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Current status of atomic ordering and phase separation in ternary and quaternary III-V compound semiconductors

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ABSTRACT The status of current understanding of atomic ordering and phase separation in epitaxial layers of ternary and quaternary III-V compound semiconductors is briefly reviewed. The formation of Cu Pt-type ordered variants and the effects of their coexistence on diffraction patterns are discussed. A model is proposed for the formation of domain boundaries in ordered layers that involves ordering at the surface during growth. In addition, it is shown that the fine scale speckle microstructure observed in many ternary and quaternary epitaxial layers occurs by surface spinodal decomposition at the growth temperature. It is argued that the coarse contrast modulations that coexist with the fine scale structure are a result of the accommodation of the two-dimensional strain associated with the latter.

1. INTRODUCTION

The ternary and quaternary III-V compound semiconductors are scientifically very interesting as well as are technologically relevant materials. They find extensive applications in light emitters, detectors, microwave devices, etc. Many applications are based on the fact that the bandgaps of these materials can be tailored by changing their composition. The most dramatic example of this is the InP/InGaAsP system where the composition of the InGaAsP active layers in light emitters can be altered to emit at wavelengths which are compatible with the spectral properties of fused silica fibers used in lightwave communication systems and layers are still lattice matched to the underlying InP substrates.

Structurally, these materials consist of two interpenetrating FCC units which are displaced from each other by 1/4 <111>. One of the units is occupied by group III atoms, where as group V atoms are located on the second unit. An obvious, but interesting, question is whether or not the atoms on the two FCC sub-lattices are randomly distributed? The answer is an emphatic "no". Within the last few years, several authors have reported the existence of long range order in different III-V ternary and quaternary layers (Kuan et al. 1985, Jen et al. 1986, Nakayama and Fujita 1986, Shahid et al. 1987, Norman et al. 1987, Gomyo et al. 1987, Ihm et al. 1987, Ueda et al. 1987, McKernan et al. 1988, Shahid and Mahajan 1988, Mahajan and Shahid 1988, and Jen et al. 1989). An interesting aspect of these observations is that ordering is seen in materials which are completely miscible at the growth temperature, such as $Ga_xAl_{1-x}As$ (Kuan et al. 1985), and those which show a miscibility gap, for example $GaAs_{0.5}Sb_{0.5}$ (Jen et al. 1986, Ihm et al. 1987 and Norman et al. 1987), $In_{1-x}Ga_xAs$ (Nakayama and Fujita 1986, Shahid et al. 1987, Shahid and Mahajan 1988 and Mahajan and Shahid 1988), $In_{0.5}Al_{0.5}As$ (Norman et al. 1987), $Ga_{0.51}In_{0.49}P$ (Gomyo et al. 1987, Ueda et al. 1987 and McKernan et al. 1988), $Ga_{0.37}In_{0.63}As_{0.82}P_{0.18}$ (Shahid and Mahajan 1988) and in $As_{1-x}Sb_x$ (Jen et al. 1989). With the exceptions of the observations of Jen et al. (1986) and Nakayama and Fujita (1986), the studies on immiscible systems indicate that ordering occurs on two of the four {111} planes (Shahid et al. 1987, Shahid and Mahajan 1988 and Mahajan and Shahid 1988). In the case of $In_{1-x}Ga_xAs$ and $In_{1-x}Ga_xAs_yP_{1-y}$ materials these planes are parallel to $\{111\}_A$ facets of a thermal decomposition induced pit observed on the surface of the underlying (001) InP substrate (Shahid and Mahajan 1988). As a result of ordering, the periodicity along the <111> directions is doubled.

