# Influence of Surface Structure on Segregation and Alloy Properties in (100)- and (311)-Oriented InGaAs/GaAs Heterostructures

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The influence of surface orientation and surface structure on indium segregation and alloy properties were systematically studied in InGaAs/GaAs quantum well structures grown by molecular beam epitaxy. (100), (311)A and (311)B surface orientations and different approaches in the growth interruption at the interfaces were used in this investigation. The segregation process and alloy parameters were obtained by photoluminescence and RHEED measurements.We find significant differences in the optical properties and growth kinetic for the three orientations. Using growth interruption we were able to change the surface structure and reduce the segregation process for all orientations.

## I. Introduction

The growth of pseudomorphic structures $In_xGa_{1-x}As$-GaAs heterostructures on GaAs substrates with different orientation has attracted much attention, due to the strong fundamental interest in growth-related phenomena and numerous practical applications[1]. For instance, the formation of self organized quantum dots of InAs during the growth has been proposed recently[2]. However, the kind of surface structure that determines the growth kinetics and morphology on high index surfaces is still unknown[3]. In addition, it is well known that in Molecular Beam Epitaxy (MBE) technique indium segregates during the growth of $In_xGa_{1-x}As$ over GaAs, which results in a gradual composition of layers. Such inherent indium segregation process has frustrated any attempt to employ abrupt interfaces up to date. Modifications introduced in the growth conditions were not enough to reduce the segregation because of the narrow growth window found for this material system.

In this work, the effect of surface orientation and step density on indium segregation were systematically studied in InGaAs/GaAs single quantum wells (SQW's) structures grown by MBE. We used the fact that the growth nucleation, surface kinetics and morphology can be changed or improved for orientations other than (100) for studying and reducing segregation phenomena in $In_xGa1-xAs$/GaAs quantum well (QW) structures. (100), (311)A and (311)B surface orientations and different approaches in the growth interruption at the interfaces were used in this investigation.

## II. Experiments

All the samples used were grown by MBE on semi-insulating (100)-, (311)A- and (311)B-oriented GaAs substrates. They were soldered side-by-side with In on molybdenum holders for comparison. The grown structures of SQW's in study consist of a 0.5 $\mu$m GaAs barrier, followed by a 3 nm $In_{0.2}Ga_{0.8}As$ well covered by 30 nm thick GaAs cap layer. Surface wells consisting of 40 nm $In_xGa_{1-x}As$ ($x = 0.1$) were used for studying In surface segregation and evaporation. The growth temperature was fixed at 540°C for part of the samples and varied from 480°C to 600°C in the case of temperature dependence experiments. Growth interruption delays varying from 0 to 120 seconds were carried out only at the inverted GaAs-to-InGaAs interface. Photoluminescence (PL) measurements were performed at about 17 K. Details of the PL setup were published elsewhere[4].

RHEED intensity oscillations were used to monitor the surface structure during the growth.

## III. Results and discussions

### III.1. Optical properties: interface and alloy properties

In Fig. 1 we display the common features of PL spectra in semilogarithmic scale for nominal 3 nm $In_{0.2}Ga_{0.8}As/GaAs$ SQW's. For this set of samples no growth interruption was carried out at the QW interfaces. Although this procedure would favor surface roughness, the emissions reveal narrow gaussian shaped (dashed lines) with 0.8 meV half width for both (311) oriented samples. In earlier work[4] we have shown that the interface scattering related to interface roughness is suppressed by segregation. In other words, the smearing of the interfaces, due to the gradual composition of the layers, makes the excitonic levels less sensitive to the interfacial fluctuations. These results show clearly that segregation and surface orientation play an important role in the QW properties. Therefore, it is important to separate the effect of surface orientation and surface structure on In segregation.

In Fig. 2 we present the luminescence emission halfwidth ($\sigma$) as a function of the well thickness ($L_z$) for all orientations. This characteristic gives a qualitative information about the mechanisms responsible for the excitonic emission broadening. The experimental points are connected by broken lines for clarity. The solid line shows the theoretical dependence of the halfwidth assuming just the contribution of the alloy disorder in the QW's. A good agreement between theoretical model and our experimental data is obtained for average cluster size present in the alloy $r_c = 1.58 r_0$, where $r_0 = 2.2$ Å is the radius associated with the atomic volume in a perfect alloy. This agreement indicates that the excitonic states are broadened only by alloy scattering mechanisms[4]. However an important feature of the results in Fig. 2 is the low alloy disorder in the (311)A oriented InGaAs wells in comparison to the (100) ones. In the case of the (31 1)B orientation the excitonic emission broadening is not limited only by alloy disorder but also by defect-related broadening mechanisms when the layer thicknesses are bigger than 4 nm [4]. For the case of no growth interruption (Figs. 1 and 2), additional scattering due to interface roughness is likely to occur in our samples. The results displayed in Figs. 1 and 2 show that this effect is suppressed because of In segregation. The smearing of the interface, due to a gradual composition of the layers, makes the excitonic levels less sensitive to the interfacial fluctuations.

