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Composition modulation in GaInNAs quantum wells: Comparison of experiment and theory

M. Herrera and D. González[a)]
Departamento de Ciencia de los Materiales e Ingenierìca Metalúrgica y Química Inorgánica, Universidad de Cádiz, Apartado 40, 11510 Puerto Real, Cádiz, Spain

M. Hopkinson, M. Gutiérrez, P. Navaretti, and H. Y. Liu
Department of Electronic and Electrical Engineering, University of Sheffield, Mappin Street, Sheffield S1 3JD, United Kingdom

R. García
Departamento de Ciencia de los Materiales e Ingenierìca Metalúrgica y Química Inorgánica, Universidad de Cádiz, Apartado 40, 11510 Puerto Real, Cádiz, Spain

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Composition modulation observed in GaInNAs quantum wells imposes an important handicap to their potential application within optical components, particularly as the indium and nitrogen contents are increased to reach longer wavelengths. In this paper, we compare our experimental results of phase separation in GaInNAs quantum wells grown at different temperatures with recent theoretical models of spinodal decomposition from the literature. This comparison has shown that the regular solution approximation, which explains the higher composition modulation compared to GaInAs samples, provides a more appropriate explanation of GaInNAs decomposition than the usual delta lattice-parameter approximation. Transmission electron microscopy shows no composition modulation contrasts with the chemical sensitive 002 dark field reflection and a strong increase in the intensity of the strain contrasts observed with 220 bright field reflection as the growth temperature increases from 360 to 460 °C. These observations can be explained by an uncoupling between N and In composition profiles forming separate In-rich and N-rich regions according to the regular solution approximation model. We therefore believe that the compositional fluctuations in GaInNAs are not only due to GaInAs decomposition, but that an uncoupled modulation of the III and V elements is also present. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1866491]

I. INTRODUCTION

One of the major interests in ternary and quaternary III-V semiconductor alloys has been the ability to continuously vary the energy gap and the strain through changes in composition in order to achieve advantageous band structure for opto- and microelectronic devices. Generally, quaternary alloys offer the possibility of selecting both the band-gap energy and the lattice parameter of the epitaxial layer independently, which constitutes an important advantage for heterostructure design. This is indeed the case of the GaInNAs system, which has received a great deal of attention in the last few years because of the giant band-gap bowing[1] that occurs when adding small amounts of N to the GaInAs alloy. However, the successful performance of such devices depends on the ability to grow epitaxially good-quality single-crystalline layers of these materials on a particular substrate. Although in principle, a random atomic distribution in the layers could be supposed, in practice a temperature-dependent miscibility gap exists in most solid solutions, leading to the apparition of composition fluctuations. The difficulties in the growth of homogeneous layers are even more complex in quaternary alloys, where two compositional degrees of freedom are present (for a review see Ref. 2).

In the past, composition modulation in semiconductor layers has been attributed to a process of spinodal decomposition. Following Cahn,[3,4] who proposed a theoretical model for bulk binary metal alloys, several authors have extended the theory of spinodal decomposition to semiconductor alloys.[5,6] However, the predicted critical temperatures were abnormally low. Since then, several authors have applied theoretical calculations to epitaxial layers, considering also the elastic energy that results from the lattice mismatch of a layer grown on a substrate.[7,8] Nowadays, theoretical models tend to include the specific characteristic of the epitaxial growth, such as the atom adsorption from the vapor to the surface, surface diffusion instead of bulk diffusion, or surface morphological undulation. From the earlier treatment of Malyshkin and Shchukin,[9] several models that introduce different kinetic instabilities coupled with morphological instabilities have been developed (see Ref. 10 for a review). However, as pointed by Millunchick and Srolovitz,[10] compositional modulations can occur even in the absence of morphological modulations if the semiconductor alloy shows a tendency for phase separation or spinodal decomposition.