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Concomitantly, the majority of the above compositions exhibit phase separation (Henoc et al. 1982, Mahajan et al. 1984, Treacy et al. 1985, Charsley and Deol 1986, Chu et al. 1985, Norman and Booker 1985, Shahid and Mahajan 1988 and Mahajan and Shahid 1988). The phase separated layers of $In_{0.53}Ga_{0.47}As$ and $In_{1-x}Ga_xAs_yP_{1-y}$ of different compositions, grown on (001) InP substrates, exhibit two types of contrast modulations (Henoc et al. 1982, Mahajan et al. 1984, Treacy et al. 1985 and Chu et al. 1985). A fine scale speckle contrast whose period is ~ 15nm is observed along the <100> directions lying in the (001) plane. In addition, these layers show coarse contrast modulations, resembling a basket-weave pattern, whose wavelength is ~ 125 nm. These modulations are also oriented along the <100> directions lying in the (001) plane (Henoc et al. 1982, Mahajan et al. 1984, Treacy et al. 1985 and Norman and Booker 1985). Henoc et al. (1982), Treacy et al. (1985) and Norman and Booker (1985) have argued that the both types of contrast modulations result from spinodal decomposition. Since the wavelength of the coarse modulations is too large for them to develop by bulk diffusion, Launois et al. (1982) have suggested that the coarse modulations could occur by surface spinodal decomposition at the growth temperature. The speckle contrast, on the other hand, could evolve from phase separation occurring during cool down from the growth temperature (Norman and Booker 1985). Mahajan et al. (1984) have presented an alternative explanation. They suggest that, because the diffusion is extremely slow in these materials, the fine scale structure could develop by phase separation, while the coarse contrast modulations may be due to the accommodation of strains associated with the fine scale modulations.

In the present paper, recent observations pertaining to the influence of growth conditions on ordering in (Ga, Al) In P epitaxial layers grown by organo-metallic vapor phase epitaxy (OMVPE), dopant diffusion-induced disordering of the ordered structures and the occurrence of phase separation in Ga In P and (Ga, Al) In P layers are presented. Also, the origins of composition-independent wavelength of the speckle contrast and rectilinear boundaries seen in $In_{1-x}Ga_xAs_yP_{1-y}$ layers grown by liquid phase epitaxy (LPE) (Mahajan et al. 1984) are discussed. Finally, an attempt has been made to synthesize various diverse aspects of ordering and phase separation in ternary and quaternary epitaxial layers of III-V compound semiconductors and thus develop a coherent picture of the microstructures of these materials.

2. ATOMIC ORDERING

Gomyo et al. (1988) and Shahid et al. (1988) have observed that the growth rate and temperature have a considerable influence on the perfection of long range order in $(Ga_xAl_{1-x})_{0.5}In_{0.5}P$ layers grown on (001) GaAs substrates by OMVPE. This is apparent from electron diffraction patterns shown in Fig. 1. Figure 1(a), obtained from the sample grown at the high growth rate, shows diffuse intensity bands that are parallel to the growth direction and pass through superlattice spots corresponding to the (111) and (11$\bar{1}$) ordered variants. On the other hand, the superlattice spots in Fig. 1(b), low growth rate, are sharp and diffuse intensity bands are absent.

When the (001) reciprocal sections of the above two samples were examined using transmission electron microscopy, diffraction patterns reproduced as Fig. 2 were observed. In addition to the diffraction spots corresponding to the zinc-blende structure, extra spots in Fig. 2(a) occur at $\pm$ (1/2, 1/2, 0) and equivalent positions. The pattern in Fig. 2(b) is different from that shown in Fig. 2(a), shows extra spots at ($\pm$ 1, $\pm$ 1, 0) and equivalent positions and is characteristic of a CuAu-I type tetragonal unit cell with its c-axis parallel to the [001] growth direction.

Following Shahid et al. (1987) and Shahid and Mahajan (1988), the observations regarding ordering in Fig. 1 can be rationalized. For the sake of discussion, let us consider the occurrence of long range order in $Ga_{0.5}In_{0.5}P$ layers grown on (001) GaAs substrates. Shahid et al. (1987) have suggested that the observed double period along the <111> direction can be developed by assuming that the alternating {111} layers within the group III sub-lattice are Ga- and In-rich. Subsequently, Shahid and Mahajan (1988) have argued that the proposed arrangement provides an optimal way to accommodate differences in tetrahedral radii of different atoms occupying a particular sub-lattice. This suggestion is borne out by observations on ternary and quaternary epitaxial layers exhibiting the Cu Pt-type ordering shown above (Shahid et al. 1987, Norman et al. 1987, Gomyo et al. 1987, Ihm et al. 1987, Ueda et al. 1987, McKernan et al. 1988, Shahid and Mahajan 1988 and Jen et al. 1989). However, the Cu Au-I type order observed in $Ga_{0.3}Al_{0.7}As$

