***IN SITU* DETECTION OF AN INITIAL ELASTIC RELAXATION STAGE DURING GROWTH OF $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}$ on GaAs (001)**

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Abstract

We have followed, *in situ* and real-time, both the relaxation and morphological evolution along [110] direction during the growth of $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}/\text{GaAs}$ by molecular beam epitaxy at low growth rates (0.2 ML/s and 0.5 ML/s). The stress measurements were performed by optical monitorization of the strain-induced substrate curvature, and the morphology evolution was assessed by means of laser light scattering. The correlation of the real-time results obtained from both *in situ* techniques allowed us to detect the existence of a growth rate dependent initial elastic relaxation regime, which is associated with the development of a long-range ordered rippled-like morphology along $[1\bar{1}0]$ direction. DOI: [10.1016/S0169-4332\(01\)00768-1](https://doi.org/10.1016/S0169-4332(01)00768-1)

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1. Introduction

Growth of heteroepitaxial layers with lattice parameter mismatch constitutes a current approach in semiconductor technology. On one hand, strained layers are incorporated in optoelectronic devices taking advantage of the modification of band structure by strain. On the other hand, accomplishment of band-gap engineering without lattice parameter restrictions requires the possibility of introduction of relaxed buffer layers in order to change the lattice parameter from that

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of the substrate to any other according to device design. In both approaches, knowledge of relaxation process and strain evolution is of fundamental importance.

During mismatched heteroepitaxial growth, several stages can be considered. First, the layer grows pseudomorphic to the substrate, is thermodynamically stable and accumulates elastic strain energy. As the growth proceeds, an increasing amount of accumulated elastic energy makes the layer to become metastable and the relaxation process begins [1, 2]. Relaxation of low-strained layers ($\varepsilon < 2\%$) usually takes place through misfit dislocations, and it is well known that the surface of these layers develops an undesired crosshatched morphology once the mechanisms for plastic relaxation start [3-5]. Furthermore, some studies on the low-mismatched SiGe/Si [6, 7] and $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{InP}$ ($x \geq 0.53$) [8] systems have shown that the surface can also roughen prior to the generation of misfit dislocations and the subsequent crosshatch development. This surface roughening is associated with an elastic relaxation process and depends strongly on the growth kinetics. However, up to now this elastic relaxation stage has not been detected in low-strained $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ layers [4].

Although extensive studies have been carried out both on morphology and relaxation, most of the experimental work has used post-growth characterization techniques, and then cannot provide information on the possible kinetic effects. In a previous work we studied the influence of kinetics on the morphological evolution during growth of $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}/\text{GaAs}$ by molecular beam epitaxy (MBE) using *in situ* laser light scattering (LLS) [5]. We found that surface roughening takes place before the dislocation multiplication processes begin to actuate. Moreover, we observed that the thickness at which the LLS intensity starts to increase, or equivalently the thickness at which surface roughness begins to develop, as well as the subsequent roughness evolution, depend on the growth rate. Our X-ray diffraction (XRD) studies on these samples failed to observe any difference in the relaxation processes for the different growth rates, but these results do not preclude the existence of growth dependent elastic relaxation processes at the surface.

In the last years, a new *in situ* stress characterization technique, based on substrate curvature optical monitorization, has been successfully used in order to study stress relaxation during the growth of SiGe/Si [9] and III-V compounds [10, 11]. This technique has been successful in providing direct experimental evidence of elastic relaxation as a consequence of the formation of 3D nanostructures. Complemented with *in situ* LLS measurements for morphology evolution, this technique could then provide information about the existence of elastic relaxation during growth of $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}/\text{GaAs}$ and how kinetics influences the process. In this paper, we present the *in situ* and real-time results obtained on the morphology and stress relaxation evolution during the first stages of growth of $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}$ on GaAs (001) by molecular beam epitaxy (MBE) at two different growth rates. Our results show a growth rate dependent initial elastic relaxation stage, which is caused by the development of surface roughness.

2. Experimental procedure

Samples were grown by MBE on on-axis Si-doped GaAs (001) epi-ready substrates with a nominal threading dislocation density of 10^4 cm^{-2} . After thermal desorption of the oxide, a 100 nm thick GaAs buffer layer was grown at substrate temperature $T_s = 580^\circ\text{C}$. $\text{In}_x\text{Ga}_{1-x}\text{As}$ layers, with a nominal In content of $x=0.2$, were grown at $T_s = 500^\circ\text{C}$ using a group V to group III flux ratio around 20/1 and at two different growth rates, 0.2 and 0.5 monolayers per second (ML/s).