There are great differences between composition modulation in metal alloys, the subject of initial theoretical studies and those of semiconductor alloys. In metals, phase separation occurs as a bulk process with large aging times, while composition modulation in semiconductor is, in general, a

[a)]Electronic mail: david.gonzalez@uca.es

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surface diffusion process and not very dependent on post-growth annealing treatments.[2] Therefore, it is not ideal to use the term spinodal to describe these two distinct types of phase separation. Nevertheless, most of the recent models applied to semiconductor compositional modulation introduce in some form the thermodynamic formulation derived from the spinodal treatment. In the following, we use the term spinodal to talk about the chemical interaction between the atoms that constitute the semiconductor alloy, although we consider that the diffusion process occurs in a thin subsurface layer during the growth and not in the bulk. Certainly, it is true that thermodynamic models do not explain completely the very complex epitaxial growth process, but they form an excellent starting point to describe the phases implicated in the compositional modulation, especially if morphological instabilities are not present. In this paper, we work with the most recent thermodynamic models proposed in the bibliography to explain the compositional modulation behavior in GaInAsN/GaAs heteroepitaxy.

In its simplified form, there are two approaches for the calculation of the critical temperature for spinodal decomposition in semiconductor alloys. One approach considers the regular solution (RS) model for the estimation of the chemical energy of the alloy, while the other takes into account the delta lattice-parameter (DLP) approximation introduced by Stringfellow.[5] The difference between these models stems from the fact that whereas in the regular solution model the chemical energy is a function of the composition of the alloy, the chemical potentials of the binaries and the interaction parameter between them, in the DLP approximation *only* the composition of the alloy is considered.

In this work, we have studied composition fluctuations in GaInNAs quantum wells grown at different temperatures. Our experimental results have been compared with two theoretical approximations for the prediction of critical temperatures for spinodal decomposition in the quaternary alloy GaInNAs: an analysis considering the DLP approximation[11] and the one using the RS model.[12] The validity of the utilization of the DLP for the GaInNAs alloy is questioned and the influence of the introduction of N in the composition fluctuations of the alloy is investigated both theoretically and experimentally.

II. EXPERIMENT

The growth of GaInNAs quantum wells (QWs) was performed in a VG V80H molecular-beam epitaxy (MBE) system equipped with an Oxford Applied Research HD25 radio-frequency plasma source for N. The N composition was controlled by monitoring the optical intensity of the atomic N plasma emission. All the quantum wells were deposited on (001)-exact GaAs substrates. The composition of the quantum wells is 0.38% of In and 0.023% of N. The samples consisted of five quantum wells, 8 nm thick sandwiched between $GaAsN_{0.007}$ barriers. The GaInNAs QWs were grown at different temperatures in each of the four samples considered: 360, 400, 440, and 460 °C, and annealed for 60 mins at 660 °C. A structure with a single $Ga_{0.7}In_{0.3}As$ quantum well grown at 480 °C has also been considered.

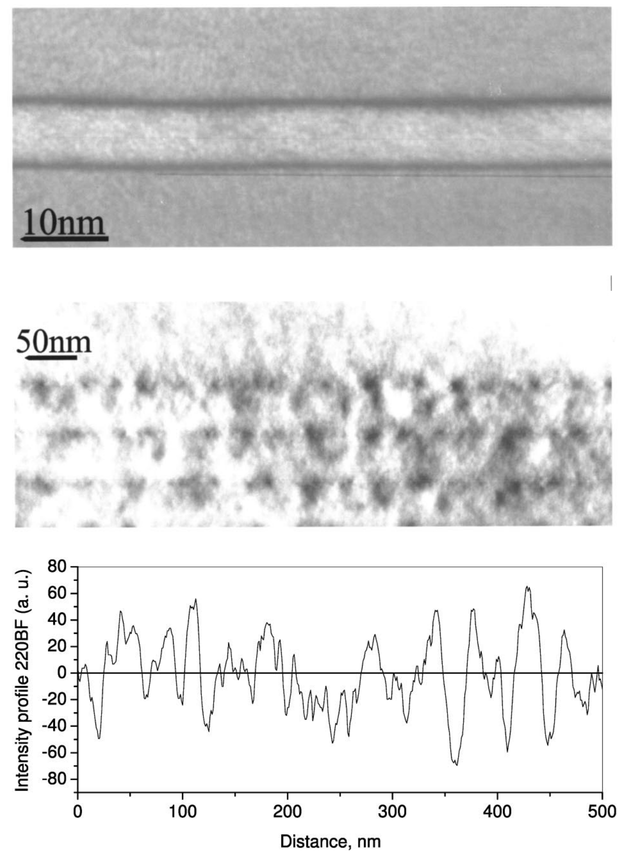

FIG. 1. TEM micrographs of the GaInNAs structures grown at 440 °C obtained with the 002DF reflection (a) and the 220BF reflection (b). Intensity profile along the upper QW with the 220BF reflection (c).