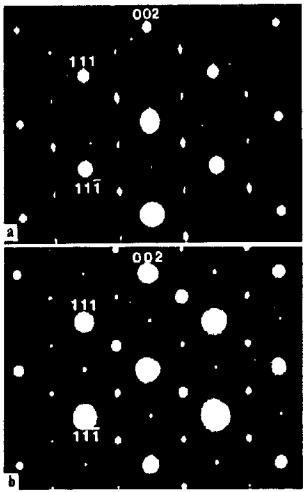

Fig. 1. Electron diffraction patterns observed from (Ga, Al)InP layers grown at (a) high, and (b) low growth rate. Note the presence of diffuse intensity bands in (a) that are parallel to the [001] growth direction and pass through superlattice reflections.

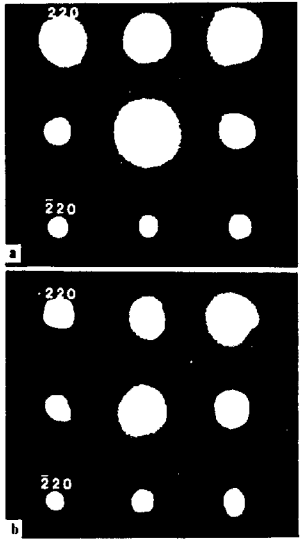

Fig. 2. Electron diffraction patterns observed from (Ga, Al)InP layers grown at different temperatures and growth rates on (001) GaAs substrates by low pressure OMVPE: (a) growth temperature is 650° C and growth rate is 1.08 μm/hr., and (b) growth temperatrue is 680° C and growth rate is 0.67 μm/hr. Note that in addition to the spots of the zinc-blende structure, extra spots occur at ± (1/2, 1/2, 0) and equivalent positions in (a) and (± 1, ± 1, 0) and equivalent positions in (b).

layers grown on (110) GaAs substrates by OMVPE and molecular beam epitaxy (Kuan et al. 1985) cannot be rationalized on this basis because the tetrahedral radii of the Ga and Al atoms are essentially identical. It could be that, in addition to the tetrahedral radii differences, another factor may be important in ordering that pertains to the tendency of two types of atoms occupying a particular sub-lattice to phase separate at the surface during growth. This suggestion is reasonable in view of the fact that the composition modulations, normal to the growth direction, are observed in $Ga_{0.3}Al_{0.7}As$ layers grown on (001) GaAs substrates (Kuan et al. 1985).

Recently, Kondow et al. (1988) and Shahid et al. (1989) have suggested that the extra spots in Fig. 2 do not represent new ordered structures and could arise because of imperfections in the two ordered variants, i.e., (111) and (11$\bar{1}$). Let us assume that, as the layer grows on the (001)

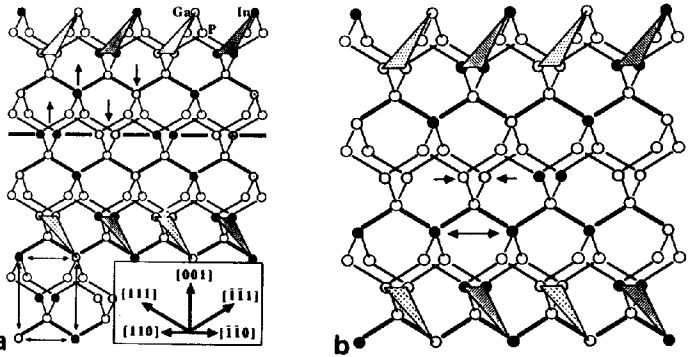

Fig. 3. Schematics showing atomic arrangement in a $Ga_{0.5}In_{0.5}P$ layer containing (111) and (11$\bar{1}$) variants of Cu Pt-type ordering. (a) The (111) and (11$\bar{1}$) variants of the trigonal structure define an abrupt interface in the (001) plane. In the volume enclosed within the (001) planes immediately one above and one below this interface, the (110) planes occur as pairs of Ga Ga(P) and In In(P) layers (indicated by arrows). (b) In this case the (111) and (11$\bar{1}$) variants define a diffuse interface. The material in this interfacial region has a distribution of Ga(4) P, In(4) P, and Ga(2) In(2) P tetrahedral units.

