

## Spinodal decomposition ranges of $\text{Al}_x\text{Ga}_{1-x}\text{N}_y\text{P}_{1-y}$ and $\text{Al}_x\text{Ga}_{1-x}\text{N}_y\text{Sb}_{1-y}$ alloys

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Spinodal decomposition ranges of low  $N$ -content  $\text{Al}_x\text{Ga}_{1-x}\text{N}_y\text{P}_{1-y}$  and  $\text{Al}_x\text{Ga}_{1-x}\text{N}_y\text{Sb}_{1-y}$  alloys grown on GaP (001) and GaSb (001) substrates, respectively, are described. The alloys are considered according to the strictly regular approximation. In the analysis we take into account the transformations of the bonds and strain, coherency strain, and elastic energies. The spinodal decomposition ranges of the alloys with compositions  $0 \leq x \leq 0.4$ ,  $0 \leq y \leq 0.05$  are demonstrated up to temperatures of  $1100^\circ\text{C}$ . It is shown that the enlargement of Al and N concentrations dramatically increases the phase separation temperature.

**Keywords:** III-V quaternary alloys; Spinodal decomposition.

Se describe un rango de descomposición espinodal de las aleaciones  $\text{Al}_x\text{Ga}_{1-x}\text{N}_y\text{P}_{1-y}$  y  $\text{Al}_x\text{Ga}_{1-x}\text{N}_y\text{Sb}_{1-y}$  con bajos contenidos de N crecidas sobre los substratos GaP (001) y GaSb (001), respectivamente. Las aleaciones son consideradas en la aproximación de estrictamente regular. En el análisis de los rangos de descomposición espinodal de las aleaciones, se consideró la transformación de enlaces y deformación, deformación de coherencia y las energías elásticas. Se muestran los rangos de descomposición espinodal de las aleaciones con composiciones  $0 \leq x \leq 0.4$ ,  $0 \leq y \leq 0.05$  hasta  $1100^\circ\text{C}$ . Se muestra que el aumento en las concentraciones de Al y N incrementa dramáticamente la temperatura de las separaciones de fase.

**Descriptores:** Aleaciones cuaternarias de compuestos III-V; descomposición espinodal.

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### 1. Introduction

The  $A_x^{III}B_{1-x}^{III}C_y^V D_{1-y}^V$  quaternary alloys with an element having dissimilar properties are widely studied now [1–3]. The study of such alloys began from  $\text{In}_x\text{Ga}_{1-x}\text{N}_y\text{As}_{1-y}$ , which were considered promising new material for active layers of laser heterostructures [4–6]. A small amount of nitrogen in these alloys considerably decreases the fundamental band gap energy [7–9]. Therefore, low  $N$ -content  $A_x^{III}B_{1-x}^{III}N_y^V D_{1-y}^V$  alloys could be used in a number of mid-infrared device applications. As was shown in Ref. 10, all the compositions of interest of the low  $N$ -content  $\text{In}_x\text{Ga}_{1-x}\text{N}_y\text{As}_{1-y}$  alloys are inside the spinodal decomposition range at the growth and lower temperatures. But it is known that stability of semiconductor material characteristics is a main criterion of a device application. Therefore, the study of the thermodynamic stability of other low  $N$ -content  $A_x^{III}B_{1-x}^{III}N_y^V D_{1-y}^V$  alloys with respect to spinodal decomposition is important. The most valuable is an analysis of a spinodal decomposition range or thermodynamically unstable states. Indeed, negligibly small phase separation fluctuations develop if an alloy is inside a spinodal decomposition range. Decomposition depends strongly on self-diffusion processes that are accelerated with a rise in temperature.

According to Gibbs's classic treatment of phase stability, spinodal decomposition begins from the changes that are large in extent but small in degree [11] and develops when

a negligibly small phase separation fluctuation decreases the free energy of an alloy.

