Fine speckle contrast in InGaAs/InP systems: Influence of layer thickness, mismatch, and growth temperature

F. Peiró
LCMM. Departament Física Aplicada i Electrònica, Universitat Barcelona, Diagonal 645-647, 08028 Barcelona, Spain and Serveis Científico-Tècnics, Universitat Barcelona, Lluis Solé i Sabarís 1-3, 08028 Barcelona, Spain

A. Cornet, A. Herms, and J. R. Morante
LCMM. Departament Física Aplicada i Electrònica, Universita Barcelona, Diagonal 645-647, 08028 Barcelona, Spain

S. A. Clark and R. H. Williams
Department of Physics, and Astronomy, University of Wales, College of Cardiff, Box 913, Cardiff CF1 3TH, United Kingdom

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This work is focused on the study of the fine speckle contrast present in planar view observations of matched and mismatched InGaAs layers grown by molecular beam epitaxy on InP substrates. Our results provide experimental evidence of the evolution of this fine structure with the mismatch, layer thickness, and growth temperature. The correlation of the influence of all these parameters on the appearance of the contrast modulation points to the development of the fine structure during the growth. Moreover, as growth proceeds, this structure shows a dynamic behavior which depends on the intrinsic layer substrate stress.

INTRODUCTION

The InGaAs-InAlAs heterostructures on InP have emerged as one of the most challenging systems for the design of optoelectronic devices based on the quantum confinement of charge carriers in wells. To assure the reliability of such devices as well as to achieve the level of integration desired for future industrial requirements, the structures have to be grown with optimum quality, with the different epilayers perfectly homogeneous and with abrupt and planar interfaces between them.[1] This approach is not easily satisfied as suggested from recent microstructural transmission electron microscopy (TEM) observations of planar view specimens of several III-V matched and slightly mismatched semiconducting alloys. Several authors[2–6] have reported two types of quasiperiodic contrast consisting of two orthogonal sets of dark bands along the ⟨010⟩ directions, one of them, like a tweed structure, with a periodicity Λ of hundreds of nm, named coarse modulation, and the other with a wavelength λ of tens of nm, named fine modulation or fine speckle. These features have been observed in III-V compounds of different compositions grown by liquid phase epitaxy (LPE) and vapor phase epitaxy (VPE).[2–5] Thermodynamic calculations[7] predict an immiscibility gap for ternary and quaternary III-V grown by quasi-equilibrium techniques like LPE and VPE, and the phase separation induced by spinodal decomposition has frequently been argued as the source of the patterns observed. So, at first it was thought that nonequilibrium techniques such as molecular beam epitaxy (MBE), could be used to grow metastable alloys which would be immiscible if grown by other techniques.[7] Nevertheless, the appearance of both the coarse and fine modulations on III-V alloys grown by MBE has also been reported.[5,6,8,9] To explain the individual origin of the two types of contrast modulation, two different approaches are envisaged. One point of view relates the coarse structure to surface atomic diffusion during growth and the fine speckle to bulk atomic diffusion when cooling to room temperature after growth.[2,3] The other point of view argues that the fine modulation occurs by phase separation on the surface when the layer is growing, and that the coarse modulation may form to accommodate stresses associated with the fine scale structure.[5,6] Some contradictions have emerged not only about the range of growth temperatures and compositions for which the contrast modulations appear but also about the modulation wavelength that these structures should have according to the atomic diffusion lengths involved in the growth process.[2–6]

Despite the large number of works on this subject, to our knowledge, a systematic study of the influence of the main parameters on the final configuration of layers with contrast modulation has not yet been done. The aim of this article is to carry out such a study in order to provide further insight into the origin of both structures. The work is focused on matched and mismatched InGaAs layers grown by MBE on InP substrates. Although our efforts have been mainly devoted to the fine scale contrast, some previous results about the appearance of the coarse modulation will be also commented on. First, we have studied the evolution of the fine structure as layer thickness increases. Second, we show the dependence of the fine speckle on the layer mismatch. Finally, the effects of the growth temperature on the appearance of the fine contrast are reported. The modification of the fine modulation wavelength according to these parameters gives valuable new experimental results for the understanding of the occurrence of such structure.

