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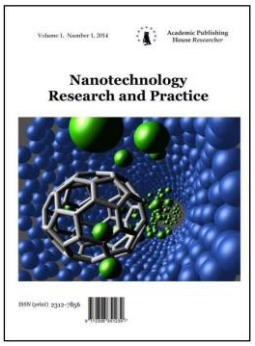

Clustering in Bismide Semiconductor Alloys

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Abstract

Interest in clustering in GaB^{V}-rich GaBi$_x$$B^{V}_{1-x}$ (B^{V} = P, As) highly lattice mismatched bismide alloys has recently grown. The main drawback associated with clusters is their heterogeneity. To resolve this problem, GaB^{V}-rich B$_x$Ga$_{1-x}$Bi$_y$$B^{V}_{1-y}$ (B^{V} = As or P) alloys are proposed. The internal strain energies due to isolated Bi atoms, 1B4Bi and 4B10Bi clusters are calculated. Formation of 1B4Bi and 4B10Bi clusters in B$_x$Ga$_{1-x}$Bi$_y$$B^{V}_{1-y}$ decreases the internal strain and bond energies. The clustering conditions were calculated up to 800 °C. 4B10Bi clustering is more favorable than 1B4Bi one. The density of 4B10Bi clusters was obtained by minimization of the free energy. The cluster formation conditions were studied for $x \geq 5\times10^{-5}$ and $y \geq 0.5x$. The majority of boron atoms should be in 4B10Bi clusters if $x > 1\times10^{-4}$ and $y > 4x$. The binding energy of a hole bound to Bi tetrahedron in GaP is estimated. The obtained results demonstrate that GaB^{V}-rich B$_x$Ga$_{1-x}$Bi$_y$$B^{V}_{1-y}$ alloys are promising semiconductors with 4B10Bi identical clusters.

Keywords: quaternary semiconducting bismuth alloys, nanomaterials, self-assembly, identical clusters.

1. Introduction

GaBi$_x$As$_{1-x}$ highly lattice mismatched bismide alloys are actively studied now (Zhang et al., 2018; Bushell et al., 2018; Valkovskii et al., 2018; Wilson et al., 2018). Such alloys are considered as perspective materials to fabricate a variety of electronic devices. However, high lattice mismatch between the constituent compounds leads to considerable alloy content inhomogeneity. It is a formidable obstacle to expending the use of these alloys.

For the first time, a way to reduce internal strains in $A^{III}B^{V}$ highly lattice mismatched alloys was described for GaAs-rich B$_x$Ga$_{1-x}$Sb$_y$As$_{1-y}$ (Elyukhin, 2009). In such alloys the formation of 1B4Sb tetrahedral clusters with boron atoms in their centers is thermodynamically favorable. Boron and Sb atoms are smaller and larger than Ga and As atoms, respectively. Therefore, the presence of 1B4Sb clusters significantly reduces the internal strains. Further, the 4B10Sb cluster formation in GaAs-rich B$_x$Ga$_{1-x}$Sb$_y$As$_{1-y}$ was also described (Elyukhin, 2011). 4B10Sb clusters are boron tetrahedrons surrounded by Sb atoms. The formation of them decreases internal strains more than that of 1B4Sb clusters. The internal strains also considerably decrease after an occurrence of 1B4Bi and 4B10Bi clusters in InAs-rich B$_x$In$_{1-x}$Bi$_y$As$_{1-y}$ alloys (Elyukhin, 2018). GaB^{V}-rich B$_x$Ga$_{1-x}$Bi$_y$$B^{V}_{1-y}$ (B^{V} = P, As) belong to the same class of alloys just as B$_x$Ga$_{1-x}$Sb$_y$As$_{1-y}$ and B$_x$In$_{1-x}$Bi$_y$As$_{1-y}$. 1B4Bi and 4B10Bi clustering in GaB^{V}-rich B$_x$Ga$_{1-x}$Bi$_y$$B^{V}_{1-y}$ should also decrease the internal

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strains since boron and Bi atoms are, accordingly, smaller and larger than Ga and As (P) atoms. Therefore, 1B4Bi and 4B10Bi clustering in GaB^{V}-rich $B_xGa_{1-x}Bi_yB^{V}_{1-y}$ alloys seems highly probable.