$N$ -low content  $\text{Al}_x\text{Ga}_{1-x}\text{N}_y[\text{P}(\text{Sb})]_{1-y}$  belong to  $A_xB_{1-x}C_yD_{1-y}$ -type quaternary alloys with a zinc blende structure. Their crystal lattice consists of the mixed cation and anion sublattices, since two kinds of the atoms fill each of them. As a result,  $A_xB_{1-x}C_yD_{1-y}$  alloys have four types of pairs of the nearest atoms corresponding to the  $AC$ ,  $AD$ ,  $BC$  and  $BD$  binary compounds. Generally, pairs of the nearest atoms are represented as bonds equivalent to the bonds in the  $AC$ ,  $AD$ ,  $BC$  and  $BD$  binary compounds [12]. Therefore,  $A_xB_{1-x}C_yD_{1-y}$  alloys were called the quaternary alloys of four binary compounds. Consequently, bond concentrations describe a chemical composition of the alloy. However, the bond concentrations and atomic composition of the alloy are connected by only three equations:

$$x_{AC} + x_{AD} = x, \quad x_{AC} + x_{BC} = y,$$

$$x_{AC} + x_{AD} + x_{BC} + x_{BD} = 1.$$

Thus, one atomic composition  $x$  and  $y$  corresponds to a vast number of sets of bond concentrations  $x_{AC}$ ,  $x_{AD}$ ,  $x_{BC}$ ,  $x_{BD}$  [13,14]. These bond concentrations are determined by the arrangement of atoms in the crystal lattice. Therefore, an exchange of lattice sites between cations or anions as a result of spinodal decomposition can transform the  $A - C$  and  $B - D$  bonds into the  $A - D$  and  $B - C$  bonds or vice

versa. The transformation of the bonds changes the chemical composition of the alloy. The sum of the cohesive energies of AlN and GaP(Sb) is significantly larger than that of AlP(Sb) and GaN [15]. As was shown in [16,17], the difference between the sums of the cohesive energies can be origin of spinodal decomposition in  $A_xB_{1-x}C_yD_{1-y}$  alloys. This difference for the  $Al_xGa_{1-x}N_y[P(Sb)]_{1-y}$  alloys is significantly larger than that for the “traditional”  $A_xB_{1-x}C_yD_{1-y}$  alloys. Therefore, the free energies of the compounds should play an important role in the spinodal decomposition of the  $Al_xGa_{1-x}N_y[P(Sb)]_{1-y}$  alloys.

The large lattice mismatch between the nitrides and phosphides, as well as antimonides, gives rise to the considerable strain energy of the  $Al_xGa_{1-x}N_y[P(Sb)]_{1-y}$  alloys that can be the next origin of spinodal decomposition. The strain energies of the “traditional”  $A_xB_{1-x}C_yD_{1-y}$  alloys estimated by the valence-force field model are significantly higher than those of the corresponding ternary alloys [18]. The regular solution model also demonstrates the same result [16]. We estimated the strain energies of the “traditional”  $A_xB_{1-x}C_yD_{1-y}$  alloys using the regular solution model with interaction parameters calculated for the ternary alloys by the valence-force field model. The values obtained are almost equal to the strain energies estimated in Ref. 18. Therefore, consideration of the strain energy of the  $Al_xGa_{1-x}N_y[P(Sb)]_{1-y}$  alloys using the regular solution model with the interaction parameters estimated by the valence-force field model seems to be reasonable.

At the same time, spinodal decomposition leads to an occurrence of the coherency strain energy due to the stress between the phases the appeared [19,20]. The shape and orientation of the phases the appeared in an alloy, with composition being in the vicinity of the spinodal decomposition boundary, correspond to the minimal coherency strain energy [19]. An increase in the internal energy owing to the coherency strain energy decreases the spinodal decomposition range. The self-diffusion transfers of the atoms at the initial stage of spinodal decomposition are of the order of a lattice parameter. Therefore, the initial stage of the phase separation in the vicinity of the spinodal decomposition boundary of  $Al_xGa_{1-x}N_y[P(Sb)]_{1-y}$  alloys grown on the GaP(Sb) (001) substrates are considered to be the appearance of two very thin layers. Compositions of the formed layers at the initial stage can be considered to be constant values due to their small thicknesses. As decomposition develops, the atom transfers and thicknesses of the layers increase as and the compositions of the layers vary with the thickness. Afterwards, the difference in the mean concentrations of the phases grows continuously.