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EXPERIMENTAL DETAILS

We have studied three groups of samples according to the three different parameters listed above. Group A consists of $In_xGa_{1-x}As$ layers ranging in thickness (t) between 0.3 and 2 μm, grown with fixed composition $x=54.3\% \pm 0.2\%$ at the same growth temperature $T_g = 515\,°C$. Group B is formed by four samples changing in composition (x), with x values of 54.1%, 54.6%, 59%, and 62.5%, respectively. For all these samples the layer thickness was 0.5 μm and $T_g=515\,°C$. Finally, to investigate the influence of the growth temperature, five samples with $x=54\%$ and $t=1.8$ μm were grown at T_g=450, 475, 500, 525, and 550 °C.

All epilayers were grown using a VG Semicon V8OH MBE system. Growth was carried out on InP (100) semi-insulating Fe-doped substrates. The substrates were etched prior to growth, at 50 °C in H_2SO_4:H_2O:H_2O_2 prepared in the ratio 7:1:1.

Plan view [100] TEM specimens were prepared by mechanical polishing of the substrate side to a thickness of 30 μm. A final thinning was carried out by ion milling using low Ar^+ intensities in a cooled stage to avoid In island nucleation. Thicker layers had to be etched to make samples thin enough for TEM studies. First, samples were etched from the substrate side and second from the layer surface. By controlling the etching times, we were able to obtain TEM specimens with their thinnest regions (about 0.3 μm) at different distances from the interface. Observations were performed using a Hitachi H-800 NA microscope operating at 200 keV. The wavelength measurements were obtained by average on different TEM micrographs and the statistical error was approximately of 15%.

RESULTS AND DISCUSSION

The most important feature of the group A is the existence of a contrast modulation roughly along both the [001] and [010] directions. The tweed-like structure has a wavelength (Λ) in the range of hundreds of nm which decreases as layer thickness increases. Moreover, this coarse pattern is located in a region of 0.5 μm above the interface. The details of our observations of this coarse modulation have been already published[8,9] so we do not consider them here.

In these samples (group A) the presence of the fine scale modulation is also noticeable. In Fig. 1(a), corresponding to sample A1 (t=0.29 μm), we can observe the coexistence of the two structures under g=022 two beam condition. Both modulations change in the same way when the imaging conditions are changed. Under g=040, only the dark bands along [001] are visible [Fig. 1(b)] whereas for g=004 the lines on [010] remain in strong contrast [Fig. 1(c)]. Similar results were obtained for the rest of samples A. The wavelength λ of the fine speckle evolved towards lower values as layer thickness increased (see graphic Fig. 2), ranging λ between 300 and 170 Å. Figure 3 corresponds to sample A3 (t=0.49 μm). By comparing Fig. 1 with 3, the reduction of the value of the periodicity when thickness increases is evident for both the fine and coarse structures.