It has long been known that isolated Bi atoms produce the localized states in GaP:Bi (Trumbore et al., 1966). The binding energy of holes bound to isolated Bi atoms in GaP:Bi is 0.04 eV (Phillips, 1969). However, the localized states formed by bismuth pairs in GaP:Bi were not observed (Christian et al, 2015). It is consistent with the theoretical results (Shen et al, 1990). The authors (Shen et al., 1990) demonstrated that different Bi pairs cannot form the localized states in the band gap of GaP due to the significant internal strains. But the estimates in (Shen et al., 1990) do not exclude the formation of the localized states caused by the larger isoelectronic impurity clusters reducing the internal strains in $B_xGa_{1-x}Bi_yP_{1-y}$. Therefore, the study of 1B4Bi and 4B10Bi clustering in GaP-rich $B_xGa_{1-x}Bi_yB^{V}_{1-y}$ would be important. The 1B4Bi and 4B10Bi clustering conditions in GaAs-rich $B_xGa_{1-x}Bi_yAs_{1-y}$ and GaP-rich $B_xGa_{1-x}Bi_yP_{1-y}$ alloys are considered here.

2. Theory

The internal strain energy of $B_xGa_{1-x}Bi_yB^{V}_{1-y}$ is presented as a sum of the internal strain energies due to boron and Bi isolated atoms and 1B4Bi, 4B10Bi clusters shown, accordingly, in Figures 1 and 2.

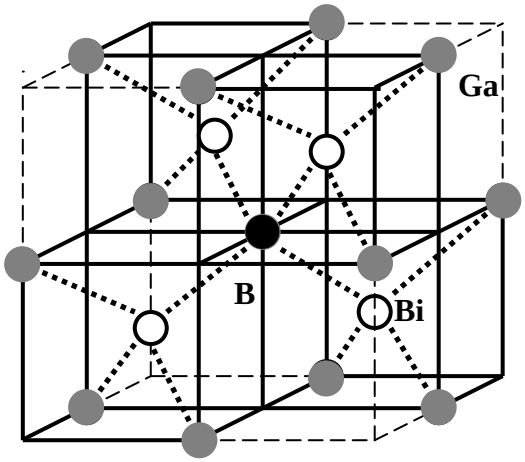

Fig. 1. 1B4Bi cluster in the GaB^{V}-rich matrix

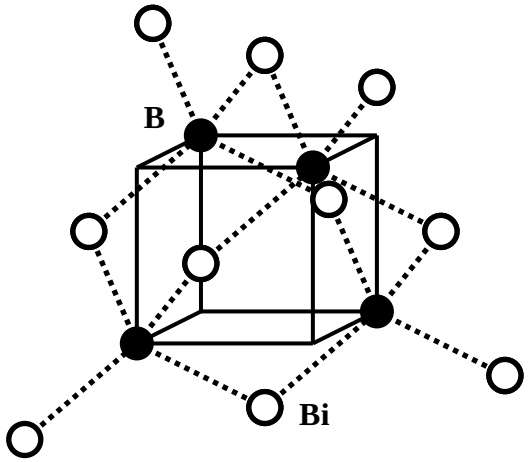

Fig. 2. 4B10Bi cluster

The internal strain energies caused by isolated boron atoms in GaAs and GaP are available (Elyukhin, 2009a; Elyukhin, 2009b). The internal strain energies due to Bi isolated atoms and clusters of both types are the sums of two terms. The first term is the energies caused by the distortions of bonds and angles between bonds formed Bi atoms or clusters. This part is described