A spinodal decomposition temperature also depends on the sign and value of the lattice mismatch between an alloy and substrate [21].

Thus, the spinodal decomposition range of the  $A_xB_{1-x}C_yD_{1-y}$  epitaxial layers depends on the transformation of the bonds and the strain, coherency strain and elastic energies. The spinodal decomposition range of the

$Al_xGa_{1-x}N_y[P(Sb)]_{1-y}$  alloys has not been considered yet. The aim of our paper is to consider the spinodal decomposition region of the  $Al_xGa_{1-x}N_y[P(Sb)]_{1-y}$  layers grown on the GaP(Sb) (001) substrates.

## 2. Theory

The initial stage of the spinodal decomposition of  $Al_xGa_{1-x}N_y[P(Sb)]_{1-y}$  is considered as a change of  $x$ ,  $y$  or both of them in two layers (phases) appearing after the phase separation. The variation of the Helmholtz free energy of the alloy can be written as

$$\delta f = \gamma_1 f(x + \delta x, y + \delta y) + \gamma_2 f(x - \frac{\gamma_1}{\gamma_2} \delta x, y - \frac{\gamma_1}{\gamma_2} \delta y) - f(x, y), \quad (1)$$

where  $\gamma_i$  is the fraction of the  $i$ -th ( $i = 1, 2$ ) layer in the decomposed part of the alloy ( $\gamma_2 = 1 - \gamma_1$ ). The boundary of the spinodal decomposition range is described by the condition  $\delta f = 0$ , [22]. The Helmholtz free energy of the  $Al_xGa_{1-x}N_y[P(Sb)]_{1-y}$  epitaxial layer grown on a substrate in the strictly regular approximation is given by [17]

$$\begin{aligned} f(x, y) = & xy\mu_{AlN}^0 + x(1-y)\mu_{AlP(Sb)}^0 \\ & + (1-x)y\mu_{GaN}^0 + (1-x)(1-y)\mu_{GaP(Sb)}^0 \\ & + x(1-x)y\alpha_{AlN-GaN} + xy(1-y)\alpha_{AlN-AIP(Sb)} \\ & + x(1-x)(1-y)\alpha_{AlP(Sb)-GaP(Sb)} \\ & + (1-x)y(1-y)\alpha_{GaN-GaP(Sb)} \\ & + u_E + RT[x \ln x + (1-x) \ln(1-x) \\ & + y \ln y + (1-y) \ln(1-y)], \quad (2) \end{aligned}$$

where  $\mu_{AlN}^0$  is the chemical potential of the pure binary compound AlN,  $\alpha_{AlN-GaN}$  is the interaction parameter between binary compounds AlN and GaN,  $u_E$  is the elastic energy, and  $R$  and  $T$  are the universal gas constant and absolute temperature, respectively. The strain energy is represented by the interaction parameters between the constituent compounds of the alloy. The interaction parameters were obtained from the strain energy of the corresponding ternary alloys. These strain energies were interpolated, for example, for  $Al_xGa_{1-x}N$  alloys as  $\alpha_{AlN-GaN}x(1-x)$ . The strain energies of the ternary alloys were estimated according to the valence-force field model and experimental results on the distortions of the crystal structure [23,24]. The valence-force field model describes the strained state of the III-V semiconductors using two constants. One constant is the elastic constant of the bond length or bond-stretching constant. Another constant is the elastic constant of the angle between the bonds or bond-bending constant. The strain energy of the unit cell of the binary compound with the zinc blende structure ac-

