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Ground-state structures in ternary III–V semiconductor alloys

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Abstract. The probability wave of the atomic configuration is used to study the ordered structures of ternary III–V semiconductor alloys. The atomic configuration of the most stable superlattices formed in the ternary III–V semiconductor alloys is discussed. Some features of the experiments are explained.

1. Introduction

Recently, a large number of long-range-ordered structures have been found in ternary III–V semiconductor alloys of type $A_{1-x}^{III}B_x^{III}C^V$ and $A^{III}C_{1-x}^V D_x^V$ grown by molecular beam epitaxy, metallorganic chemical vapour deposition and vapour levitation epitaxy. Since the first ordered structure $L1_0$ was observed in $Al_{1-x}Ga_xAs$ by Kuan *et al* [1], ordering has also been found in other families of compounds. The alloys with a ‘common anion’ known to order are $Al_{1-x}In_xP$ [2, 3], $Ga_{1-x}In_xP$ [4–7], $Al_{1-x}Ga_xAs$ [1], $Al_{1-x}In_xAs$ [8] and $Ga_{1-x}In_xAs$ [9–12]. Additionally, alloys with a ‘common cation’, namely $GaP_{1-x}As_x$ [13–15], $GaAs_{1-x}Sb_x$ [16–18] and $InAs_{1-x}Sb_x$ [19] and $InAs_{1-x}P_x$ [20] have been reported.

The usual expected structure of a ternary alloy is zincblende (space group, $F\bar{4}3m$), a disordered phase in which, in the ‘common-cation’ alloy $A_{1-x}^{III}B_x^{III}C^V$, atoms A^{III} and B^{III} randomly occupy one set of face-centred cubic (FCC) positions and atoms C^V occupy the other set and, in the ‘common-anion’ alloy $A^{III}C_{1-x}^V D_x^V$, atoms C^V and D^V randomly occupy the anion FCC positions, while atoms A^{III} occupy just the cation FCC positions. In describing ordered structures derived from the alloy $A_{1-x}^{III}B_x^{III}C^V$ we call the zincblende compounds $A^{III}C^V$ and $B^{III}C^V$ the ‘parents’, and the alloy $A_{1-x}^{III}B_x^{III}C^V$ the ‘family’. Then there is an infinite set of possible ordered compounds; for example, for a superlattice, one varies the periodicity of the lattice. The newly reported ordered structures all involve ordering of either the cations (for the alloy $A_{1-x}^{III}B_x^{III}C^V$) or the anions ($A^{III}C_{1-x}^V D_x^V$) on one FCC sublattice.

The ordered structures can be derived from some phenomenological models. Khachatryan [21] has developed a statistical thermodynamic theory of the order–disorder transition in crystal solid solutions which transforms the real space into the reciprocal space of the crystal lattice and has set up the self-consistent equations for the order parameter. Clapp and Moss [22] and de Fontaine [23] discussed ordering in the FCC lattice according to the special-point approach. Teng *et al* [24] used five special k -point ordered structures to calculate the band structures of ternary III–V

semiconductor alloys. Ferreira *et al* [25] used six ordered structures to determine the interaction parameters. We have used [26] the theory of the probability wave of atomic configuration (PWAC) to study the occupation probabilities of different kinds of atom at lattice sites in multi-component solid solutions. In this paper, we shall apply this method to study the ground states in multi-component semiconducting alloys.

2. The crystal structures of ground states

The Hamiltonian of a solid is the summation of the kinetic energy of all particles in the solid and the interaction potential energy between them. By means of the adiabatic approximation, the electronic degrees of freedom are reduced so that we have a system of ions with an effective Hamiltonian. The ion-ion interaction can be divided into two parts: $H_{\text{ion-ion}}^0$, which describes the interactions between the ions that are in equilibrium positions, and H_{ph} , which is caused by the deviation from equilibrium positions of ions:

$$H_{\text{ion}} = H_{\text{ph}} + H_{\text{ion-ion}}^0. \quad (1)$$

In the Hamiltonian in equation (1), we consider the term $H_{\text{ion-ion}}^0$, which is different for different compositions and atomic distributions in multi-component crystal structures and determines the crystal structures of multi-component solid solutions.