by using the valence force field model (Martin, 1970). The bond stretching and bond bending elastic constants of BBi and GaBi were taken from (Elyukhin, 2018; Elyukhin, 2019). The second terms of the sums are the deformation energies of GaB^{V}- rich matrix. The internal strain energies due to isolated Bi atoms, 4B10Bi and 1B4Bi clusters are equal, respectively, to u_{Bi} = 29.29 kJmol^{-1}, u_{4B10Bi} = 254.88 kJmol^{-1} and u_{1B4Bi} = 119.41 kJmol^{-1} in GaAs-rich alloys and u_{Bi} = 51.19 kJmol^{-1}, u_{4B10Bi} = 587.53 kJmol^{-1} and u_{1B4Bi} = 230.62×10^{5} kJmol^{-1} in GaP-rich alloys. All these deformation energies were obtained by the minimization method. The strain energies due to 4B10Bi and 1B4Bi clusters demonstrate a tendency to the formation of these clusters. In addition, the 4B10Bi cluster formation decreases the strain energy much more. It is due to the fact that only 2 bond/atom formed by boron or Bi atoms or both of them are in 4B10Bi clusters, but in 1B4Bi clusters there are 3.2 bond/atom. The formation of polyatomic boron or Bi clusters in GaB^{V}-rich alloys is improbable due to the high lattice mismatch of boron and Bi atoms. An occurrence of other boron and Bi clusters is also unlikely as it should lead to the larger internal strains than that of 1B4Bi and 4B10Bi clusters in the alloys with the same composition.

4B10Bi and 1B4Bi clustering is presented by 4B10Bi cluster order parameter α (4B10Bi COP) and 1B4Bi cluster order parameter β (1B4Bi COP). These parameters are the ratios between numbers of boron atoms in 4B10Bi and 1B4Bi clusters and total number of boron atoms, correspondingly. The alloys are considered in the boron and Bi dilute limits. In such a case, boron and Bi atoms are isolated in the alloys before clustering (disordered alloys). The internal strain energies of $B_xGa_{1-x}Bi_yB^{V}_{1-y}$ with clusters (cluster ordered alloy) and disordered alloy are, respectively,

$$u^{O(IS)} = u_B x + u_{Bi} y + \left(0.25u_{4B10Bi} - u_B - 2.5u_{Bi}\right)\alpha x + \left(u_{1B4Bi} - u_B - 4u_{Bi}\right)\beta x,$$

$$u^{D(IS)} = u_B x + u_{Bi} y,$$

where u_B, u_{Bi}, u_{4B10Bi} and u_{1B4Bi} are the strain energies due to isolated boron and Bi atoms and clusters of both types.

The 4B10Bi and 1B4Bi cluster formation in the disordered alloy decreases the entropy and it is favorable if the free energy of the disordered alloy reduces after an occurrence of the clusters. The entropies of the alloys with and without clusters, respectively, are

$$s^{O} = -RT(1-\alpha)x\ln(1-\alpha)x/(1-\alpha x) - RT(1-x)\ln(1-x)/(1-\alpha x)$$
$$- RT\left(y - 2.5\alpha x - 4\beta x\right)\ln\frac{y - 2.5\alpha x - 4\beta x}{1 - 2.5\alpha x - 4\beta x} - RT(1-y)\ln\frac{1-y}{1-2.5\alpha x - 4\beta x}$$
$$- 0.1RT\alpha x\ln 1.35\alpha x - RT\left(0.074 - 0.1\alpha x\right)\ln(1 - 1.35\alpha x)$$
$$- RT(1-\alpha-\beta)x\ln\left(1-\alpha-\beta\right)/\left(1-\alpha\right) - RT\beta x\ln\beta/\left(1-\alpha\right),$$

$$s^{D} = -R\left[x\ln x + (1-x)\ln(1-x) + y\ln y + (1-y)\ln(1-y)\right].$$

The entropy of the alloy with clusters is presented as a function of a number of atomic configurations. This number is a product of two factors. The first factor is a number of permutations of atoms when isolated boron and Bi atoms as well as 1B4Bi clusters are distributed randomly at a fixed location of 4B10Bi clusters. The second multiplayer is a number of the arrangements of 4B10Bi clusters. Such entropy corresponds to the alloys with the boron and Bi in the dilute limits. The entropy of the disordered alloy describes the random alloy.

