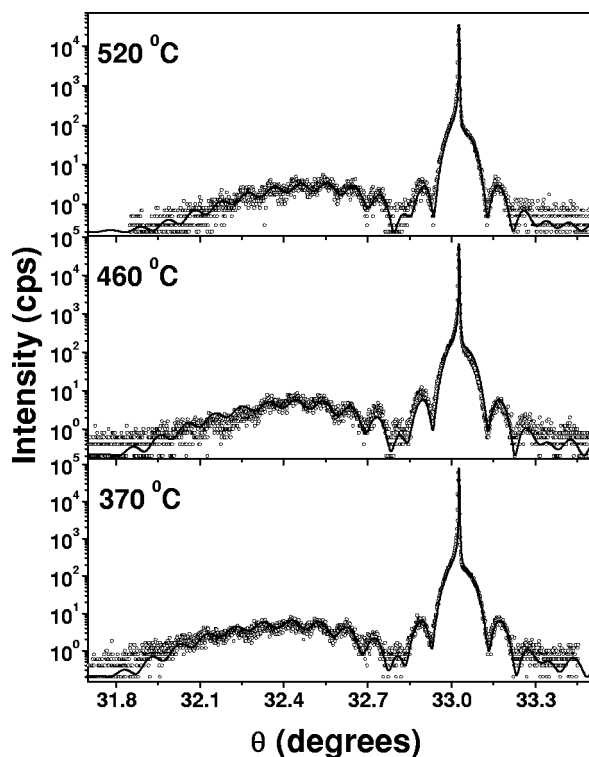


FIG. 1. $\omega/2\theta$ scans measured around the (004) Bragg diffraction peak (open circles) of $\text{In}_{0.15}\text{Ga}_{0.85}\text{As}/\text{GaAs}$ QWs grown at different temperatures. The solid lines represent the best fits to the data using a sample model that takes segregation into account. The values of the segregation coefficient R obtained from the best fits are 0.85, 0.74 and 0.62 for the QWs grown at 520, 460 and 370 °C, respectively.

co-workers.⁶ Their phenomenological model has been shown to describe most of the manifestation of segregation up to now^{3,10–12} and simply assumes that a constant fraction R of the In atoms present on the top layer of the crystal always migrates toward the next layer while the rest of them ($1 - R$) are incorporated into the bulk crystal each time a new monolayer is completed. The In composition in the n th layer is given by

$$x_n = x_0(1 - R^n) \quad (\text{for the well}), \quad (1)$$

$$x_n = x_0(1 - R^N)R^{n-N} \quad (\text{for the barrier}), \quad (2)$$

where x_0 and N are, respectively, the nominal In concentration and the total number of $\text{In}_x\text{Ga}_{1-x}\text{As}$ monolayers (MLs)

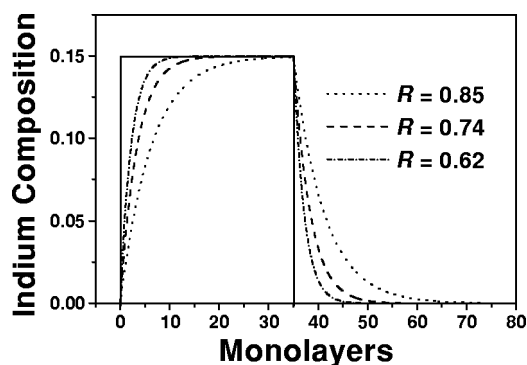


FIG. 2. In-composition profile of an $\text{In}_{0.15}\text{Ga}_{0.85}\text{As}$ QW with segregation coefficients $R=0.85$, 0.74 and 0.62. The solid line represents the nominal square profile ($R=0$).

in the well. For the calculations, the sample was divided into four parts: the GaAs substrate, N ML of $\text{In}_x\text{Ga}_{1-x}\text{As}$ with the In content described by Eq. 1 (well region), N ML of $\text{In}_x\text{Ga}_{1-x}\text{As}$ with the In content given by Eq. 2 (barrier region), and a GaAs cap layer. All layers were assumed to be completely strained to the GaAs substrate, i.e., they had the same in-plane lattice constant throughout the whole structure. Considering that the nominal composition x_0 and well width N were precisely obtained from our *in situ* RHEED measurements (approximately 0.15 and 35 ML, respectively), the segregation coefficient R became the main fitting parameter of our method. Finally, a routine calculated the x-ray spectra of each sample for values of R ranging from 1 to 0 and compared them with the experimental HRXRD results using a least-squares routine that deals with the logarithm of the data.

The best fits to the experimental scans are plotted in Fig. 1 and were obtained with values of R equal to 0.85, 0.74 and 0.62 for the samples grown at 520, 460 and 370 °C, respectively. The segregation coefficient increases with a rising growth temperature, as expected, and is in excellent agreement with the values in the literature.¹³ Simulations of our HRXRD data were also performed with two segregation coefficients, R_w (in the well) and R_b (in the barrier), in order for the calculations to converge toward different values of these parameters as a consequence of the different amounts of In and opposite behavior of the surface In population in both cases.¹¹ We found that the best fits were obtained when R_w and R_b were identical to the values estimated previously with a single R parameter, which is consistent with the fact that segregation efficiency does not depend on the number of In atoms present at the surface nor on the way the In material is supplied to the epitaxial layers.¹⁴ These values of R were also positively cross checked by *in-situ* RHEED measurements performed on the samples, since it was recently demonstrated that the strong decay of RHEED oscillations during InGaAs deposition was directly connected to the presence of the In population on the surface of the sample and was thus an excellent probe of the segregation phenomenon.¹¹ All these results definitely confirm the physical basis of our model and indicate that the technique is really able to probe the In-profile modification resulting from the segregation phenomenon.