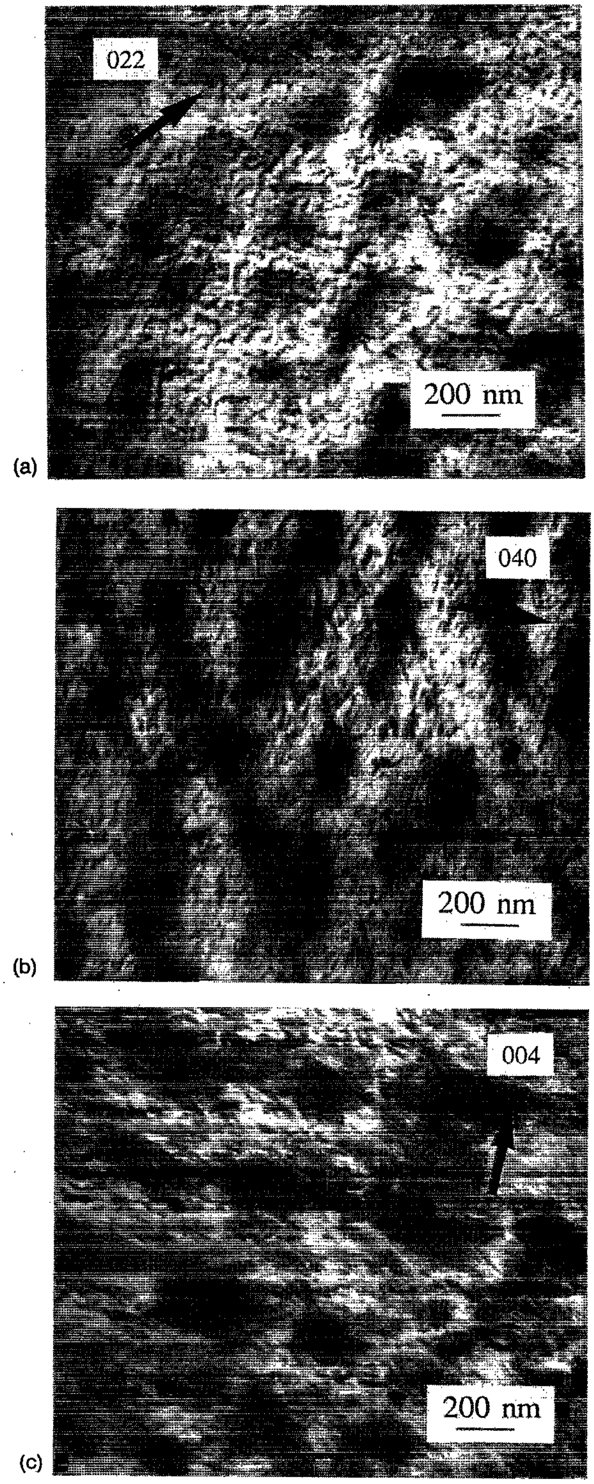


FIG. 1. Plan view micrographs of sample A1 (t=0.3 μm). (a) The coarse and fine modulations are in strong contrast under g=022. (b) When the sample is imaged under g=040 for both structures, only the set of dark bands orthogonal to g remain visible. (c) For g=004 the bands parallel to g vanish.

We have also studied the evolution of the coarse contrast upward from the interface in thick InGaAs layers. In

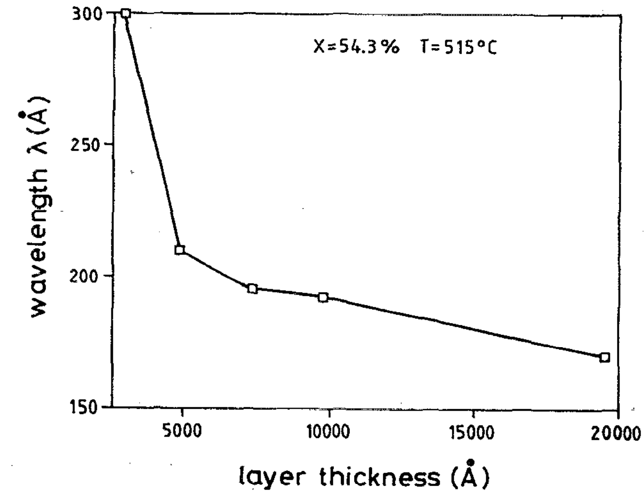


FIG. 2. Plot showing the evolution of the fine wavelength λ as a function of the InGaAs layer thickness t.

the upper regions of the layer the coarse structure disappeared.[9] Figure 4(a) corresponds to a thin foil of sample A5, milled so as to have the most suitable region for TEM examination near to the layer-substrate interface. There, the coarse and fine modulations were both present. In contrast, imaging the top of the epilayer [Fig. 4(b)], only the fine structure was observed. We must point out that although the spots on the transmission electron diffraction (TED) pattern presented some diffuse intensity, additional satellite spots were not observed in any sample.

For the second group of samples, the parameter subject to variations is the initial intrinsic mismatch (f) of the epilayer, resulting from the change in the In fraction, from $x=54.1\%$ to $x=62.5\%$. The fine contrast modulation is present in all the layers and shows essentially the same features as for the samples of group A. We also found a reduction of the fine speckle wavelength as mismatch increases, as shown in Fig. 5.

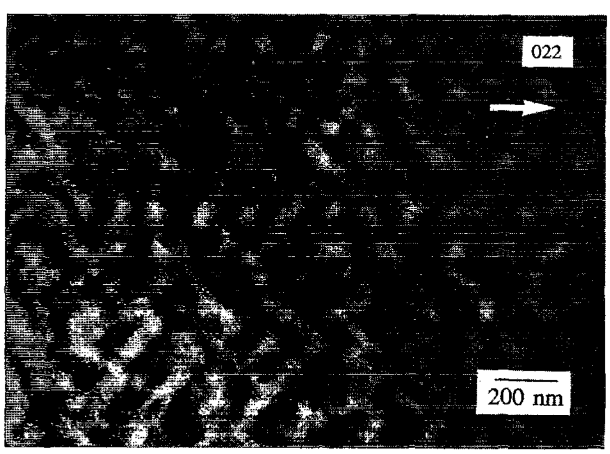


FIG. 3. Micrograph obtained from sample A3 ($t=0.74$ μm) under dynamical $g=022$ bright field condition. The reduction of the wavelengths Λ and λ is evident.