$B_xGa_{1-x}Bi_yB^{V}_{1-y}$ are alloys with B-Bi, B-B^{V}, Ga-Bi and Ga-B^{V} chemical bonds. The cluster formation leads to transformation of B-B^{V} and Ga-Bi bonds into B-Bi and Ga-B^{V} ones. This transformation may change the bond energy of the alloy. The bond energies of the cluster ordered and disordered alloys are, respectively,

$$u^{O(B)} = \Delta h_{B^{V}}(\alpha+\beta)(1-y)x + \Delta h_{B^{V}} xy + h_{GaB^{V}} + \left(h_{BB^{V}} - h_{GaB^{V}}\right)x + \left(h_{GaBi} - h_{GaB^{V}}\right)y,$$

$$u^{D(B)} = h_{BBi}xy + h_{BB^{V}}x(1-y) + h_{GaBi}(1-x)y + h_{GaB^{V}}(1-x)(1-y),$$

where $\Delta h_{B^{V}} = h_{BBi} + h_{GaB^{V}} - h_{BB^{V}} - h_{GaBi}$ and $h_{GaB^{V}}$ is the enthalpy of GaB^{V}. The closeness of the values of the enthalpy and internal energy for condensed matter (Elyukhin,

2016a; Elyukhin, 2016b) was taken into account in the last formulae. The value of $\Delta h_{B^{V}}$ is equal to the negative value of a similar relation between the enthalpies of formation $\Delta h^{0f}_{B^{V}} = h^{0f}_{\mathrm{BBi}} - h^{0f}_{BB^{V}} - h^{0f}_{\mathrm{GaBi}} + h^{0f}_{\mathrm{Ga}B^{V}}$ since the enthalpies of the atoms and molecules involved in the enthalpies of formation are self-cancelled. The enthalpies of formation are available (Elyukhin, 2018; Elyukhin, 2019; Landolt-Börnstein, 1984; Dumont, Monteil, 2006). The calculated values of the relations between the enthalpies of formation $\Delta h^{0f}_{\mathrm{P}} = -11\,\mathrm{kJmol}^{-1}$ and $\Delta h^{0f}_{\mathrm{As}} = -57\,\mathrm{kJmol}^{-1}$ demonstrate the primary BBi and GaB^{V} bonding over the BB^{V} and GaBi one, respectively.

The values of COPs were obtained by minimization of the free energy of the alloys. The free energy is a sum of three terms $f^{O} = u^{O(B)} + u^{O(IS)} - Ts^{O}$. The first summand is the bond energy, the second item is the internal strain energy and the third item is the entropy term.