ording to the valence-force field model is given as

$$u = \frac{1}{2}\alpha \left(\frac{3}{4R^2}\right) \sum_{i=1}^4 [\Delta(r_i^1 \cdot r_i^1)]^2 + \frac{1}{2} \sum_{s=1}^2 \beta^s \left(\frac{3}{4R^2}\right) \sum_{i,j>i} [\Delta(r_i^s \cdot r_j^s)]^2,$$

where  $\alpha$  and  $\beta$  are the bond-stretching and bond-bending elastic constants, respectively,  $R$  is the undistorted bond length,  $\Delta(r_i^1 \cdot r_i^1) = R_i^2 - r_i^2$  and

$$\Delta(r_i^s \cdot r_j^s) = R^2 \cos \varphi_0 - r_i^s r_j^s \cos \varphi$$

are the scalar variations, where  $r_i^s$  and  $r_j^s$  are bond vectors about atom  $s$ ,  $\varphi_0 = 109.47^\circ$  and  $\varphi$  are the angles between the bonds in the unstrained and strained crystals, respectively [25].

The strain energy of the ternary alloys was estimated as the sum of the deformation energies of the tetrahedral cells of the same size. The tetrahedral cells consist of four atoms from the mixed sublattice in the corners and one atom from another sublattice in the center, as shown in Fig. 1. The displacements of the central atoms were calculated by the minimum condition of the deformation energy of the cells.

As stated in Ref. 26, there is no nucleation barrier when an alloy is unstable with respect to negligibly small decomposition fluctuations. Therefore, the energies of the interfaces between the layers appeared, as well as these layers and homogeneous alloy, are not taken into account here. The phase separation in cubic crystals in the vicinity of the spinodal decomposition range should occur in the  $\{001\}$  planes since the relation between stiffness coefficients  $2C_{44} - C_{11} + C_{12} > 0$  is fulfilled [19]. The stiffness coefficients of the  $Al_xGa_{1-x}N_y[P(Sb)]_{1-y}$  alloys were estimated as to be

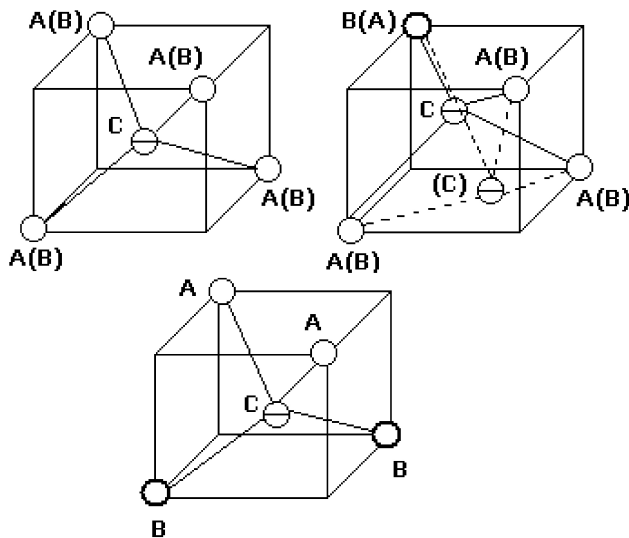


FIGURE 1. 4A(B)1C, 3A(B)1B(A)1C and 2A2B1C tetrahedral cells.

$$C_{ij} = xyC_{ij}^{AlN} + x(1-y)C_{ij}^{AlP(Sb)} + (1-x)yC_{ij}^{GaN} + (1-x)(1-y)C_{ij}^{GaP(Sb)}.$$