Samples were prepared for transmission electron microscopy (TEM) by mechanical thinning followed by ion milling for cross-section TEM (XTEM) and planar view observation TEM (PVTEM). The TEM study was performed using JEOL 1200EX and JEOL 2011 transmission electron microscopes.

III. RESULTS AND DISCUSSION

Our transmission electron microscopy results with 002 dark field (DF) reflection have shown that the GaInNAs QWs samples grown at 360, 400, and 440 °C are perfectly flat. Furthermore, 220 bright field (BF) reflection shows an absence of dislocations or other structural defects in all of the samples. However, on increasing the growth temperature to 460 °C, the wells become undulated and some threading dislocations have been observed. The most important feature observed in these samples is the absence of contrast modulations with the chemical sensitive 002DF reflection and the apparition of high strain contrasts along the wells in all the structures with the 220 BF reflection (see Fig. 1 for the structure grown at 440 °C). These contrasts are more pronounced when increasing the growth temperature and we can associate these strain fields with differences regarding the nominal compositions of the QWs.[13] This correlation between strain contrasts and growth temperature is more noticeable when taken into account the intensity profiles of the strain contrasts. Figure 2 shows the intensity amplitude of the strain contrasts normalized to the GaAs layer in the upper well of

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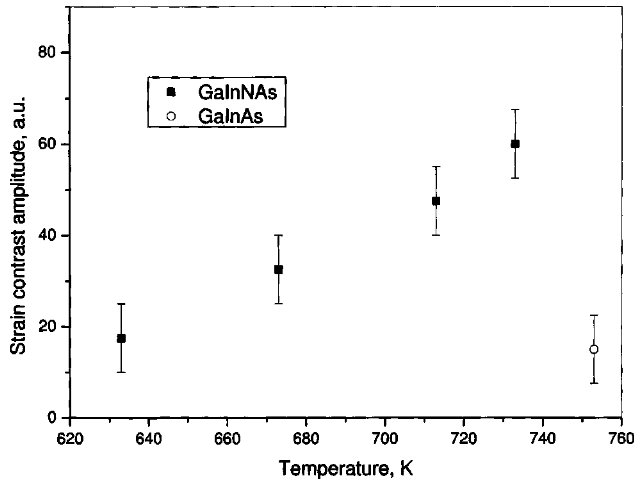

FIG. 2. Intensity amplitude of normalized contrasts from micrographs acquired with 220BF in the upper well of the four GaInNAs samples vs growth temperature. For comparison, the circle shows the intensity contrast amplitude for GaInAs.

the four samples considered, taken from micrographs acquired with the 220BF reflection. As it can be observed, the amplitude of the profiles is higher when increasing the growth temperature from 360 to 400, 440, and 460 °C. In the same figure, we add the intensity amplitude registered for the $Ga_{0.7}In_{0.3}As$ sample growth at 480 °C. As we can observe, the extent of composition modulation for the GaInNAs samples is remarkably high with respect to the $Ga_{0.7}In_{0.3}As$ sample, the latter being only comparable to the GaInNAs grown at the lowest growth temperature (360 °C)

Planar view TEM specimens reveal that strong composition contrasts are aligned to the $\langle 110 \rangle$ directions. It is well known that the lateral composition modulation can line up in the $\langle 100 \rangle$ direction in which the elastic strain energy is minimum or in the $\langle 110 \rangle$ directions where the atomic diffusion is faster.[14] This fact highlights the importance of the surface diffusion process and therefore the deposition variables for this system, but also reinforces our hypothesis that the profiles obtained in the $\langle 110 \rangle$ cross-section micrographs can offer a good and realistic image of the strength of composition modulation in the alloy. For all of the samples, except the highest-temperature (460 °C) sample, compositional modulation occurs without undulations in the QW structure. It has been reported that morphological instabilities are in many cases the origin of phase separation due to a different size or mobility of the atoms in the alloy.[15–17] Although the sample grown at 460 °C shows that a higher composition modulation is coupled with a surface modulation, the samples grown at lower temperatures present perfectly flat interfaces with a well-defined composition modulation. This is evidence to support the intrinsic trend of GaInNAs alloys towards phase separation, probably due to spinodal decomposition. Although it is true that kinetic factors, such as growth rate, must be included in a more realistic model and that it is probable that phase separation occurs within a few monolayers close the surface, it is also clear that thermodynamical aspects, such as chemical interactions between atoms, play an important role in the description of phase separation in this system.[18]