In situ and real-time strain measurements were performed during the growth by following the stress-induced substrate curvature using the deflection of a laser beam. For this purpose, the 350 μm thick GaAs substrates were shaped as cantilevers along [110] direction and mounted on a special substrate holder which allowed free bending of the sample. Substrate curvature and accumulated stress in the layers ($\Sigma\sigma$) are related by the Stoney's equation [9, 10]. The layer relaxation and the In composition were obtained from the *in situ* accumulated stress measurements and also from *ex situ* X-ray diffraction. The X-ray diffraction measurements were performed in a diffractometer with 4 crystal Ge (220) as incident beam optic. The (+ -) Bragg arrangement for the (004) reflection and the ($\theta+\Phi$) ($\theta-\Phi$) arrangement for (115) reflections were used. These four diffractograms were taken in both [110] and $[\bar{1}\bar{1}0]$ directions for each sample. From the recorded data and by using a dynamical simulation program we have obtained the alloy composition and the strain in the InGaAs samples.

For the *in situ* LLS measurements, the sample surface was illuminated with a He-Ne laser ($\lambda = 633$ nm) at an angle of incidence $\theta_i = 50^\circ$ and the scattered light was collected at an angle $\theta_s = 0^\circ$, with respect to the surface normal. We employ lock-in detection in order to reject spurious signals coming from the hot effusion cells and to improve the signal-to-noise ratio [5, 12]. The *ex situ* surface morphology characterization has been carried out by atomic force microscopy (AFM).

3. Results

Figure 1 shows the *in situ* and real-time measurements of accumulated stress evolution during the growth of the first 100 nm of $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}$ layers at two different growth rates, 0.5 ML/s (Fig. 1(a)) and 0.2 ML/s (Fig. 1(b)). The scattered light intensity for the first 50–60 nm has also been included in this figure. Both $\Sigma\sigma$ and LLS data were taken along the [110] sample direction.

As we can see in Fig. 1, the accumulated stress has a first stage where it increases linearly with the thickness. Then we observe a clear slope change, followed by a stage where $\Sigma\sigma$ remains constant. The initial linear behaviour corresponds to the pseudomorphic growth regime. In this regime, each deposited monolayer incorporates the same amount of stress, which corresponds to the misfit strain ε_0 . Consequently, the accumulated stress increases linearly with thickness and its slope allows to determine the composition x of the $\text{In}_x\text{Ga}_{1-x}\text{As}$ layers [9, 13]. For the samples shown in Fig. 1, the composition calculated from the initial slope of the $\Sigma\sigma$ curve are given in Table 1, together with the composition determined by XRD measurements. These samples were grown up to a thickness around 400 nm (not shown in Fig. 1). In table 1, the degree of relaxation at the final thickness obtained from both techniques are also given. We want to emphasize the agreement between *ex situ* and *in situ* measurements ensuring the quantitative validity of the *in situ* measurements during the whole growth process.

After the initial linear increase in the $\Sigma\sigma$ curve we observe in Fig. 1 a slope change, which indicates the occurrence of a relaxation process. This process cannot correspond to the Matthews relaxation mechanism [1], based on the bending of a threading dislocation originating from the substrate which create a misfit dislocation segment at the interface, for two main reasons. First, the critical thickness value at which this process occurs is $h_c \sim 6$ nm in $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}/\text{GaAs}$, as has been

Fig. 1. *In situ* and real-time measurements of accumulated stress ($\Sigma\sigma$) and laser light scattering (LLS) signal evolution during growth of $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ (001) layers, with nominal indium content $x=0.2$, at different growth rates: (a) $r_g = 0.5$ ML/s; (b) $r_g = 0.2$ ML/s. The dashed lines represent the linear fits of the initial part of the $\Sigma\sigma$ curves, from which the composition can be calculated. The solid circles correspond to the relaxation degree value obtained from post-growth X-ray diffraction measurements.

calculated theoretically [1] and demonstrated experimentally [14]. Second, the relaxation associated with the Matthews mechanism depends on the quality of the substrate. Then, the maximum relaxation that could be achieved in our layers is as low as $\Delta\varepsilon = 10^{-4}$, and this small change in the layer strain, not detectable by XRD [15], would produce a change in the experimental $\Sigma\sigma$ curve slope much smaller than the measured one. Moreover, this $\Delta\varepsilon = 10^{-4}$ is too small for the sensitivity of our present *in situ* configuration, where we use a 350 μm thick cantilever. This explains why we cannot detect the Matthews relaxation mechanism and still consider the growth as pseudomorphic for layer thickness much larger than h_c .

Table 1

Indium composition x , and relaxation degree R , as obtained from the *in situ* and real-time stress measurements and post growth X-ray diffraction (XRD) characterization for $\text{In}_x\text{Ga}_{1-x}\text{As} / \text{GaAs}$ (001) layers grown at two different growth rates.