3. Results and discussion

$B_xGa_{1-x}Bi_yAs_{1-y}$ epitaxial films with compositions x = 0.0022, 0.0044, 0.0089, 0.012 and y = 0.016 were successfully grown at 320 °C via MBE (Beaton et al, 2012). The 4B10Bi cluster formation during the growth at 320 °C is improbable since it demands the boron and Bi self-diffusion processes in the crystal lattice. Such processes in III-V semiconductors are very slow at such growth temperature. As it was shown (Kurtz et al., 2001; Müller et al., 2011), the thermal annealing of GaAs-rich $In_xGa_{1-x}N_yAs_{1-y}$ epitaxial films at 670 °C - 700 °C during 15-30 min is an efficient method to change the arrangement of In and nitrogen atoms over long distances. $B_xGa_{1-x}Bi_yB^{V}_{1-y}$ belong to the same type of the alloys just as $In_xGa_{1-x}N_yAs_{1-y}$. That is why the temperature range from 0 °C to 800 °C was considered. The small decrease of the bond energy and considerable diminution of the internal strain energy are two causes of clustering. The fulfilled estimates for $B_xGa_{1-x}Bi_yB^{V}_{1-y}$ demonstrate that all or almost all boron atoms should be in 4B10Bi clusters up to 800 °C if $x > 1\times10^{-4}$ and $y > 4x$. However, the fraction of boron atoms in the clusters in the GaP-rich alloys has to be smaller than that in $B_xGa_{1-x}Bi_yAs_{1-y}$. It is due to the favorable BBi and GaB^{V} bonding over the BB^{V} and GaBi one in GaP-rich alloys is five times less than in GaAs-rich ones ($\Delta h^{0f}_{\mathrm{P}} = -11\,\mathrm{kJmol}^{-1}$ and $\Delta h^{0f}_{\mathrm{As}} = -57\,\mathrm{kJmol}^{-1}$). 1B4Bi clustering also decreases the free energy but less than that of 4B10Bi clusters. Such clusters can be formed earlier than 4B10Bi clusters at a thermal treatment since an occurrence of 1B4Bi clusters demands the redistribution of the smaller number of atoms. Cooling of the alloys with 1B4Bi clusters will lead to the “frozen” atomic distribution as self-diffusion processes at RT are very slow. Accordingly, the last mentioned alloys can also be used for devices applications.

The other cluster formation conditions for the boron and Bi contents in $B_xGa_{1-x}Bi_yB^{V}_{1-y}$ were studied for $x = y = 1\times10^{-4}$, 7.5×10^{-5}, 5×10^{-5} and $x = 2y = 1\times10^{-4}$, 7.5×10^{-5}, 5×10^{-5}. The temperature dependencies of 4B10Bi COPs for $B_xGa_{1-x}Bi_yAs_{1-y}$ with these contents are shown in Figures 3 and 4, correspondingly.

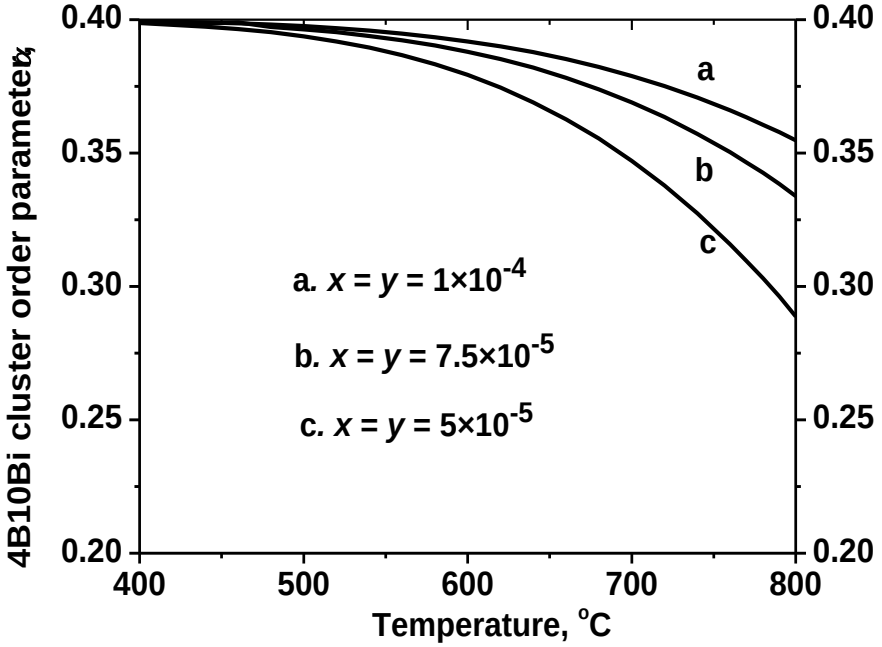

Fig. 3. The temperature dependencies of 4B10Bi COPs in $B_xGa_{1-x}Bi_yAs_{1-y}$ with $x = y = 1\times10^{-4}$, 7.5×10^{-5}, 5×10^{-5}