Let us now compare our experimental results of phase separation with the most recent models developed for our system. Several factors are usually included in the calculation of the miscibility gap for epitaxial semiconductor layers: the chemical energy, the coherency strain energy, the gradient energy, and the elastic energy due to the lattice mismatch. Some of these factors are sometimes considered negligible. In this work, we are going to consider the theoretical equations for the occurrence of spinodal decomposition proposed for the GaInNAs alloy, based on the RS model[12] and the DLP model.[11] Both models consider as the main factors for the calculation the chemical energy of the alloy and the strain energy due to the lattice mismatch with the substrate. However, these approaches differ from each other essentially in the treatment of the chemical energy of the system. These expressions for the chemical energy read, respectively, as follows:

$$G_{\text{chem}}^{\text{RS}} = xy\mu_{\text{InN}}^{0} + x(1-y)\mu_{\text{InAs}}^{0} + (1-x)y\mu_{\text{GaN}}^{0} + (1-x)\times(1-y)\mu_{\text{GaAs}}^{0} + x(1-x)y\alpha_{\text{InN-GaN}} + xy(1-y)\alpha_{\text{InAs-InN}} + x(1-x)(1-y)\alpha_{\text{InAs-GaAs}} + (1-x)y(1-y)\alpha_{\text{GaAs-GaN}} + A, \quad (1)$$

$$G_{\text{chem}}^{\text{DLP}} = -K(\Delta a)^2 a^{-4.5} + A, \quad (2)$$

where

$$A = RT[x\ln(x) + (1-x)\ln(1-x) + y\ln(y) + (1-y)\ln(1-y)], \quad (3)$$

where μ^0 is the chemical potential of the pure binaries, a is the lattice constant, α is the interaction parameter between the binary compounds, K is a constant for all systems, R is the universal gas constant, and x and y are the In and N composition, respectively.

As it can be seen, the expression of the regular solution model includes the calculation of the chemical potentials and the interaction parameters of the pure binaries. The DLP, on the other hand, allows us to simplify the calculation, approximating these terms by the expression $K(\Delta a)^2 a^{-4.5}$. We are going to evaluate the validity of this approximation for our alloy GaInNAs. The second term included in these models of spinodal decomposition is the elastic energy due to the lattice mismatch with the substrate, which in this case is

$$G_{\text{strain}} = \Omega(C_{11} - C_{12})(C_{11} + 2C_{12})\varepsilon^2/C_{11}, \quad (4)$$

where Ω is the molar volume, C_{11} and C_{12} are the elastic constants, and ε is the elastic strain.

We have computed both models. For the RS model, the chemical potentials of the pure binaries are estimated using the expression,

$$\mu_{AB}^{0} = H_{AB} - TS_{AB} + \int_{298.15}^{T} C_P^{AB} dT - T\int_{298.15}^{T} C_P^{AB} dT/T, \quad (5)$$

where H and S are the enthalpy and the entropy at the standard state, respectively, and C_p is the specific-heat capacity at a constant pressure.