GaAs substrate, initially ordering on group III sublattice of $Ga_{0.5}In_{0.5}P$ occurs on the (111) plane. At some point during the layer growth, the ordered variant may change to (11$\bar{1}$). The switch-over produces a sharp (001) interface between the (111) and (11$\bar{1}$) variants of the trigonal structure that is schematically shown in Fig. 3(a). A close examination of the interfacial region in Fig. 3(a) shows that the (110) planes constitute a sequence of ...Ga Ga In In... double layers in the [110] direction. It is therefore suggested that spots observed at ± (1/2, 1/2, 0) and equivalent positions, Fig. 2(a), could arise from the (001) interfaces between the (111) and (11$\bar{1}$) ordered variants. The fact that these extra spots are observed predominantly in $Ga_{0.5}In_{0.5}P$ layers grown at a higher rate suggests that the (001) interfaces between the two ordered variants may form more readily in these layers.

When $Ga_{0.5}In_{0.5}P$ layers are grown at a low rate, interfaces between the (111) and (11$\bar{1}$) variants need not be sharp. One such interface is schematically shown in Fig. 3(b). Here, the interface is much wider and not so well defined. The interlacing of the two variants produces a platelet of In (or equivalently Ga) atoms. This platelet, like the abrupt interface in Fig. 3(a), lies on the (001) plane. Also, note that unlike the alternate stacking of Ga(3) In(1)P and Ga(1) In(3)P tetrahedral units along the <111> directions of the ordered trigonal structures, the interfacial region in Fig. 3(b) exhibits a distribution of Ga(4)P, Ga(2)In(2)P or equivalently In(4)P and Ga(2)In(2)P tetrahedral units. In other words there is a definite increase in the distribution of Ga(2)In(2)P type units which could produce extra diffraction spots at positions characteristic of the Cu Au-I type ordering. Thus the extra spots observed at ± (1, 1, 0) and equivalent positions in Fig. 2(b) can also be rationalized in terms of an "interfacial defect" between the two variants.

The influence of growth conditions on the perfection of as-grown $(Ga_xAl_{1-x})_{0.5}In_{0.5}P$ (x=0.7-1) layers is also manifested in the domain structures. Fig. 4(a) shows fairly small ordered domains in a sample grown at the higher rate, whereas the average size is fairly large, ~ 0.5 μm, in layers grown at the slow rate, Fig. 4(b).

The evolution of the domains may be rationalized using a schematic shown in Fig. 5. In Fig. 5 ABCD delineate a step of height $\frac{a}{4}$ at the (001) GaAs surface. The surfaces AB and CD are

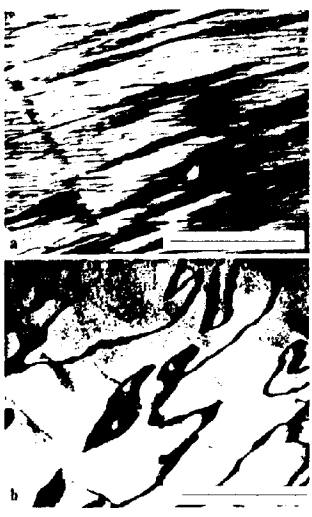

Fig. 4. Domain boundaries observed in (Ga, Al) InP layers grown at different rates on (001) GaAs substrates by low pressure OMVPE: (a) high, and (b) low growth rate. Reflections used for forming images in (a) and (b) are, respectively, 111 and $3\bar{1}1$ superlattice spots.