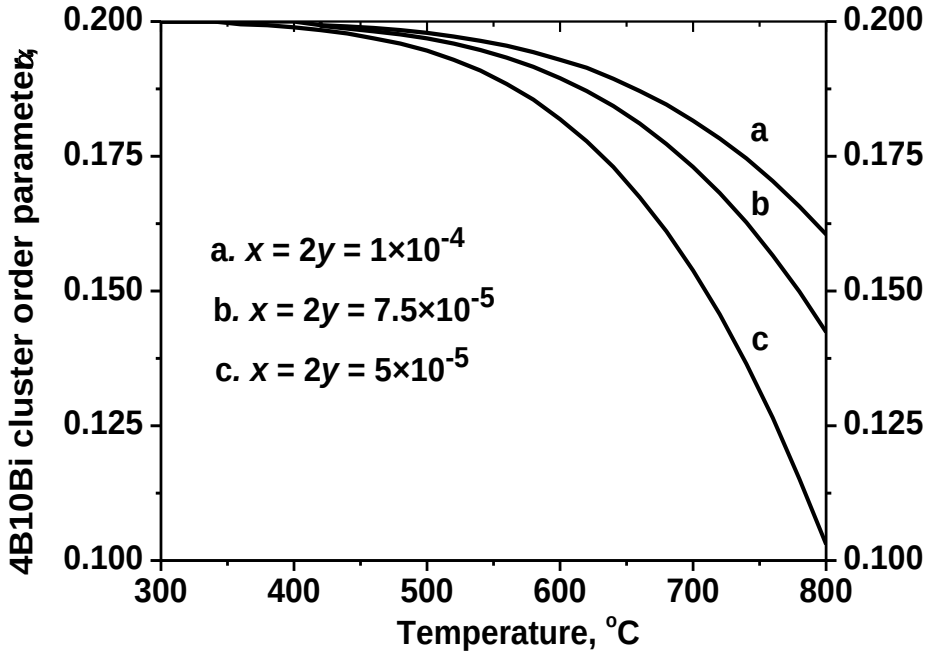

Fig. 4. The temperature dependencies of 4B10Bi COPs in $B_xGa_{1-x}Bi_yAs_{1-y}$ with $x = 2y = 1\times10^{-4}$, 7.5×10^{-5}, 5×10^{-5}

As seen in Figure 3, the smaller concentrations decrease the fraction of boron and Bi atoms in 4B10Bi clusters at high temperatures. The reason is that the clusters decrease the entropy more if the boron and Bi concentrations are smaller. It may be seen also that the larger fraction of boron

atoms is in 4B10Bi clusters in $B_xGa_{1-x}Bi_yAs_{1-y}$ with $x = y = 1\times10^{-4}$ at high temperatures. The significant diminution of Bi atoms which might be in 4B10Bi clusters is in the alloys with $x = 2y = 1\times10^{-4}$, 7.5×10^{-5}, 5×10^{-5} at temperatures above 700 °C. Thus, if a small density of 4B10Bi clusters is desired then alloys with $x > y$ could be used. However, in such a case, isolated highly lattice mismatched boron atoms will produce the considerable internal strains. 1B4Bi COPs should be insignificant in $B_xGa_{1-x}Bi_yAs_{1-y}$.

The temperature dependencies of 4B10Bi COPs for $B_xGa_{1-x}Bi_yP_{1-y}$ with $x = y = 1\times10^{-4}$, 7.5×10^{-5}, 5×10^{-5} and $x = 2y = 1\times10^{-4}$, 7.5×10^{-5}, 5×10^{-5} are shown in Figures 5 and 6, correspondingly.

