In-situ **Determination of Indium Segregation in InGaAs/GaAs Quantum Wells Grown by Molecular Beam Epitaxy**

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The surface segregation of indium atoms during the growth of InGaAs/GaAs heterostructures has been investigated *in situ* by reflection high-energy electron diffraction (RHEED). We pointed out that the strong damping of the RHEED oscillations during the deposition of InGaAs on GaAs was related to the segregation strength of indium atoms in the InGaAs layer. A simple model shows that the decay constant of the RHEED oscillations may be used to determine accurately the segregation coefficient R, as confirmed by photoluminescence (PL) measurements.

I Introduction

Since its discovery [1, 2], surface segregation in III-V semiconductor alloys has been the subject of numerous studies, in particular in InGaAs/GaAs quantum wells (QWs) [3, 4] because of their importance to the optoelectronics and microwave industries. It is now well established that segregation represents the ultimate limitation to obtain perfectly abrupt interfaces necessary for the production of high-performance devices [5]. This phenomenon has already been investigated by various surface-sensitive methods such as x-ray photoelectron spectroscopy (XPS) [6], Auger electron spectroscopy (AES) and RHEED [7, 8], or more indirectly through the fit of QW optical-transition energies from composition-profile models [3]. However, until now none of the physical theories reported in the literature was able to explain the origin of the effect in a satisfactory way for all the different materials and systems investigated.

In this work, we propose a simple and very efficient way to determine, *in situ* and in real time, the surface segregation of indium (In) atoms during the growth of InGaAs layers on GaAs(001) substrates. Our method, that is based on the interpretation of the intensity variation of the RHEED oscillations, differs from the other few RHEED studies by the fact that they only provide qualitative results [7] or are restricted to a specific material or growth technique [8].

II Experimental Details

The sample investigated here was grown in a Mod. Gen. II molecular beam epitaxy (MBE) system from Varian and consisted of a 0.25μm-thick GaAs buffer grown at 570 ^{o}C and a QW structure grown at 520 ^{o}C. After the GaAs buffer layer, the substrate temperature was ramped up to 610 ^{o}C during 2 minutes to smooth the GaAs surface and then ramped down to 520 ^{o}C to deposit the 50Å-thick $In_{0.14}Ga_{0.86}As$ layer. A growth interruption of 20 s was realized at the second interface and, finally, a 50Å-thick GaAs layer was deposited at 520 ^{o}C, followed by 450Å of GaAs grown at 570 ^{o}C. The growth rates of the materials were determined by RHEED measurements that yielded a value of 0.79 and 0.68 monolayer per second (ML/s) for the $In_{0.14}Ga_{0.86}As$ and GaAs layers, respectively. The sample was not rotated during the growth of the structure in order to allow RHEED measurements during the deposition of the InGaAs layer and the second GaAs barrier. The RHEED measurements were carried out with a 8kV electron gun and a glancing angle of 1.0^o along the [110] direction of the GaAs(001) surface. After the growth, the sample was analyzed by PL spectroscopy at 1.4K using conventional lock-in techniques, a cooled GaAs Photomultiplier and the 5145Å line of an argon laser.

III Results and Discussion

Fig. 1 shows a typical RHEED oscillation during the deposition of GaAs layers on a GaAs(001) surface.

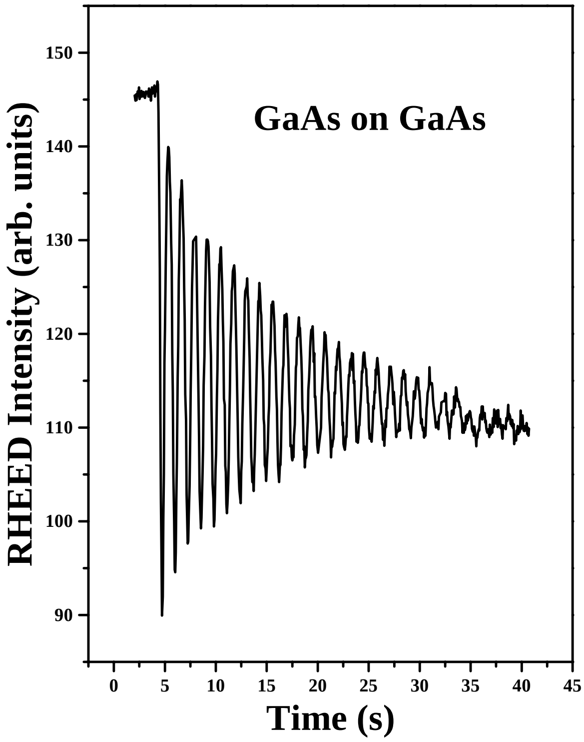

Figure 1. Typical RHEED oscillations during the deposition of GaAs on a GaAs(001) substrate at 570 °C.

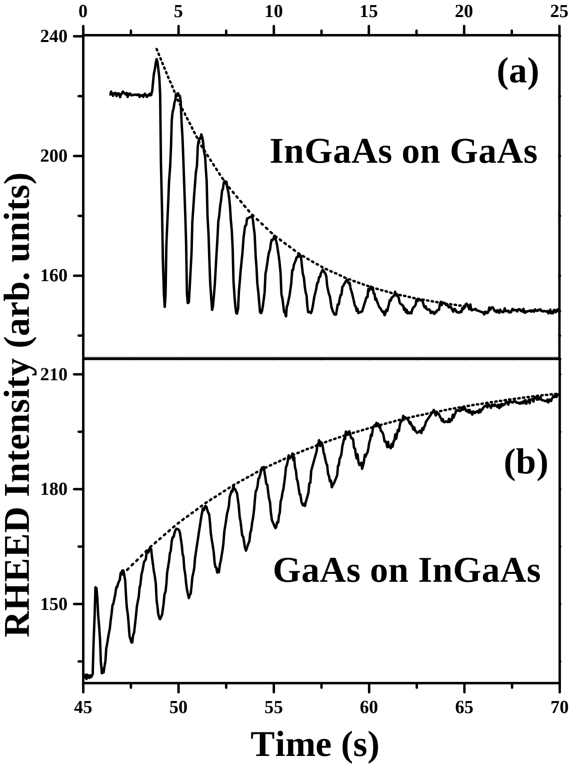

Figure 2. RHEED oscillations (solid lines) detected a) at the first interface of the InGaAs QW; b) at the second interface of the InGaAs QW. The dotted lines show the best fit of the damping using equation 1.