distinctly different: surface AB is occupied by the As Atoms, whereas Ga atoms define the surface CD. Now imagine that the stepped surface is subjected to a flux of In, Ga and P atoms - the atoms required to grow a layer of $Ga_{0.5}In_{0.5}P$. Following Suzuki et al. (1988) it may be argued that As atoms along the row B would bond to a row of Ga atoms, position G in Fig. 5, and the rows of In and Ga atoms would alternate as shown. The crucial question is which atoms do occupy the row marked X in Fig. 5? If this row is occupied by the In atoms as indicated, then the alternating arrangement of the two types of atomic rows would be commensurate with each other on either side of the step. However, if row is occupied by the Ga atoms, then a volume of Ga(4)P tetrahedral units is produced. As a result of the atomic attachment, the layer will grow and the initial step will assume the position EFGH. Invoking arguments for atomic attachment at positions K and Y similar to the ones used for positions G and X, it can be seen that two distinct situations develop: (i) the $(11\bar{1})$ ordered regions on either side of the step are commensurate with each other, and (ii) the $(11\bar{1})$ ordered regions are separated from each other by the tubes of disordered material. The position of these tubes shift laterally during growth. It is suggested that these tubes are responsible for the domain contrast observed in Fig. 4.

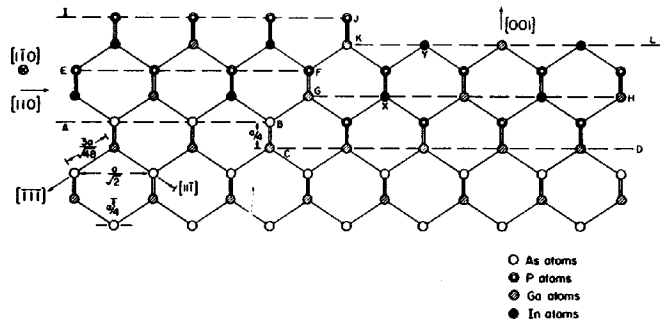

Fig. 5. Schematic illustrating the role of a surface step in ordering and in forming domain boundaries in a $Ga_{0.5}In_{0.5}P$ layer grown on a (001) GaAs substrate.

A word of caution is in order. These domain boundaries are neither anti-phase boundaries in the sense of ordered metallic alloys nor anti-site boundaries that are possible in the growth of GaAs on (001) Si, i.e., the growth of a polar semiconductor on a non-polar one (Kroemer 1987). This stems from the fact that the degeneracy in site assignation that exists in the case of the GaAs growth on (001) Si is not present in the growth of $Ga_{0.5}In_{0.5}P$ on GaAs.

With the exception of a single observation where long range order has been observed in layers grown by LPE (Nakayama and Fujita 1986), ordering has only been seen in layers deposited by vapor levitation epitaxy (VLE) (Shahid et al. 1987 and Shahid and Mahajan 1988), OMVPE (Jen et al. 1986, Gomyo et al. 1987, Ihm et al. 1987, Ueda et al. 1987, McKernan et al. 1988, Gomyo et al. 1988, Suzuki et al. 1988, Jen et al. 1989 and Shahid et al. 1989) and MBE (Kuan et al. 1985 and Murgatroyd et al. 1986). These observations imply that surface mobilities of atoms constituting a layer play a crucial role in the evolution of long range order in ternary and quaternary epitaxial layers.

The ordered structures are thermally quite stable (Gavrilovic et al. 1988) unless they are annealed at temperatures higher than the critical temperature for ordering (Plano et al. 1988). However, they can be disordered at much lower temperatures by dopant diffusion (Gavrilovic et al. 1988 and Plano et al. 1988). If ordering involves atoms on both sub-lattices and dopant atoms diffuse only on one of the two sub-lattices, then complete diffusion-induced disordering cannot be achieved. This suggestion is borne out by the recent work of Plano et al. (1988) who observe spots due to ordering in InGaAsP layers even after the Zn-diffusion.

The presence of long range order in $Ga_{0.5}In_{0.5}P$ and $(Ga_xAl_{1-x})_{0.5}In_{0.5}P$ layers lowers the bandgap of these semiconductors by ~ 90 meV (Gomyo et al. 1988 and Gavrilovic et al. 1988). This differential can be eliminated by dopant-diffusion induced disordering (Gavrilovic et al. 1988).