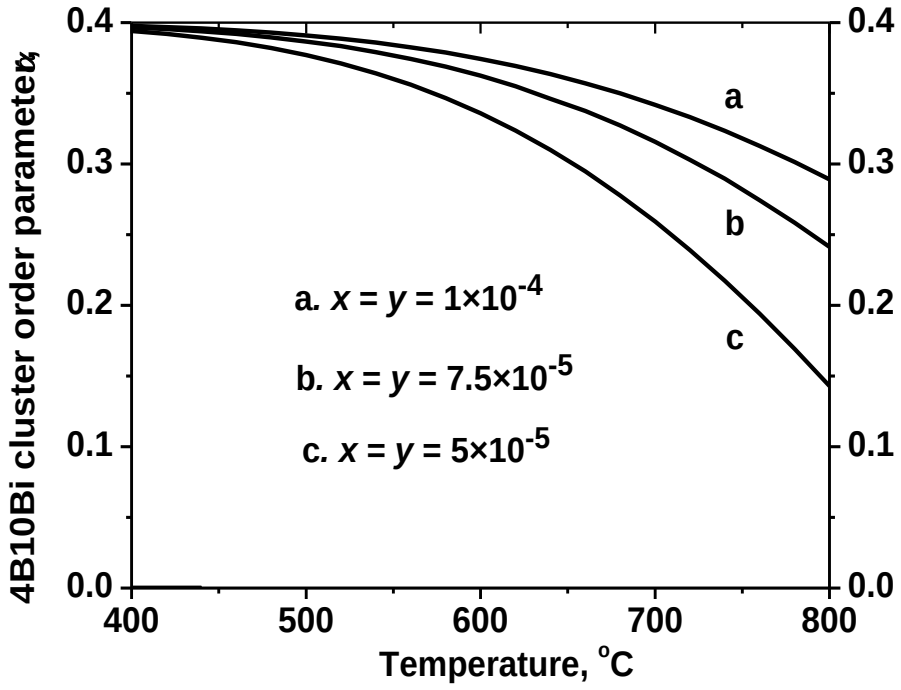

Fig. 5. The temperature dependencies of 4B10Bi COPs in $B_xGa_{1-x}Bi_yP_{1-y}$ with $x = y = 1\times10^{-4}$, 7.5×10^{-5}, 5×10^{-5}

4B10Bi COPs in the GaP-rich alloys (shown in Figures 5 and 6) are smaller than those in the arsenides (Figures 3 and 4). It is also due to the small preference of BBi and GaB^V bonding in phosphides.

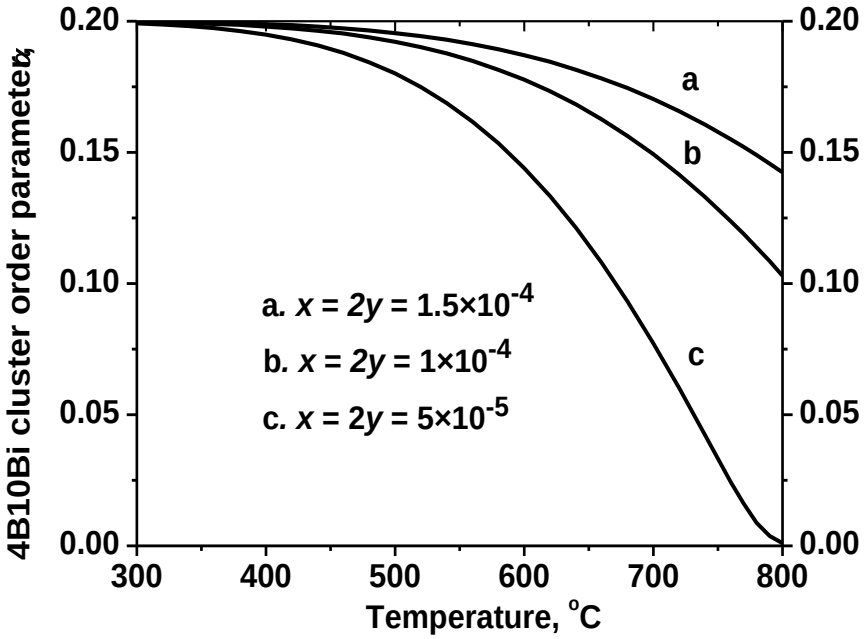

Fig. 6. The temperature dependencies of 4B10Bi COPs in $B_xGa_{1-x}Bi_yP_{1-y}$ with $x = 2y = 1\times10^{-4}$, 7.5×10^{-5}, 5×10^{-5}