Once the best value of R was found for each sample, the real In profile could be determined by substituting R in Eqs. 1 and 2; this led to the graphs shown in Fig. 2. One can observe that the width of both interfaces of the QWs increases at higher growth temperature as a consequence of the larger segregation length of the In atoms that strongly modifies their In profile with respect to the nominal square profile. Lowering the growth temperature reduces this difference, and the In profile approaches that of the nominal one, thus leading to optical and electronic properties that are closer to the expected ones. However, it should be emphasized here that, when an experimental physical property (that is a function of segregation) matches its theoretical predicted value without taking segregation into account, it definitely does not mean that the segregation coefficient is zero, as often claimed in the literature. In fact, when the best fits of the

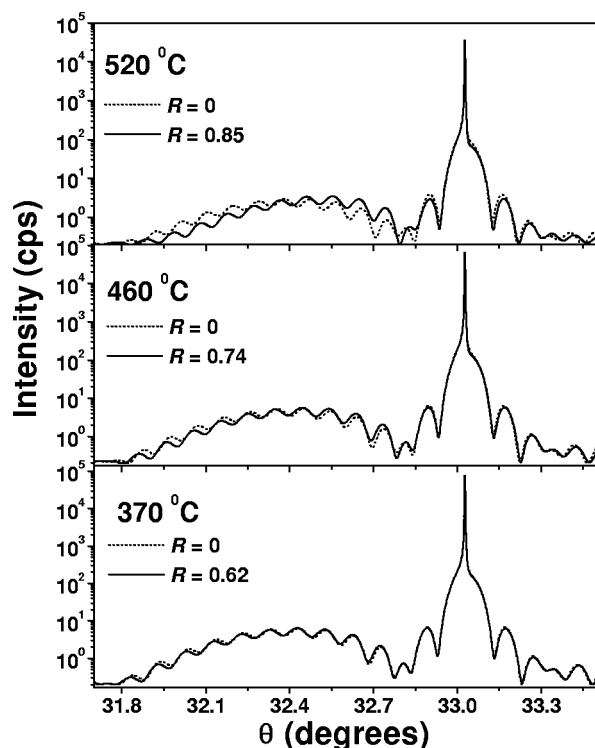


FIG. 3. Comparison between the best fits presented in Fig. 1 (solid lines) and the spectra calculated assuming no segregation ($R=0$) for the $\text{In}_{0.15}\text{Ga}_{0.85}\text{As}/\text{GaAs}$ QWs grown at 520, 460 and 370 °C.

HRXRD data shown in Fig. 1 are compared with the same simulations where R was assumed to be zero, as in Fig. 3, it is clear that, as the growth temperature decreases, the respective simulation tends toward the one with $R=0$. For $T=370$ °C, both curves are almost undistinguishable, but the corresponding In profiles are still very different as illustrated in Fig. 2. These results suggest that x-ray diffraction might no longer be sensitive to any In-profile variation when R is smaller than 0.6, but it does not mean that $R=0$ each time both simulations match.⁸ Since the segregation phenomenon is thermally activated, its contribution to the HRXRD spectra decreases at low temperature, and the uncertainty of the absolute value of R determined in that way increases. Therefore, below 370 °C, the curves become indistinguishable and this technique no longer provides a reliable value of the segregation coefficient. The same situation is also frequently encountered in photoluminescence (PL) measurements in the literature where it is claimed that R is zero each time the optical-transition energy of InGaAs QWs no longer changes with the growth temperature or is equal to the expected theoretical value without segregation.^{4,15} This is not necessarily true and only means that the difference in In profile with respect to the ideal square potential is no longer enough to influence significantly the energy levels within the structure and, as a consequence, the optical properties of the system.

The method introduced here is a very powerful and versatile tool for the investigation of segregation because it relies simply on the knowledge of the structural parameters of the samples, which are usually provided by the growers, and on the simulation of the experimental x-ray diffraction data

using the segregation coefficient R as the main fitting parameter. Since high-resolution x-ray diffractometers are now readily available, are able to analyze any type of material without requiring specific sample preparation, and are commercially available with user-friendly simulation software, such a technique should provide very useful data on the subject, especially when no information about the RHEED-intensity decay is available, as is most often the case. It is also more straightforward and accurate than PL measurements, because optical experiments need different types of optics and detectors as a function of the material constituting the samples, and their data must be compared with theoretical calculations that are very sensitive to the theoretical model, mathematical approximations and physical parameters used to compute the electronic properties of the structure designed, thereby yielding large variation of the possible values of R for a specific sample.

CONCLUSION

We showed that HRXRD is able to provide an accurate estimate of the segregation coefficient R when phenomenological equations that describe segregation are introduced in the simulation of the experimental data. This is of great relevance for investigation of the phenomena in the InGaAs alloy that became so important in optoelectronics. Our results also suggest that, below a certain value of R , segregation seems to no longer have any significant effect on the physical properties of the samples, but it does not mean that it is absent.

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