3. PHASE SEPARATION

As indicated in the Introduction Section, in addition to long range atomic ordering most of the ternary and quaternary compositions exhibit phase separation. To highlight the current status of our understanding of this microstructural inhomogeneity, the following aspects will be considered:

(i) composition-independence of fine scale modulations in InGaAsP layers grown by LPE (Mahajan et al. 1984), (ii) origins of coarse contrast modulations and rectilinear boundaries in InGaAsP epitaxial layers (Henoc et al. 1982, Mahajan et al. 1984 and Treacy et al. 1985), and (iii) the occurrence of phase separation in $(Ga_xAl_{1-x})_{0.5}In_{0.5}P$ layers grown by OMVPE.

Figure 6, reproduced from the work of Mahajan et al. (1984), shows microstructural features observed in InGaAsP layers of different compositions grown in the temperature range of 638-636°(C) by LPE on (001) InP buffer layers; arrows in each micrograph delineate the <100> directions lying in the (001) plane. The emission wavelengths of the layers in Fig. 6(a), (b), (c) and (d) are, respectively, 1.25, 1.3, 1.37 and 1.37 μm. The near-equilibrium growth technique was used to grow the layer shown in Fig. 6(c), whereas the two-phase melt was utilized to grow the layer in Fig. 6(d).

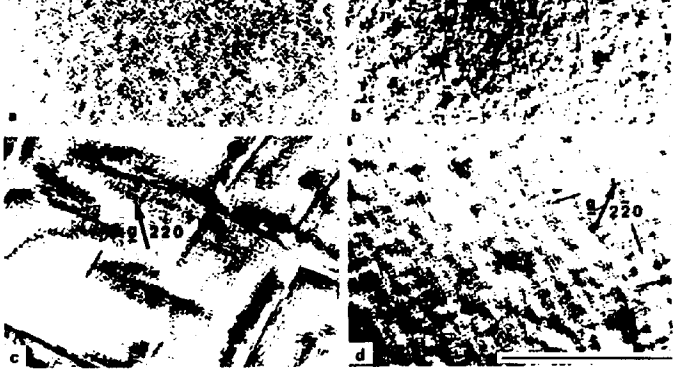

Fig. 6. Electron micrographs obtained from InGaAsP layers of different compositions grown by LPE on (001) InP substrates: (a) $\lambda = 1.25$, (b) $\lambda = 1.30$, (c) $\lambda = 1.37$ and (d) $\lambda = 1.37$ μm. The last two samples differ in the schemes used to grow the quaternary layers. Arrows in each micrograph mark the <100> directions lying in the (001) plane. Marker represents 1 μm.

It is clear that the characteristics of the quasi-periodic fine scale structure are the same in the four micrographs. The wavelength of the fine scale structure is ~ 15nm. The other distinguishing microstructrual features are rectilinear boundaries aligned along the <100> directions, Fig. 6(c), and a barely discernible basket-weave pattern, Fig. 6(d), whose periodicity is ~ 125 nm.

To ascertain the direction of strain associated with the rectilinear boundaries shown in Fig. 6(c), diffraction contrast experiments were carried out, and these results are reproduced as Fig. 7. The arrow marked A identifies the same region in the four micrographs. It is clear that the nearly vertical boundaries, parallel to the [010] direction, are in contrast for the 2$\bar{2}$0, 220 and 400 reflections and exhibit a residual contrast for the 040 reflection. On the other hand, the boundaries which are nearly parallel to the [100] direction are visible for the 2$\bar{2}$0, 220 and 040 reflections and exhibit a residual contrast for 400. These observations indicate that the principal strain is normal to the habit planes of the boundaries.