Since, under conventional growth conditions, homoepitaxy proceeds in the layer-by-layer growth mode, the oscillations are usually explained in terms of the roughening of the growth front by the nucleation of small two-dimensional(2D) islands that merge to complete the layers one after the other one. This periodic roughening of the surface induces a periodic scattering of the electron beam that yields a minimum reflection of the specular beam when half of the layer is deposited and a maximum reflection when the layer is complete, as shown in Fig. 1. However, as the total roughness of the growth front increases slowly with time, the amplitude of the oscillations decreases as the growth proceeds.

Fig. 2a shows the RHEED oscillations when the $In_{0.14}Ga_{0.86}As$ layers of the 50Å-wide QW were deposited on the first GaAs barrier at 520 °C. A strong and asymmetric damping of the oscillations is observed (unlike in Fig. 1) and is usually related in the literature to the strained growth originating from the lattice mismatch between GaAs and InAs [9]. In these conditions, the initial stage of the deposition occurs under strain and the InAs layer adopts the lateral lattice parameter of the GaAs substrate, storing elastic energy in the epitaxial layer. The growth still occurs in a 2D manner, but now nucleation of new islands on the top layer before the completion of the underlying layer is energetically more favorable, unlike in the case of homoepitaxy, because they allow a partial relaxation of the strain at the island edge and contribute to decrease the total energy of the surface. As a consequence the surface becomes rough faster, yielding a stronger damping of the oscillations. However, such arguments are not totally satisfactory because they do not explain why the damping varies with temperature and why it still appears during the growth of lattice-matched heterostructures [8].

In order to better understand the origin of the phenomenon, we investigated the influence of the growth conditions on the behavior of the RHEED oscillations during deposition of InGaAs on GaAs and observed that their damping was stronger at higher temperature and at lower growth rate and V/III flux ratio. Coincidentally, In segregation is known to be very effective in this ternary alloy and strongly depends on the surface mobility of the group-III adatoms that varies exactly in the same way with respect to these growth parameters. On the basis of this analogy, we tried to fit the decay of the RHEED oscillations in order to extract any parameter that could be related to the strength of In segregation in the InGaAs layers. The expression

$$I = I_0 + I_1 exp(\pm n/\tau) \tag{1}$$

was used, where n is the number of oscillations (*i. e.* of MLs) and τ the decay constant that is the relevant fitting parameter expressed in MLs. The best fit of the experimental data is shown in Fig. 1a. The real physical meaning of τ can be better understood when the segregation coefficient R is expressed as [3]

$$R = exp(-1/\delta) \tag{2}$$

where δ is a characteristic length (expressed in MLs) over which segregation is effective. If the damping of the RHEED oscillations is a manifestation of In segregation (as we suggest), we should be allowed to substitute δ by τ and calculate R. Using the experimental data of Fig. 2a and their best fit from equation 1, we obtained R=0.83, which is a typical value for the growth conditions used here [10]. When the second GaAs barrier is deposited on top of the InGaAs layers, the large number of In atoms accumulated on the surface is progressively incorporated into the GaAs barrier and the envelope of the RHEED oscillations recovers its normal intensity as shown in Fig. 2b. Since this behavior also results from the segregation of In atoms, we calculated R (using the + sign in equation 1) and obtained 0.84, a value that is in excellent agreement with the other one previously determined at the first interface, showing the consistency of our model. Other similar RHEED studies were carried out changing the growth parameters (growth temperature, growth rate, V/III flux ratio) in order to verify their influence on the numerical value of R. In all the cases, we obtained extremely coherent and reproducible results compatible with the data available in the literature [11].

To cross check the validity of our model, PL measurements were carried out on the same sample. In segregation generally produces a blueshift of the optical QW emission with respect to the expected electronic transition of a square QW because of the non-ideal potential profile at both interfaces. Using the effective masses of electron and heavy holes for a biaxially-strained InGaAs layer, we could obtain their confinement potentials and solve the Shrodinger equation. Assuming an exciton binding energy of 7 meV, the optical transition of the square QW was estimated in 1.4229 meV, showing a redshift of 5.3 meV with respect to the experimental PL energy (1.4282 meV)[11]. To determine R, we used the composition-profile model of Muraki [3] according to which the In composition x_n of the nth layer is

$$x_n = x_0(1 - R^n) \qquad n \leq N \tag{3}$$

where x_0 and N are the nominal In composition (0.14) and the well width in MLs (18), respectively. The segregation coefficient R is adjusted in order to provide the right value of the In content that has to be used to calculate the electron and heavy-hole confinement energies that will fit the experimental PL data [4]. For the optical emission detected in our sample, we obtained a value of R=0.83, which is in perfect agreement with the previous results provided by the RHEED technique.

IV Conclusion

We showed that the damping of the RHEED oscillations during the MBE growth of InGaAs layers is related to the strength of In segregation that can be quantitatively estimated in a very accurate way. This powerful and versatile method allows real-time and *in-situ* investigations of the phenomenon and will surely provide new interesting data in this field.

Acknowledgments

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