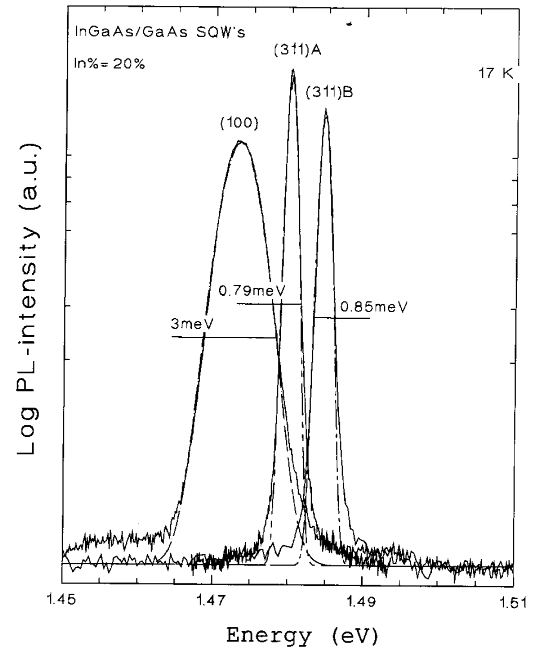


Figure 1. 17 K PL spectra from 3 nm $In_{0.2}Ga_{0.8}As/GaAs$ SQW's having the (100), (311)A and (311)B orientations.

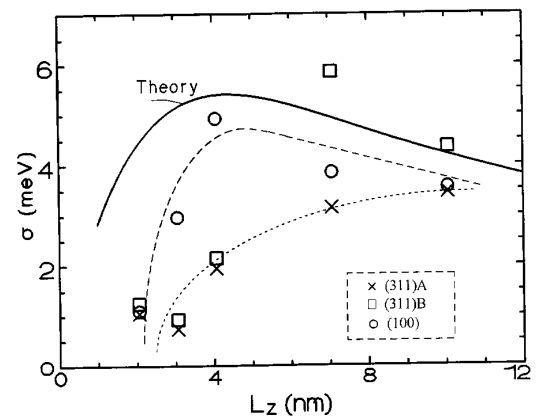


Figure 2. Well width dependence of the PL half width for single QW's having the (100)-, (311)A- and (311)B-orientations. The continuous line is the calculated line width due to alloy disorder. Broken lines are guides for the eye.

### III.2 Effect of surface orientation

The degree of In segregation in the three orientations was obtained qualitatively through the change of the potential profile with the growth temperature. In Fig. 3a we show the energy shift of the QW emissions as function of the inverse of the substrate temperature. As before, no growth interruption was taken at the interfaces for these samples. Fig. 3b displays for consistence and comparison the In composition $x$, which was calculated from emissions of the 40 nm InGaAs surface wells grown with nominal In concentration of 10%. As one can see (Fig. 3b), indium incorporation decreases monotonically for temperatures above $\sim$ 550 °C. Below this temperature In evaporation can be regarded as negligible, but we still observe a shift of the PL emissions in the quantum wells as a consequence of segregation effects (Fig. 3a). However, this shift is small for the (311)A orientation, which means that In surface segregation is strongly suppressed for these samples. In this temperature regime, where there is no significant In evaporation, the composition $x$ in the (311)A surface wells corresponds to the 10% expected nominally. On the contrary, $x$ is smaller (about 6%) in the case of the (100) and (311)B orientations. These results just confirm the low segregation rates found for the (311)A orientation. The fraction of deposited In which does not segregate is actually incorporated into the final (311)A structure. This result is in agreement with the activation energies $E_A$ of 3.14 eV and 2.83 eV for the indium desorption obtained for (311)A and (311)B surfaces, respectively, from Fig. 3b. These values for $E_A$ are in agreement with data previously published for (100) surfaces[5,6]. The high value of the activation energies for the (311)A orientation is accounted for the very reactive double dangling bounds As site which stick efficiently the In atoms. On contrary, the group III element has a single-dangling bond on the (311)B surface, explaining the low affinity for the In atoms.

The reduction of the In floating layer on the growth front should also improve the alloy quality[7], which is in agreement with the small linewidth observed before. Sangter[8] also suggested that on (m11) surfaces there is a high density of steps compared to the atomicaly flat (100) surface. Such additional surface structures (steps and/or corrugations), together with the surface roughness due to the absence of interruption, would favor kinetically the indium incorporation in the (311)A surface, resulting in the reduction of segregation.