4B10Bi COPs in the GaP-rich alloys shown in Figures 5 and 6 are smaller than those in the arsenides (Figures 3 and 4). It is also due to the small preference of BBi and GaB^V bonding in the phosphides. 1B4Bi COPs should be also insignificant in $B_xGa_{1-x}Bi_yP_{1-y}$.

The binding energy due to isolated Bi atoms in GaP:Bi (Trumbore et al, 1966) allows us to estimate the binding energy that may be caused by tetrahedral Bi cluster, by using the approach developed in (Goede, Hennig, 1983). The binding energy of a hole bound to Bi tetrahedron in GaP:Bi should be (Goede, Hennig, 1983)

$$\varepsilon_4 = \frac{1}{2}T_{VB}\left[\frac{J}{T_{VB}} - \frac{7}{6} + \left(\frac{J}{T_{VB}} + \frac{1}{6}\right)\sqrt{1+\frac{3T_{VB}}{2J}}\right]$$

where T_{VB} is an average energy related to the valence band and J is the Koster-Slater parameter (Koster, Slater, 1954). The average energy related to the valence band of GaP was estimated as $T_{VB} = -1.4$ eV (Baldereschi, Hopfield, 1972). The Koster-Slater parameter J is obtained from the experimental data on the binding energy of a hole bound to an isolated Bi atom and calculated value of T_{VB} by using the formula (Goede, Hennig, 1983) $\varepsilon_1 = J\left(1-\frac{TVB}{4J}\right)^2$, where ε_1 = 0.04 eV is the binding energy of a hole bound to an isolated Bi atom. The calculated binding energy of a hole bound to Bi tetrahedron is ε_4 = 0.2598 eV. The binding energy of a hole that should be bound to an unstrained Bi dyad in GaP calculated by using the same approach is only ε_2 = 0.1222 eV. The fulfilled estimates show that the formation of the localized states due to 1B4Bi clusters in $B_xGa_{1-x}Bi_yP_{1-y}$ cannot be excluded in spite of the internal strains. 1B4Bi clusters may be formed earlier than 4B10Bi clusters since the 1B4Bi cluster occurrence demands redistribution lesser number of atoms. Moreover, the self-diffusion processes at RT are very slow. Therefore, the considered alloys with 1B4Bi clusters may be also used for device applications.

4. Conclusion

To conclude, 4B10Bi clustering is favored thermodynamically in GaAs-rich $B_xGa_{1-x}Bi_yAs_{1-y}$ and GaP-rich $B_xGa_{1-x}Bi_yP_{1-y}$ alloys in the wide boron and Bi content ranges. The occurrence of 1B4Bi clusters is also favorable but less than 4B10Bi clusters. However, 1B4Bi clusters may occur earlier than 4B10Bi clusters since their formation demand smaller distances for self-diffusion processes. The alloys with 1B4Bi clusters should be in the "frozen" state at RT. The considerable compensation of the internal strains and favorable BBi and GaAs (GaP) bonding over BAs (BP) and GaBi one are the causes of clustering. The calculations demonstrate that the significant fraction of boron and Bi atoms should be in 4B10Bi clusters even at high temperatures. Therefore, the thermal treatment is offered as an effective way to reach the state of GaAs-rich $B_xGa_{1-x}Bi_yAs_{1-y}$ and GaP-rich $B_xGa_{1-x}Bi_yP_{1-y}$ alloys with the clusters. Thus, the studied semiconductors are promising candidates to obtain the identical polyatomic clusters in the GaB^V-rich $GaBi_xB^V_{1-x}$ (B^V = P, As) highly lattice mismatched bismide alloys.

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