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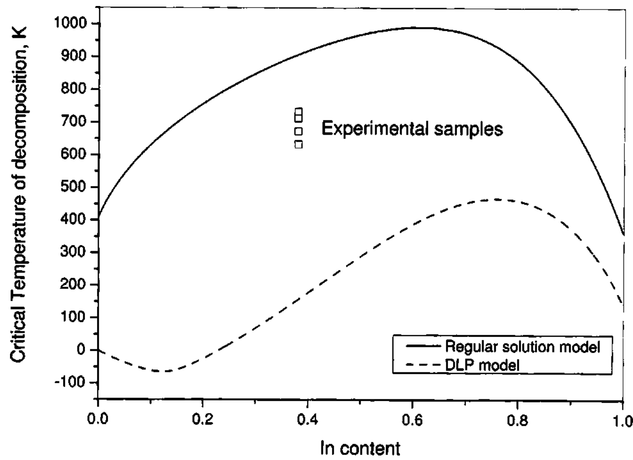

FIG. 3. Critical temperatures for spinodal decomposition in $Ga_xIn_{1-x}As_{0.977}N_{0.023}/GaAs$ structures using the DLP and RS models. Our experimental results are included.

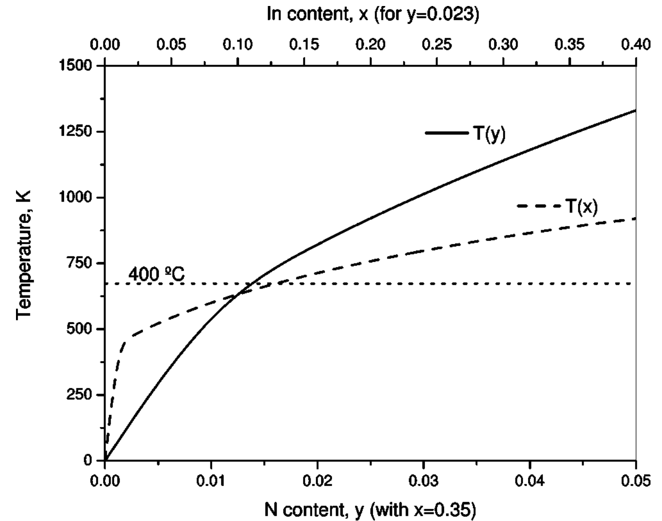

FIG. 4. Critical temperatures for spinodal decomposition of the GaInAsN/GaAs samples using the RS model for In composition (keeping the N content constant at the value of our samples, $y=0.023$) and for N composition (keeping the In content constant at $x=0.38$).

We have taken the experimental dependences of the specific-heat capacity with the temperature from a different source[19,20] than the ones[21] used by Asomoza *et al.* in Ref. 12. This has produced deviations in the slope of the curve of the critical temperature with regard to that proposed by Asomoza *et al.*[12] Moreover, we have found a high sensitivity in the calculated critical temperatures when changing slightly the expressions considered by us for the specific-heat capacity. This means that we should be very careful with the absolute values of the critical temperatures obtained in this calculation, given that the expression for C_p has been obtained experimentally and could be affected by some errors. However, these variations do not affect the main tendencies and conclusions obtained in the present work.

Figure 3 shows a plot of the theoretical calculations of critical temperatures for spinodal decomposition from the DLP and RS models. Our experimental results are also included. As can be clearly seen, the curve using the DLP model shows critical temperatures for spinodal decomposition considerably lower than that corresponding to our samples. Therefore, this model predicts that our GaInNAs quantum wells should be homogeneous, a conclusion that differs from our experimental observations. On the other hand, the RS model is in good agreement with our results, showing that spinodal decomposition should have taken place in our samples. It follows that the DLP model of Stringfellow[5] is not appropriate for the calculation of the miscibility gap in GaInNAs alloys.

In view of the results above, we focus only on the RS model in the subsequent discussion. Figure 4 exhibits a plot of the critical temperatures for spinodal decomposition versus variations in In (keeping the N content constant at the value of our samples, 0.023) and in N (keeping the In content constant at 0.38). As can be observed, the curve corresponding to the N is much higher than that for the In. Higher critical temperatures in the miscibility gap mean higher instability with regard to spinodal decomposition. In this way, it seems that N is the alloy constituent which will show a higher tendency to suffer spinodal decomposition and phase separation. This fact is a direct consequence of the higher interaction parameter of InAs–InN and GaAs–GaN regarding the other combinations.[12] The higher interaction parameters of $\alpha_{GaAs-GaN}$ (2.16×10^5 J/mole) and $\alpha_{InAs-InN}$ (1.46×10^5 J/mole) regarding $\alpha_{InN-GaN}$ (4.53×10^4 J/mole) and $\alpha_{InAs-GaAs}$ (1.89×10^4 J/mole) explain the trend of nitrogen to separate into GaN and other phases. Following this reasoning, the RS model predicts a separation into N-rich regions (GaN) and In-rich regions (InAs). It is this additional separation of the N component which can explain the higher composition modulation of GaInNAs with regard to the GaInAs samples. The interaction parameters involved in the spinodal decomposition of GaInAsN samples are an order a magnitude higher that in the GaInAs case.