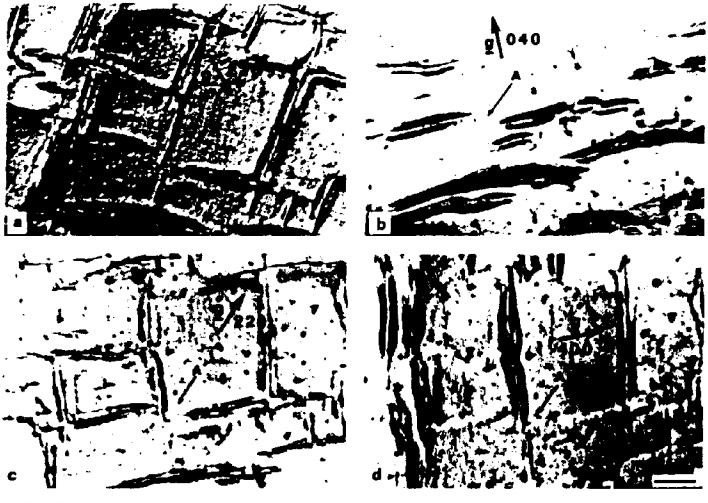

Fig. 7. Micrographs illustrating the contrast behavior of rectilinear features observed in Fig. 6(c). The arrow marked A identifies the same region in the four micrographs. In each case, the plane of the micrograph is ~ (001). Marker represents 1 μm.

Figure 8, reproduced from the work of Treacy et al. (1985), shows well-developed coarse contrast modulations in an $In_{0.72}Ga_{0.28}As_{0.63}P_{0.37}$ layer grown by LPE on (001) InP substrates. Like in the cases of the fine scale structure and the rectilinear boundaries, these modulations also lie along the <100> directions lying in the (001) plane.

Microstructures observed in cross-sections of $Ga_{0.5}In_{0.5}P$ and $(Ga_xAl_{1-x})_{0.5}In_{0.5}P$ layers, grown on (001) GaAs substrates by OMVPE, are shown in Fig. 9. The presence of fine scale structure is again evident in both micrographs. Superimposed on the fine scale structure are coarse contrast modulations aligned parallel to the growth direction.

Lattice matched layers of $In_{0.53}Ga_{0.47}As$ and InGaAsP (Shahid et al. 1987) and InAlAs (McDevitt 1989), grown respectively by VLE and MBE, are phase separated. Taken together the preceding results show that the growth techniques do not appear to influence the occurrence of phase separation in immiscible ternary and quaternary III-V compound semiconductors. This suggestion is at variance with the ideas of Stringfellow and co-workers (Stringfellow and Cherng 1983, Cherng et al. 1984 and Cherng et al. 1986) who have argued that phase separation in immiscible compositions can be prevented by growing them using MBE and OMVPE because of kinetic limitations on the speed with which the constituent atoms can redistribute themselves on the surface during growth.

To rationalize the preceding results, let us first consider the hypothesis that the fine scale microstructure evolves during cool down from the growth temperature (Norman and Booker 1985). If this were true, decomposition in (001) layers should occur along the three <100> directions, the softest directions in the zinc-blende lattice. In addition, the phase separation along

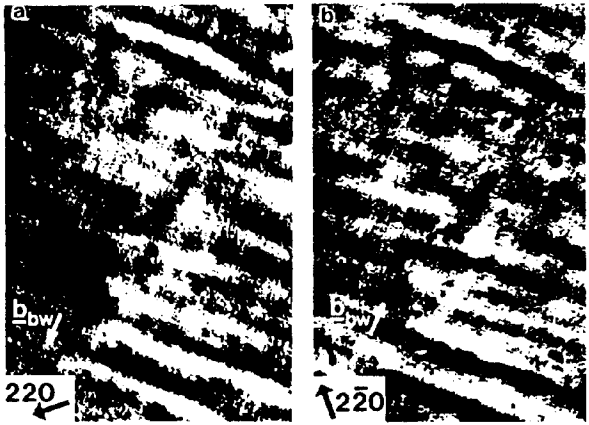

Fig. 8. Dark-field electron micrographs obtained from an $In_{0.72}Ga_{0.28}As_{0.63}P_{0.37}$ sample, approximately 250 nm thick: (a) (220) dark-field and (b) ($2\bar{2}0$) dark-field (Treacy et al. 1985).