Growth rate (ML/s)	In composition (x)		Thickness (nm)	R (%)	
	<i>In situ</i> stress measurements	<i>Ex situ</i> XRD Measurements		<i>In situ</i> stress measurements	<i>Ex situ</i> XRD measurements
0.5	0.20 ± 0.02	0.20 ± 0.005	450	82 ± 3	82 ± 2
			47	9 ± 1	0
0.2	0.18 ± 0.02	0.21 ± 0.005	370	79 ± 3	80 ± 2
			50	15 ± 1	0

The Matthews relaxation process failing to account for the detected slope change in $\Sigma\sigma$ curve, one can think in other plastic relaxation mechanisms involving nucleation of new dislocations. In order to check this possibility, we have grown samples with thickness slightly above the critical value where the slope change in the accumulated stress occurs (50 nm for the 0.2 ML/s grown sample and 47 nm for the 0.5 ML/s). We have characterized these samples by post-growth XRD measurements, and the obtained relaxation values are given in Table 1, together with the relaxation values calculated from the *in situ* measurements for the corresponding thickness. We have also represented in Fig. 1 by two solid circles the $\Sigma\sigma$ values calculated from XRD measurements on these layers. It can be seen that in both cases the samples are fully strained for XRD sensitivity (the experimental diffraction peaks overlap with the peaks simulated for a layer coherent to a GaAs substrate), whereas the *in situ* measurements indicate a relaxation degree of 9 or 15 %. Since plastic relaxation of this value should have been detected by XRD measurements, this *in situ* detected relaxation mechanism may originate from a different process, which we think it is related to the inhomogeneous elastic relaxation via surface roughness development.

This conclusion is supported by the results on surface morphology evolution shown in Fig. 1. It can be observed that both the thickness at which the $\Sigma\sigma$ curve slope changes and that where LLS signal takes off (directly related with surface roughening) depend on the growth rate. Moreover, surface roughening occurs at smaller thickness for smaller growth rate, just as the detected relaxation. As a more significant argument, we want to point out that the onset of the LLS increase and the change of the $\Sigma\sigma$ slope take place at the same thickness (39 nm and 31 nm for 0.5 and 0.2 ML/s respectively).

As growth proceeds, the nucleation and multiplication mechanisms begin to actuate, which corresponds to the region where the $\Sigma\sigma$ curve remains constant [2, 13]. When the plastic relaxation stage is reached, the *ex situ* XRD relaxation values are again in agreement with the *in situ* results (see Table 1 for around 400-nm-thick layers), and the morphology develops a characteristic crosshatched pattern [5].

Figure 2 shows an AFM image of the 50-nm-thick sample grown at 0.2 ML/s. For this particular thickness and growth rate, the LLS signal has already increased and the *in situ* stress measurement has detected 15% elastic relaxation although no plastic relaxation was found by XRD (see Table 1). In the image an undulated or rippled-like anisotropic roughness along $[1\bar{1}0]$ direction can be seen together with a straight and high (1.7 nm) ridge at the left-hand side. In a previous work [5], we associated the increase in scattered light to the development of these high ridges built on top of the misfit dislocations formed by the Matthews mechanism. However, our present *in situ* stress results show an elastic relaxation stage taking place during the growth, which cannot be accounted for by the low density of ridges formed at the surface. Instead, we now propose that the rippled-like roughness is likely to contribute significantly to the increase of LLS signal as well as be responsible for the detected elastic relaxation stage. Moreover, this roughness is aligned along $[1\bar{1}0]$ direction (the preferential surface diffusion direction in (001) oriented III-V semiconductor surfaces) and so it would relax stress along $[110]$, which is the direction measured in the *in situ* stress experiments. *In-situ* measurements along $[1\bar{1}0]$ in order to check the anisotropic nature of this elastic relaxation mechanism and *ex situ* angle resolved light scattering characterization [12] for a fully understanding of the *in situ* LLS signal evolution are being carried out.

Figure 2. AFM image of the 50-nm-thick $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}$ layer grown at 0.2 ML/s. A line profile is presented at the upper right part of the figure. Below it, an average profile of the whole image along [110] direction, excluding the high ridge appearing at the left side, is shown. Both profiles have been plotted with the same height scale.

In Fig. 2 it is shown two surface profiles along [110] direction (single and averaged over the whole image) taken without including the highest ridge. From these profiles we obtain the mean period for the ripples to be 200 nm. These profiles evidence that there is a long-range order in the surface morphology, as can be deduced from the perfectly defined ridges in the profile averaged over a distance as large as 3 μm . In previous works on the $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}/\text{GaAs}$ system [4], this rippled-like morphology has not been observed. The fact that our samples built this kind of initial roughness could be due to the lower growth rate (0.2 and 0.5 ML/s) we have employed. As has been established, morphological instabilities can form whenever the growth rate of the film is smaller than the rate of development of the instability. [16].

4. Summary and conclusions

We have monitored *in situ* both the relaxation and morphological evolution along [110] direction during the growth of $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}/\text{GaAs}$ by MBE. For the low growth rates employed (0.2 ML/s and 0.5 ML/s), the real-time stress measurements show a growth rate dependent initial elastic relaxation regime previous to the plastic relaxation one. A simultaneous surface roughness development was detected by complementary *in situ* LLS characterization. Correlation of the results obtained by both *in situ* techniques demonstrates that surface roughening and elastic relaxation are intrinsically linked processes.

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