Thus, the initial stage of spinodal decomposition of the alloys is considered to be the appearance of two thin layers oriented in the (001) plane. The elastic energy of the alloy grown on the substrate with orientation (001) is given as

$$u_E = \frac{1}{2}\nu(2\sigma_{||}\varepsilon_{||} + \sigma_{\perp}\varepsilon_{\perp}), \tag{3}$$

where  $\nu$ , and  $\sigma_{||}$ ,  $\sigma_{\perp}$  and  $\varepsilon_{||}$ , and  $\varepsilon_{\perp}$  are the molar volume and the parallel and perpendicular components of the stresses and strains, respectively. The axis perpendicular to the layer-substrate interface is characterized by vanishing forces in this direction ( $\sigma_{\perp} = 0$ ) [27]. Therefore, the stresses parallel to the layer-substrate interface can be written as

$$\sigma_{||} = [(C_{11} - C_{12})(C_{11} + 2C_{12})/C_{11}] \varepsilon_{||}.$$

Thus, the elastic energy of the lattice-mismatched alloy can be rewritten as

$$u_E = \nu \left[ \frac{(C_{11} - C_{12})(C_{11} + 2C_{12})}{C_{11}} \right] \left( \frac{a - a_s}{a_s} \right)^2, \tag{4}$$

where  $a$  and  $a_s$  are the lattice parameters of the phase of the alloy and substrate, respectively. The lattice parameters of the GaP and GaSb substrates are  $a_{GaP} = 5.45 \times 10^{-10}$  m and  $a_{GaSb} = 6.096 \times 10^{-10}$  m, respectively. The molar volume is given by  $\nu = a^3 N_{Av} / 4$  and

$$a = xy a_{AlN} + x(1-y) a_{AlP(Sb)} + (1-x)y a_{GaN} + (1-x)(1-y) a_{GaP(Sb)},$$

where  $N_{Av}$  is the Avogadro number. The stiffness coefficients of the binary compounds were taken from Refs. 28, 29. The minimum value of the elastic energy of the decomposed alloy is reached at the condition  $\gamma_1 = \gamma_2$ . The variation of the Helmholtz free energy (1) at the initial stage using the Taylor's expansion and the condition mentioned above can be expressed as

$$\delta f = \frac{1}{2} \times \frac{\partial^2 f(x, y)}{\partial x^2} \delta x \delta x + \frac{\partial^2 f(x, y)}{\partial x \partial y} \delta x \delta y + \frac{1}{2} \times \frac{\partial^2 f(x, y)}{\partial y^2} \delta y \delta y. \tag{5}$$

This quadratic form ceases to be positive when one of the next quantities

$$\frac{\partial^2 f(x, y)}{\partial x^2} = 0 \tag{6}$$

or

$$\frac{\partial^2 f(x, y)}{\partial x^2} \times \frac{\partial^2 f(x, y)}{\partial y^2} - \left[ \frac{\partial^2 f(x, y)}{\partial x \partial y} \right]^2 = 0 \tag{7}$$

becomes equal to zero [30]. These conditions can be rewritten, respectively, as

$$-2y\alpha_{\text{AlN-GaN}} - 2(1-y)\alpha_{\text{AlP(Sb)-GaP(Sb)}} + \frac{\partial^2 u}{\partial x^2} + \frac{RT}{x(1-x)} = 0, \quad (8)$$

and

$$\begin{aligned} & \left[ -2y\alpha_{\text{AlN-GaN}} - 2(1-y)\alpha_{\text{AlP(Sb)-GaP(Sb)}} + \frac{\partial^2 u_E}{\partial x^2} \times \frac{RT}{x(1-x)} \right] \\ & \times \left[ -2x\alpha_{\text{AlP(Sb)-AlN}} - 2(1-x)\alpha_{\text{GaP(Sb)-GaN}} + \frac{\partial^2 u_E}{\partial y^2} \times \frac{RT}{y(1-y)} \right] \\ & - \left[ \mu_{\text{AlN}}^0 - \mu_{\text{AlP(Sb)}}^0 - \mu_{\text{GaN}}^0 + \mu_{\text{GaP(Sb)}}^0 + (1-2x)(\alpha_{\text{AlN-GaN}} - \alpha_{\text{AlP(Sb)-GaP(Sb)}}) \right. \\ & \left. + (1-2y)(\alpha_{\text{AlP(Sb)-AlN}} - \alpha_{\text{GaP(Sb)-GaN}}) + \frac{\partial^2 u_E}{\partial x \partial y} \right]^2 = 0. \quad (9) \end{aligned}$$