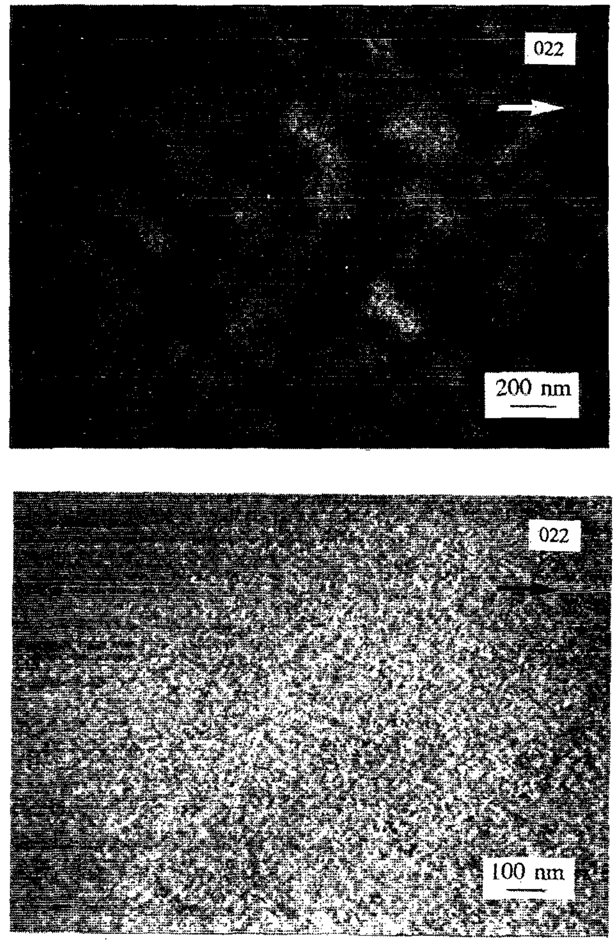


FIG. 4. Plan view $g=022$ images of the sample A5 ($t=2\mu$m). (a) Coarse and fine modulations near the interface layer substrate. (b) Top of the InGaAs layer. The coarse modulation has disappeared whereas the fine modulation is still present.

Finally, we investigated the influence of the growth temperature on the appearance of the fine structure in InGaAs layers. For this purpose, five specimens were grown at a fixed composition $x=54\%$ and a layer thickness $t=1.8$ μm, but at different growth temperatures, rang-

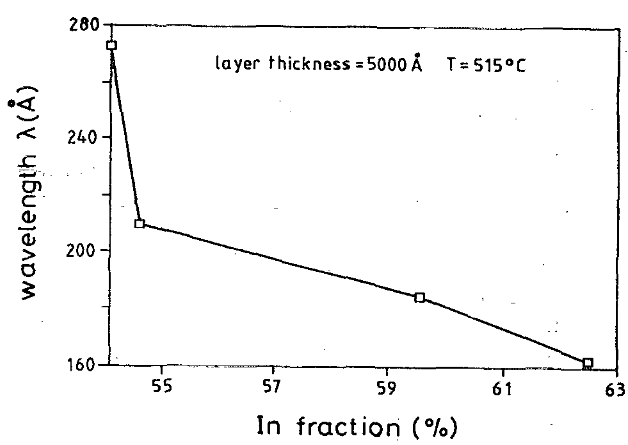


FIG. 5. Graphic of the dependence of the fine scale modulation wavelength on the layer/substrate mismatch f.

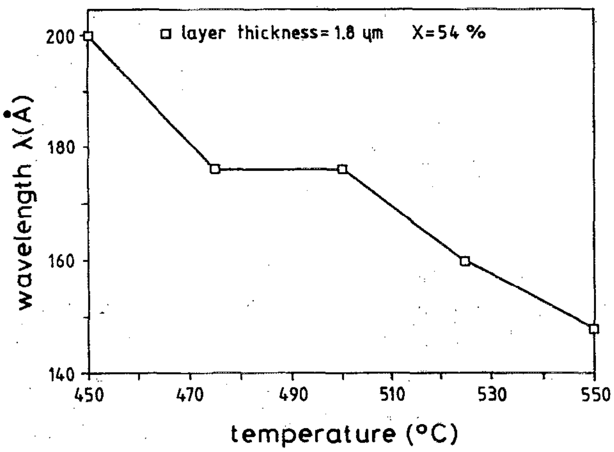


FIG. 6. Evolution of the wavelength λ vs the growth temperature.

ing from 480 °C to 550 °C. All the samples show the speckle contrast under $g=022$ and $g=0\bar{2}2$ two beam condition, with a wavelength ranging between 200 and 150 Å. Figure 6 shows the evolution of the wavelength λ when increasing T_g. We can observe a slight influence of the T_g values for the range studied.