Up to now, several authors have already reported experimental evidence of lateral composition modulations in the GaInNAs QWs using 002DF image analysis,[22–24] although this has been only attributed to In segregation. Albrecht*et al.*[23] reported In-concentration fluctuations of ~5% while Patriarche *et al.*[24] found a fluctuation of ~7% between In-depleted and In-rich zones in a similar quaternary alloy using normalized 002DF intensity profiles for the as-grown samples. In both reports, there was no indication of lateral fluctuations of N concentration. For the annealed samples, they also do not observe fluctuations in the 002DF profiles. However, spinodal decomposition curves predict a high thermodynamical trend for N fluctuations. The experimental results by TEM show that no modulation contrasts appear with the chemical sensitive reflection 002DF, whereas a high degree of lateral deformation with the 220BF reflection is observed. The chemically sensitive 002DF reflection under kinematical conditions depends mainly on differences between the atomic scattering factors of III and V elements, being relatively independent of sample thickness for these contents.[22] The intensity of the 002DF reflection (I_{002}) for a zinc-blende structure is given by

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$$I_{002} = C|F|^2 = 4C(f_{\mathrm{III}} - f_{\mathrm{V}})^2 = 4C[x f_{\mathrm{In}} + (1-x) f_{\mathrm{Ga}} - y f_{\mathrm{N}} - (1-y) f_{\mathrm{As}}]^2, \quad (6)$$

where C is a factor that depends on thickness and imaging conditions, F is the structure factor, and f are the atomic scattering factors. If we imagine a region of the material A, where the In and N contents are increased by an arbitrary quantity Δx and Δy, respectively, it is necessary for there to be a region B nearby in which these compositions are decreased in the same proportion. To obtain the same dark field intensity in the two different regions, A and B, we need that

$$I_{002}^{A}(x+\Delta x, y+\Delta y) = I_{002}^{B}(x-\Delta x, y-\Delta y). \quad (7)$$

Solving, we obtain

$$\Delta x = -\frac{f_{\mathrm{As}} - f_{\mathrm{N}}}{f_{\mathrm{In}} - f_{\mathrm{Ga}}}\Delta y = -2.208\Delta y. \quad (8)$$

Therefore, to obtain the invariant 002DF intensity profiles in the GaInNAs QWs that we have observed experimentally, it is necessary that an increase in the N composition Δy must be accompanied of a simultaneous decrease in the In content Δx in the proportion $\Delta x \approx -2.21\Delta y$. The formation of N-rich regions with a depletion of the In content into In-rich and N-depleted regions obeying the rule $\Delta x \approx -2.21\Delta y$ could explain the absence of contrast modulation with the 002DF reflection. Consequently, we suggest that there are two compositional modulation profiles, one for N and another for In, which is 90° out of phase to the In distribution. In addition, the uncoupling of the In and N composition profiles could also explain the high strain fields that appear with the 220BF reflection. For this reflection, the image is dominated by the strain field contrast (**gR**), where **R** is the displacement vector regarding the regular position of the atoms. In a simplified form, if we consider two close coherent regions with different composition, for example, A and B, the displacement field for this situation is

$$\mathbf{R} = \frac{3K\delta}{3K + 2E(1-\nu)}, \quad (9)$$

where K is the bulk modulus for the phase A, E and ν are the elastic modulus and Pisson's ratio of phase B, and δ is the lattice misfit. Although the compositional profile follows a sinusoidal form, we can calculate the maximum δ that corresponds to the higher contrast as

$$\delta = \frac{a^{A}(x+\Delta x, y+\Delta y) - a^{B}(x-\Delta x, y-\Delta y)}{a^{B}(x-\Delta x, y-\Delta y)}, \quad (10)$$