The value  $\mu_{\text{AlN}}^0 - \mu_{\text{AlP(Sb)}}^0 - \mu_{\text{GaN}}^0 + \mu_{\text{GaP(Sb)}}^0$  was estimated as

$$\begin{aligned} & \mu_{\text{AlN}}^0 - \mu_{\text{AlP(Sb)}}^0 - \mu_{\text{GaN}}^0 + \mu_{\text{GaP(Sb)}}^0 \\ & = \Delta h - T\Delta s + \int_{298.15}^T \Delta c dT - T \int_{298.15}^T \frac{\Delta c}{T} dT, \end{aligned}$$

where

$$\begin{aligned} \Delta h &= h_{\text{AlN}} - h_{\text{AlP(Sb)}} - h_{\text{GaN}} + h_{\text{GaP(Sb)}}, \\ \Delta s &= s_{\text{AlN}} - s_{\text{AlP(Sb)}} - s_{\text{GaN}} + s_{\text{GaP(Sb)}}, \\ \Delta c &= c_{\text{AlN}}^P - c_{\text{AlP(Sb)}}^P - c_{\text{GaN}}^P + c_{\text{GaP(Sb)}}^P, \end{aligned}$$

$h_{\text{AlN}}$ ,  $s_{\text{AlN}}$  y  $c_{\text{AlN}}^P$  are the enthalpy, the entropy at STP (standard temperature and pressure) and the specific heat capacity at constant standard pressure of AlN, respectively. The data used in the calculations were taken from Refs. 23, 28, 29 and 31 to 33. The interaction parameters  $\alpha_{\text{AlP-AlN}}=240$  kJ/mol and  $\alpha_{\text{AlSb-AlN}}=597$  kJ/mol were estimated from the strain energies of the  $\text{AlN}_x[\text{P(Sb)}]_{1-x}$  ternary alloys. The strain energy of the  $\text{AlN}_x[\text{P(Sb)}]_{1-x}$  ternary alloys was calculated according to the valence-force field model [24] with elastic constants from Refs. 23 and 32.

### 3. Results and discussion

The estimated spinodal decomposition regions of  $\text{Al}_x\text{Ga}_{1-x}\text{N}_y\text{P}_{1-y}$  and  $\text{Al}_x\text{Ga}_{1-x}\text{N}_y\text{Sb}_{1-y}$  alloys with compositions  $0 \leq y \leq 0.05$ ,  $x = 0, 0.1, 0.2, 0.3, 0.4$  are shown in Figs. 2 and 3, respectively. The spinodal decomposition ranges are determined by condition (7b), because it is fulfilled at a higher temperature than condition (8). The spinodal decomposition temperature was estimated as a function of  $x$  and  $y$  which were used as given values. Consideration of a wider temperature range is invalid since the P- and Sb-rich alloys are epitaxially grown at temperatures lower than

1100°C [34]. In order to analyze the results, the spinodal decomposition range was also calculated, ignoring the transformations of the bonds. In this case the term  $\mu_{\text{AlN}}^0 - \mu_{\text{AlP(Sb)}}^0 - \mu_{\text{GaN}}^0 + \mu_{\text{GaP(Sb)}}^0$  was not taken into account in Eq. (7b). The bond transformation during phase separation provides a dramatic decrease in the estimated temperature of spinodal decomposition. This effect can be explained as an increase of the Helmholtz free energy of the alloy after decomposition, due to transformation of the Al-N and Ga-P bonds in the less thermodynamically profitable Al-P and Ga-N bonds. Exactly the same situation occurs for the N-low content  $\text{Al}_x\text{Ga}_{1-x}\text{N}_y\text{Sb}_{1-y}$  alloys, where Al-N and Ga-Sb bonds are transformed into more thermodynamically profitable Al-Sb and Ga-N bonds.