To date, several authors have attempted to establish the origin of the fine contrast modulation and much controversy has been evident from their opposed conclusions. The issue of greatest debate is whether the fine speckle develops during the growth or during the cooling process. According to the presence or not of satellite spots on TED patterns, Norman and Booker[3] have suggested that the fine modulation develops in two different ways. When such satellites are evident, they associate them to a well defined speckle induced by phase separation produced by bulk atomic diffusion during growth. Conversely, when spots are not observed, they consider alloy decomposition as occurring in the specimen when cooling after growth. If these assumptions were correct, the fine modulation of our samples should have been developed when cooling, since we never saw satellite spots. However our cross sectional observations make this approach inconsistent. When imaging the sample under $g=0\bar{2}2$ two beam condition [Fig. 7(a)], a weakly columnar structure with contrast modulation orthogonal to the growth direction [100] was clearly visible. The contrast disappears in $g=200$, indicating that there was no modulation in the growth direction. These results are in agreement with other works. Based on the appearance of the fine structure for the (100) and (111) substrate orientation, McDevitt *et al.*[5] argued that if decomposition were to occur during cooling after growth, the modulation would dominate along the growth direction, since the relaxation of decomposition induced stresses would be easier normal to the substrate (the shortest dimension). In view of these results we consider that the decomposition of the alloy during the cooling process is not the cause of the fine modulation. Besides, the evolution of the wavelength modulation with the layer thickness (Fig. 2) suggests a dynamic nature of the fine speckle, i.e., the modulation may modify its λ value as growth proceeds.

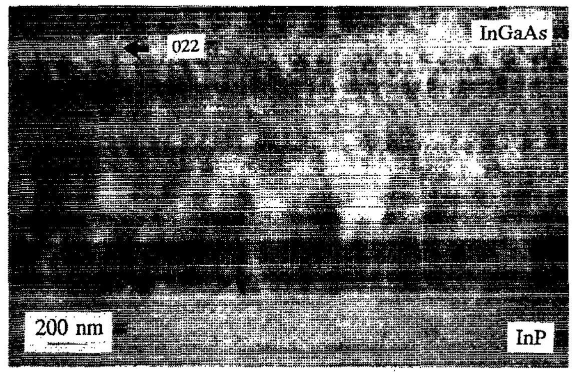
Another point of discrepancy is the range of compositions within which the fine modulation occurs. Following the experiments reported in quaternary alloys as $In_xGa_{1-x}As_yP_{1-y}$ grown by LPE and MBE lattice matched to InP, and taking into account that the surface atomic diffusion lengths depend on substrate orientation, method of growth, and growth temperatures, Mahajan *et al.*[6] and McDevitt *et al.*[10] suggest that if the growth conditions are identical, the wavelength of the fine speckle contrast should be approximately the same even in layers with different composition. This does not apply to our samples of group B, since we have shown that the value of λ depends on the layer composition, i.e., the x values (Fig. 5), despite the fact that the growth conditions are exactly the same for all the epilayers. This apparent contradiction may be explained by considering that it is the stress induced by the intrinsic mismatch between the layer and the substrate that strongly affects the occurrence of the fine modulation during growth and not the x variations themselves.[11,12] Therefore, $In_xGa_{1-x}As_yP_{1-y}$ layers matched to InP and grown under identical conditions would present the same λ values whatever the x and y values,[10] whereas x,y variations introducing a mismatch in the system would induce also wavelength modifications.