where a^A and a^B are the lattice parameters of regions A and B. If we imposed the condition derived from Eq. (8), we can obtain the lattice misfit amongst regions with the same chemical contrast in the 002DF condition (see Fig. 5). In the same figure, we also show the lattice misfit for the GaInAs case ($y=0$) with the same Δx. As we can see, the lattice misfit for the same amplitude of In modulation Δx is always higher in the GaInNAs samples than in the GaInAs samples. This result is in agreement with the contrast amplitude ratio experimentally measured with the 220BF reflection (see Fig. 1). The system would show homogeneous contrasts with the 220BF (no strain fields) if the In and N composition profiles were in phase (matched conditions, $\Delta x \sim 3\Delta y$), that is to say, the N-rich regions coincide with In-rich regions. Only the formation of separate In-rich and N-rich regions could explain the higher strain fields observed with the 220BF and the absence of fluctuation contrast with the 002DF reflection.

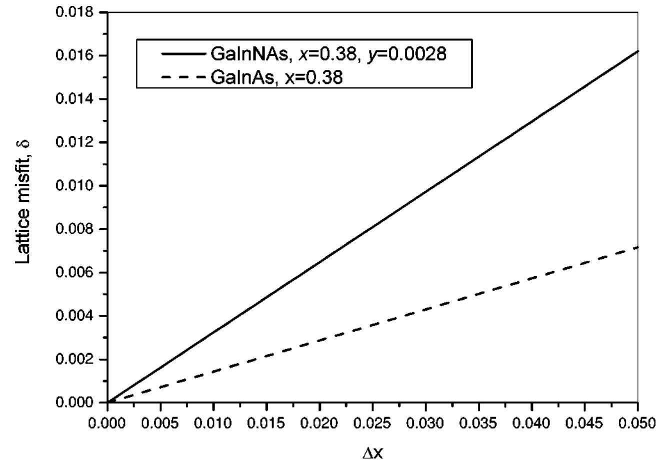

FIG. 5. Lattice misfit for the GaInNAs regions with a constant 002DF chemical contrast. In the same figure, we also show the misfit for the GaInAs case ($y=0$) with the same Δx.

We should point out that the proportion $\Delta x \approx -2.21\Delta y$ needs not to be precisely followed in order to observe an absence of contrast with the 002DF reflection. Probably there exists a range of proportions of indistinguishable contrasts for which we are not sensitive. The use of 002DF intensity contrasts has been studied in detail for GaInAs. Cagnon *et al.*,[25] Glas,[26] or Patriarche *et al.*,[24] using different models for 002DF image analysis, have shown that there does not exist a perfect agreement between experimental and theoretical results, although their experimental results indicate that the relative 002DF contrasts are more sensitive than theoretical predictions when In contents are in the 30%–40% range. As example, Patriarche *et al.*[24] using an experimental fit of normalized contrast propose contrast sensitivity for indium measurement of 0.02%. With regard to the effect on the 002DF contrast of N incorporation, there have been no experimental studies in the bibliography from which we can estimate the relative sensitivity of the technique. However, if we use kinetical model and compare this to known observations we believe that we should be able to detect differences of 0.5% of N for the same In content (relative intensities higher than 10%). The large difference between the N atomic factor and the other atoms (Ga, In, and As) explains this relatively high sensitivity. These predictions are confirmed experimentally through 002DF TEM analysis of GaAsN/GaAs QWs with ~1% of nitrogen which show remarkably strong contrast with respect to the GaAs barriers. In addition, as the same way that that elastic strain raises the experimental 002DF contrast with regard to theoretical predictions,[25] we suppose that the nitrogen 002DF analysis could be more sensitive because the inhomogeneity of N incorporation introduces a higher strain field. However, it should be noted that we do not propose exact measurement of composition modulation

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amplitude in our work, but rather we think that contrast differences should be more appreciable for this N and In composition.