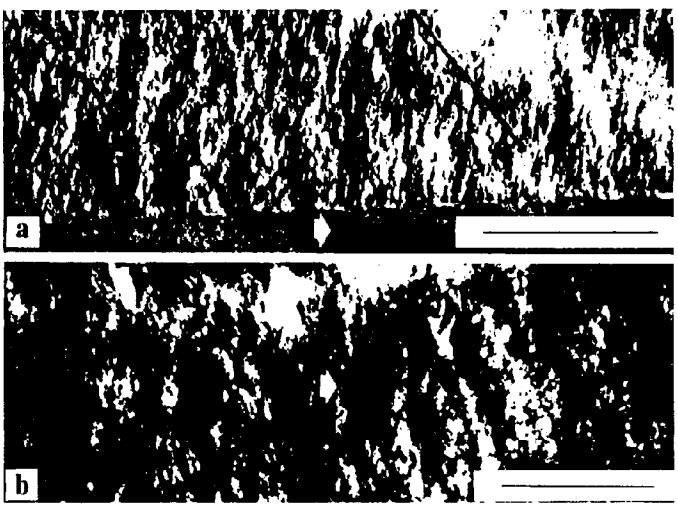

Fig. 9. Electron micrographs obtained from cross-sections of (a) GaInP and (b) (Ga, Al)InP epitaxial layers grown on (001) GaAs substrates by low pressure OMVPE. Plane of the micrograph in each case is – – (110) and arrows mark the 220 operating reflection. Markers in (a) and (b) represent 0.5 and 0.2 μm, respectively.

the growth direction should dominate because transformation-induced strains can easily be relaxed due to its small thickness. This is, however, not borne out by the experimental results of Chu et al. (1985) and Norman and Booker (1985) who do not observe any diffraction contrast in (001) InGaAsP epitaxial layers using 004 reflection. This implies the absence of decomposition along the growth direction, i.e., the decomposition is two-dimensional. Recently, McDevitt (1989) has observed that the decomposition in $In_{0.53}Ga_{0.47}As$ layers, grown ($\bar{1}\bar{1}\bar{1}$)P InP substrates by MBE, occurs only along the directions lying in the growth plane, again indicating the two-dimensional nature of the decomposition-induced microstructure.

The preceding results can be understood if it is invoked that the fine scale microstructures evolves by surface spinodal decomposition at the growth temperature. Launois et al. (1982) were the first ones to propose the idea, but they assumed that the coarse contrast modulations develop in this fashion. Consider the case of (001) layers. In this situation, phase separation would occur along the [100] and [010] directions and two-dimensional strains would develop in the surface regions. Alerhand et al. (1988) have argued that such a system could lower its energy by the formation of elastic-stress domains at the surface. It is proposed that this effect is responsible for the observed coarse contrast modulations. Furthermore, if growth conditions are such that the surface of an epi-layer undergoes permanent periodic distortion, then rectilinear boundaries shown in Figs. 6 and 7 could form.

The wavelength of the fine scale structure that evolves by surface spinodal decomposition would depend on the surface diffusion lengths of atoms constituting the layer. The diffusion lengths, in turn, will depend on the substrate orientation, method of growth and growth temperature. This implies that the wavelength of the speckle contrast should be approximately the same in layers of different compositions, containing the same types of atoms, grown at the same temperature by the same growth technique, an assessment consistent with the results presented in Fig. 6. On the other hand, the composition amplitude of the modulations should depend on the layer composition.

de Cremoux et al. (1981), Stringfellow (1982) and Onabe (1982) have used bulk thermodynamics to compute miscibility gaps in a number of ternary and quaternary III-V compound semiconductors. The work of Chu et al. (1985) indicates that the fit between the compositions undergoing phase separation and those predicted by theory is not good. The observed discrepency is not surprising because the surface thermodynamics has not been used in the computations.

It is apparent from the preceding discussion that both long range atomic order and phase separation in ternary and quaternary layers occur at the surface during growth and could evolve concomitantly.

4. SUMMARY

The characteristics of CuPt-type ordered structures observed in ternary and quaternary epitaxial layers of III-V compound semiconductors are highlighted. It is suggested that these structures evolve to lower the strain energy in systems where one of the two sub-lattices is occupied by atoms differing in their tetrahedral radii. It is shown that "defect structures" resulting from the presence of two {111} ordered variants can produce diffraction effects. In addition, following Suzuki et al. (1988), a model is proposed for the formation of domain boundaries in ordered layers that involves ordering at the surface during growth.

It is argued that the fine scale speckle contrast observed in many ternary and quaternary epitaxial layers develops by surface spinodal decompositon at the growth temperature. On the other hand, the coarse contrast modulations occur to accommodate the two-dimensional strains associated with the fine scale structure.

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