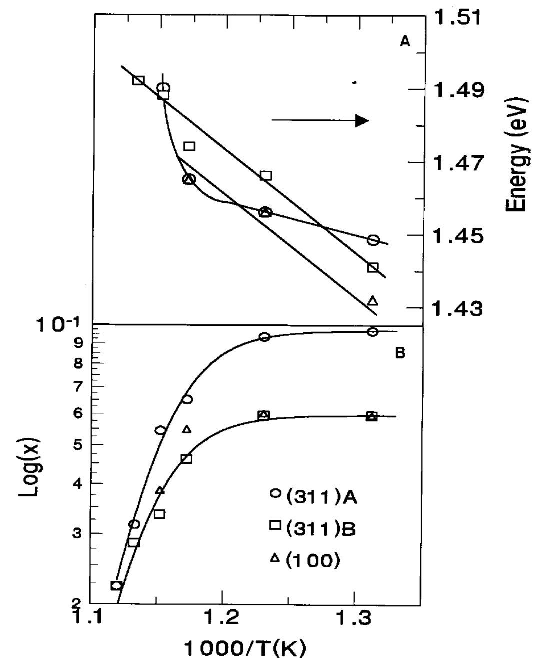


Figure 3. (a) PL emission energy for 3 nm wide QW $In_{0.2}Ga_{0.8}As/GaAs$ and (b) indium composition obtained from the PL spectra of a 40 nm surface well as a function of the inverse of substrate temperature.

### III.3 Effect of surface structure

In order to confirm the effect of the surface structure on segregation we carried out growth interruption at the lower GaAs-to-InGaAs interface. With this procedure we expect to modify both step structure and surface roughness. Fig. 4 shows the results of this experiment for 3 nm SQW's grown simultaneously on the (100), (311)A and (311)B surfaces. Interruption delays of 0 and 120 seconds were considered for the samples in this Fig. 4. The main effect of the growth interruption is the significant shift to higher energies for all orientations. This observed shift in the PL emissions can only be explained in terms of the increase of the segregation length (flatness and smearing of the QW profile) produced by the change of the surface properties.

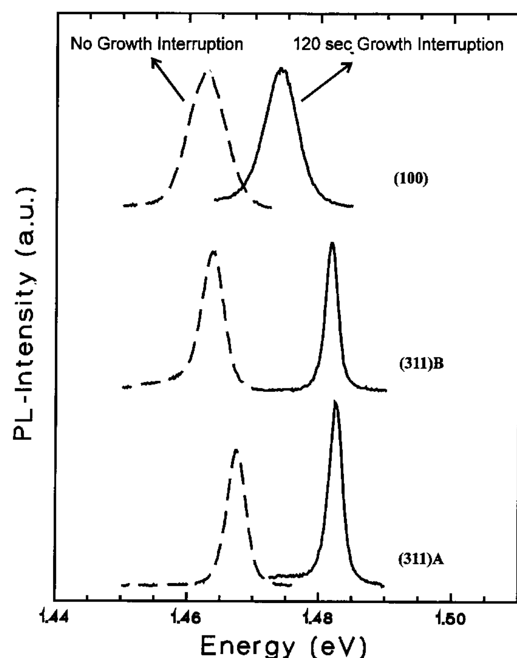


Figure 4. Exciton PL spectra of 3 nm SQW's grown simultaneously on (100), (311)A and (311)B surfaces showing the growth interruption experiments. The interruption delays used for this set of samples were 0 and 120 seconds and were carried out only at the inverted interface (GaAs-to-InGaAs).

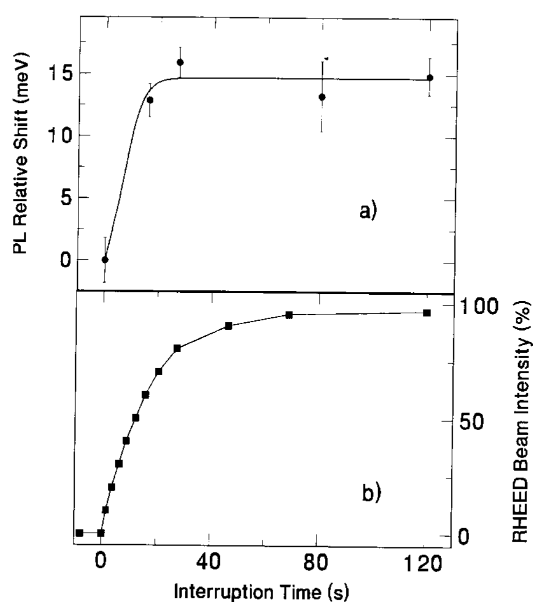


Figure 5. Comparison between the relative shift in the PL emissions for the case of a 3 nm SQW (a) and RHEED specular beam intensity recovering as a function of the interruption time at the inverted interface (GaAs-to-InGaAs). The shifts are determined relative to the emissions from QW's with no growth interruption.