There are few works in the bibliography concerning measurements of In and N in QWs with the required compositional sensitivity on a nanometer scale. Using electron-energy-loss spectroscopy[27] (EELS) or annular dark field scanning transmission electron microscopy[28] (STEM) there have been no studies reported which can resolve the N distribution to the required compositional and spatial sensitivity. However, for N-rich samples, the existence of a composition profile, which is 90° out of phase, has been confirmed using STEM energy dispersive x-ray (EDX) elemental maps. Bullough *et al.*[29] confirm the presence of differentiated cells rich in In, Ga, and As atoms that tend to become In-rich towards the cell center separated by thin second phase regions, with show an increase in the Ga and N contents, and a deficiency in In and As. These results are consistent our experimental findings.

The comparison between the experimental and theoretical results implies a phase-separation trend towards InGaAs–GaN regions. Therefore, the phase separation of the GaInNAs QWs is different with regard to that of the GaInAs QWs. In the GaInAs samples, the composition modulation is governed by the lower InAs–GaAs interaction parameter. For the GaInNAs samples, there are two compositional modulations: one for the N (V element) and another for the In (III element) which are 90° out of phase. The higher degree of modulation in GaInNAs compared to GaInAs is due to uncoupling between the In and N profiles, because of the higher interaction parameter that governs this decomposition.

It is well known that In modulation occurs in GaInAs structures and also the existence of N modulation in GaAsN (Refs. 30 and 31) has been observed. We have therefore examined possible mechanisms, which might account for the formation of a 90° out-of-phase compositional contrast. One possible explanation is that during the growth process, chemical bonding aspects are dominating at the growing surface, which favor Ga-N bonds instead of In-N bonds[32] and this state is then frozen in during the nonequilibrium epitaxial growth process. The natural trend of In atoms to form a modulation at the surface of GaInAs structures could produce nitrogen accumulation in the Ga-rich phase. However, local strain effects have higher importance in bulk, favoring In-rich nearest-neighbor configurations of N. Therefore, the frozen nonequilibrium bulk state can be moved towards to the equilibrium bulk state by certain annealing processes.[33] The diffusion of the N- to In-rich regions therefore decreases the elastic N-induced perturbation, leading to smaller band-structure changes. This behavior explains very well the blueshift produced by the annealing of the GaInNAs structures, although the very low bulk diffusion coefficients ($\sim 10^{-19}$ $\mathrm{cm^2/s}$) limit this process and therefore high post-growth annealing temperatures are required (typically $\sim 800\ ^\circ\mathrm{C}$).

Finally, it should be noted that a discrepancy can be found between our experimental results and critical temperature curves for spinodal decomposition. We have observed an increase in the intensity of the strain contrasts when raising the growth temperature of the GaInNAs quantum wells, a fact that has been related to an increase in the magnitude of composition fluctuations in the alloy for higher growth temperatures. However, theoretical models predict a higher stability with regard to spinodal decomposition for higher growth temperatures. This disagreement occurs because we do not consider the kinetics of the spinodal model. In a real system, kinetic factors, such as the influence of growth temperature on the surface diffusion of the different constituents of the alloy, can play an essential role in the formation of composition fluctuations, explaining the experimentally observed enhancement of the phase separation when raising the growth temperature. Therefore, it is concluded that, to better explain our experimental results of the phase separation in the GaInNAs quantum wells, it is indispensable to consider models of spinodal decomposition that include kinetic factors.

IV. CONCLUSIONS

We have studied by transmission electron microscopy the composition fluctuation of GaInNAs quantum wells grown at different growth temperatures and compared these results with two theoretical models of spinodal decomposition proposed for this alloy. The TEM results show an increase in the intensity of the strain contrasts observed with the 220BF reflection for GaInNAs wells when increasing the growth temperature and an absence of chemical sensitive contrast with the 002DF reflection. A comparison between theoretical calculations based on the DLP and RS models of spinodal decomposition has showed that only the second one agrees with our experimental results and that therefore the DLP approximation is not appropriate for the GaInNAs alloy system. In the RS model, differences between the interaction parameters lead to a separation of In and N, giving a higher composition modulation with regard to the GaInAs samples. The formation of two uncoupled composition profiles of In and N can explain the absence of contrast modulations with the chemical sensitive 002DF reflection and the high lateral strain fields evidenced with the 220BF reflection. A thermodynamic model using the RS approximation gives an excellent prediction of the phases involved in the composition fluctuations of GaInAsN alloys, although it is necessary introduce kinetical parameters to explain the dependence of the composition amplitude with temperature.

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