Once assumed that the fine modulation develops during growth and that its characteristics are influenced by the stress, we agree with other authors[11,12] that the origin of the fine speckle is a not random distribution of III-V type atoms. This idea accounts for the fact that the fine structure has been universally observed in III-V alloys for a wide range of compositions. Following the assumptions of Letardi *et al.*,[11] the fine speckle could be explained by the differences in bond lengths of the atoms constituting the layer. As a consequence, the four possibilities[11] for the tetrahedral unit cell are not equally probable and hence, the atoms distribute to decrease the Gibbs free energy of the system leading to nonuniformities on the lattice parameter, even in matched layers. The existence of this equilibrium state is thought to be due to the ability of such a configuration to accommodate two dissimilar bond lengths (In–As and Ga–As, 7% different) in a coherent fashion[11] leading to In rich or Ga rich zones. Following this hypothesis, as may be intuitively expected, the stress-free bond length distribution in the layer would be seriously affected by the stress in the case of mismatched structures, as already suggested by the theoretical models of Letardi *et al.*[11] and Glas.[12] Thus for coherent epitaxial growth, the elastic energy involved in growing lattice mismatched layers on a given substrate would determine the preferential sites of incorporation of the atoms when they reach the surface. Since the elastic energy of the system rises as layer thickness increases, a redistribution in order to minimize the energy of mixing will be favored, conferring dynamic behavior to the fine structure. In fact, we have provided experimental evidence that the parameters directly related to the increase of the energy (namely, layer thickness and stress induced by the x values) have a significant effect on the fine modulation. As far as the growth temperature is concerned, we notice that this parameter has the lowest influence on the fine modulation wavelength. In any case, the slight tendency of λ towards lower values as T_g increases is in contradiction with the results of McDevitt *et al.*,[10] who found an increase in the wavelength with T_g in InGaAsP matched layers. More experiments in a wider range of T_g values should be performed to explain the influence of the growth temperature.

Let us now briefly refer to the relation between the coarse and the fine contrast modulations. McDevitt *et al.*[10,13] have recently argued that the fine modulation is the direct cause of the coarse modulation. They suggest that the fine speckle evolves by two-dimensional phase separation occurring at the surface during growth. Conversely, they found the coarse structure to be an artifact of thin foils, appearing as a result of the accommodation of the biaxial stress state, which produces a breakup of the foil surface into domains when the underlying substrate is removed. Our results show that it is possible to have fine modulation and not the coarse structure in specimens with the underlying substrate totally removed (Fig. 4). McDevitt *et al.*[10,13] also gave an explanation to account for the fact that the coarse structure was not observed in some cases. They argued that the coarse modulation would not be observed in thicker and thinner regions of the specimen since, for thin regions, the stress associated with the fine speckle is not high enough for buckling to occur, whereas the thicker regions require much higher buckling stresses. In our case, the observations were made on specimens with the same foil thickness (approximately of 0.3 μm) but at different distances from the interface. Therefore, the extinction of the coarse structure on the top of the layer is not related to the thickness of the region under examination. This assumption is corroborated by XTEM observations. In Fig. 7(a), besides the fine modulation, there are some dark bands along the [100] direction in a region 0.5 μm above the interface, whereas in the upper regions, only the fine speckle remains. Figure 7(b) corresponds to the 0.3-μm-thick InGaAs layer, for which both the fine and coarse modulations are present in the whole film. These results show that a buckling of the thin film due to stresses associated with the fine modulation does not originate the coarse structure.

CONCLUSIONS

We have studied the appearance of the fine contrast modulation present in InGaAs layers. We have shown the dependence of this modulation on the layer-substrate mismatch. Our results show a tendency of the modulation wavelength towards lower values as growth temperature, layer thickness, and mismatch increase. We have found that the wavelength depends on the layer thickness and mismatch more strongly than it does on the growth temperature. The hypothesis of element distribution according to differences in bond lengths accounts for the evolution of λ with x,t. Our results have demonstrated that the buckling of the thin TEM foil due to stresses associated with the fine modulation is not the cause of the coarse pattern.



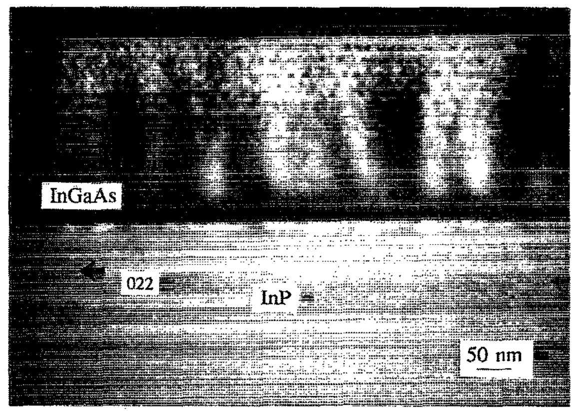


FIG. 7. (a) XTEM [0$\bar{1}$1] image of sample A5 ($t=2$ μm). Under $g=022$, the fine contrast is modulated orthogonally to the growth direction and the coarse modulation is visible only up to 0.5 μm. (b) Sample A1 ($t=0.3$ μm), dark field $g=022$. The coarse modulation covers all the layer.

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