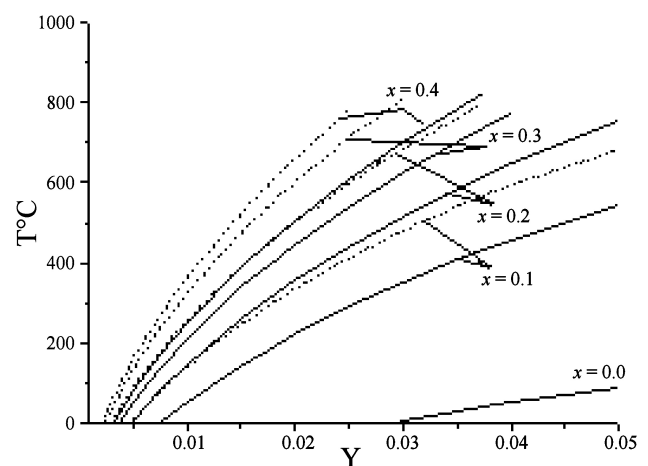


FIGURE 2. Spinodal decomposition ranges of the N-low content  $\text{Al}_x\text{Ga}_{1-x}\text{N}_y\text{P}_{1-y}$  alloys grown on the GaP (001) substrates. Solid curves indicate the range estimated considering the transformation of the bonds. Dotted curves demonstrate the results of calculations ignoring the transformation of the bonds.

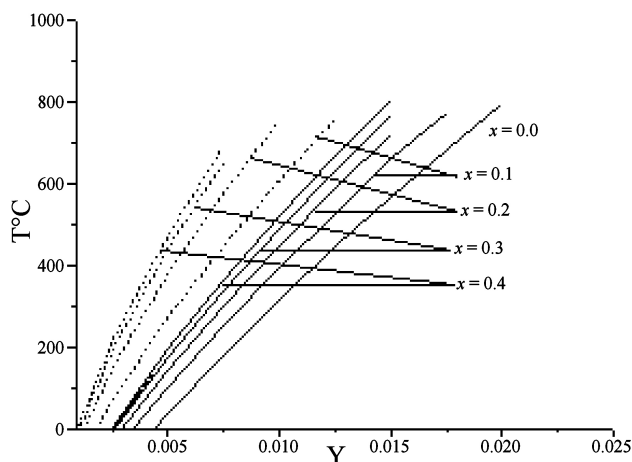


FIGURE 3. Spinodal decomposition ranges of the N-low content  $\text{Al}_x\text{Ga}_{1-x}\text{N}_y\text{Sb}_{1-y}$  alloys grown on the GaSb (001) substrates. Solid curves indicate the range estimated considering the transformation of the bonds. Dotted curves demonstrate the results of calculations ignoring the transformation of the bonds.

As can be seen from Figs. 2 y 3, the increase in the Al and N concentrations raises the temperature of the phase sep-

aration. A substitution of Ga and P or Ga and Sb for Al and N, respectively, increases the concentrations of the Al-N and Ga-N bonds. The bond-stretching and bond-bending elastic constants of Al-N and Ga-N bonds are significantly higher than those of Al-P and Al-Sb bonds [23,32]. A rise of Al and N concentrations increases the strain energy. The elastic and coherency strain energy decreases the spinodal decomposition range of  $\text{Al}_x\text{Ga}_{1-x}\text{N}_y[\text{P}(\text{Sb})]_{1-y}$  alloys. Therefore, in spite of the significant strain energy, the alloys considered in the large composition regions are outside the spinodal decomposition range at their growth temperatures.

#### 4. Conclusions

The spinodal decomposition ranges of the low N-content  $\text{Al}_x\text{Ga}_{1-x}\text{N}_y\text{P}_{1-y}$  and  $\text{Al}_x\text{Ga}_{1-x}\text{N}_y\text{Sb}_{1-y}$  alloys in the wide temperature region are presented. Transformations of the bonds and coherency strain energy significantly suppress the spinodal decomposition ranges. The elastic energy weakly affects the spinodal decomposition of alloys grown on the GaP (001) and GaSb (001) substrates.

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