To clarify which surface process is responsible for the PL shift during the growth interruption, RHEED specular beam intensity recovering was used for the (100) surface. Fig. 5b gives the RHEED intensity recovery after the growth of 0.5 $\mu$m thick GaAs layer at 540 °C as a function of the interruption time. The monotone increase of the RHEED intensity reflects itself the reduction of the equilibrium density of growth nuclei on the growing surface terraces. For comparison, we include in Fig. 5 (part a) the relative shift of the PL emissions for 3 nm SQW's grown with different interruption delays at the inverted GaAs-to-InGaAs interface. The shift in Fig. 5a was determined relative to the emission of the SQW grown with no interruption. Surprisingly a qualitative agreement is obtained between the relative shift of the PL emissions and the change of the surface nuclei structure.

The effect of the surface structure on the peak position in the PL spectra for the (100) samples can be explained in term of the morphology evolution during the interruption time (Fig. 6). At the relative low substrate temperatures used in this work (~540°C) the 2D nucleation growth mode dominates. In this case, the surface morphology is the result of the equilibrium between the densities of isolated 2D nuclei of different sizes and steps on the growing surface. With the growth interruption 2D nuclei coalesces with the terrace steps. As a consequence of this process, RHEED intensity increases exponentially and an equilibrium terrace lengths of about 50 nm are expected for the $(100) \pm 30'$ oriented GaAs surfaces after 30 seconds of interruption, as can be seen in Fig. 5a. The presence of such large terraces increases the effective life time of the isolated In atoms on the terraces and, consequently, would favor its segregation. This growth picture confirms our assumption that surfaces with high density of 2D nuclei reduce In segregation on the (100) surfaces and is in agreement with the previous results in Figs. 4 and 5.

The growth kinetic of InGaAs on the high index (311) surfaces is very different when compared with the (100) surfaces. Accordingly, the non reconstructed (311) surface exhibits alternate (100) terraces and (111) step edge atom with period of 6.64Å [9]. Under these ideal conditions the MBE growth should be predominantly governed by the step flow propagation mode and no morphology change on the small terraces is expected. As a consequence, RHEED oscillations do not take place and surface segregation would not change. However, the PL shifts (Fig. 4) suggest other surface

structures different from the ideal one for the (311) samples which change during the growth interruption.

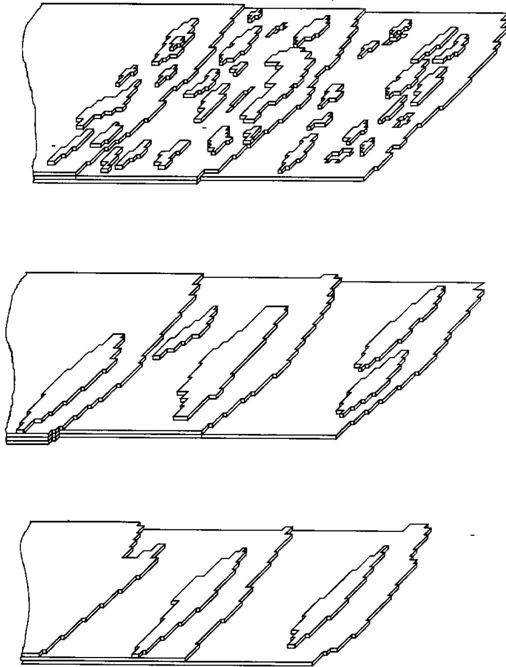

Figure 6. Schematic illustration of the structural changes on a (100) GaAs surface during the interruption procedure.

According to Briones et al.[3] the As stabilized (311)B surface has the same coverage and probability reconstructs in similar way as the (100). This would explain the same In concentration for (311)B and (100) surfaces in Fig. 3b and similar PL segregation dependence on the growth temperature in Fig. 3a. Different authors[10,11] also suggest the existence an energetically stable surface based on macrosteps and macrofacets (corrugation) on the (311)A surface which could explain the results found in this work. The kind of surface structure that determine our results for the (311) orientations is still an open question. However, more growth experiments are underway trying to clarify these points.

## IV. Conclusions

The present work exhibit significant differences in the optical properties and growth kinetic in InGaAs/GaAs SQW's grown on (100), (311)A and (311)B surface orientations. No growth interruption at the interfaces allows us to reduce the segregation effects in QW's, although quite sharp gaussian shaped excitonic transitions with line width of about 0.8 meV are still obtained. Our results show smaller indium surface segregation and alloy disorder in the (311)A orientations than in the (311)B and (100) ones. We found a strong dependence of the luminescence energy of QW's on the interruption time at the GaAs-to-InGaAs interface. This luminescence shift is explained in terms of the segregation profile change for due to the surface morphology evolution during growth interruption, which was monitored by the recovering of the RHEED specular beam intensity.

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