We consider the ordering of two kinds of atom: A and B. The Hamiltonian of its crystal structure is a random variable and the configuration average of the Hamiltonian can be determined by the augmented-space formalism which was introduced formally by Mookerjee [27] to evaluate the Green function in real space. The randomness in $H_{\text{ion-ion}}^0$ can be conveniently characterized with the help of an indicator function $n(\mathbf{R}, \text{S})$ ($\text{S} \equiv \text{A}, \text{B}$) which takes the values 1 and 0 depending on whether the lattice site \mathbf{R} is occupied by an atom of type S or not, respectively. The Hamiltonian $H_{\text{ion-ion}}^0$ can thus be written as

$$H_{\text{ion-ion}}^0 = \frac{1}{2} \sum_{\mathbf{R}, \mathbf{R}'} \sum_{\text{S}, \text{S}'} W_{\text{SS}'}(\mathbf{R} - \mathbf{R}') n(\mathbf{R}, \text{S}) n(\mathbf{R}', \text{S}') \quad (2)$$

where $W_{\text{SS}'}(\mathbf{R} - \mathbf{R}')$ represents the effective interaction energy between the S ion at the \mathbf{R} site and the S' ion at the \mathbf{R}' site ($\text{S} \equiv \text{A}$ or B , $\text{S}' \equiv \text{A}$ or B), and \sum' means exclusion of the term corresponding to $\mathbf{R} = \mathbf{R}'$ in the sum.

Rather than thinking of the indicator functions $n(\mathbf{R}, \text{S})$ as random quantities we interpret them as quantum mechanical operators characterized by their eigenfunctions $|RA\rangle$ and $|RB\rangle$ [28]. Thus,

$$\hat{n}(\mathbf{R}, \text{S})|RS'\rangle = \delta_{\text{SS}'}|RS'\rangle \quad (3)$$

where we have used a caret to distinguish the operator $\hat{n}(\mathbf{R}, \text{S})$ from the random quantity $n(\mathbf{R}, \text{S})$. The replacement $n \rightarrow \hat{n}$ in equation (2) defines a Hamiltonian $\hat{H}_{\text{ion-ion}}^0$ in the space spanned by the functions $\prod_{\mathbf{R}} |RS_{\mathbf{R}}\rangle$ where $S_{\mathbf{R}} \equiv \text{A}, \text{B}$. A state in configuration space is specified by selecting the set $\{S_{\mathbf{R}}\}$ in exactly the same way as the occupation of all sites to characterize a particular configuration. The average can

be similarly calculated as expectation values in appropriate augmented-space states. The state

$$|R0\rangle = \sqrt{P_A(\mathbf{R})}|RA\rangle + \sqrt{P_B(\mathbf{R})}|RB\rangle \quad (4)$$

describes a situation where the lattice site \mathbf{R} is occupied with probability P_A by an A atom and with probability P_B by a B atom with

$$\sum_{\mathbf{R}} P_A(\mathbf{R}) = N_A \quad (5)$$

$$\sum_{\mathbf{R}} P_B(\mathbf{R}) = N_B \quad (6)$$

where N_A and N_B are the total numbers of A atoms and B atoms, respectively, in solids. Hence,

$$E = \hat{H} = \langle f | \hat{H} | f \rangle \quad (7)$$

where

$$|f\rangle = |\dots R0\dots\rangle. \quad (8)$$

Using equations (3) and (4), the average value of the Hamiltonian is given by

$$E = U_0 - \frac{1}{2}N_A \sum_{\mathbf{R}}' W(\mathbf{R}) + \frac{1}{2} \sum_{\mathbf{R}, \mathbf{R}'}' W_{AA}(\mathbf{R} - \mathbf{R}') P_A(\mathbf{R}) P_A(\mathbf{R}') \quad (9)$$

where

$$W(\mathbf{R} - \mathbf{R}') = W_{AA}(\mathbf{R} - \mathbf{R}') + W_{BB}(\mathbf{R} - \mathbf{R}') - 2W_{AB}(\mathbf{R} - \mathbf{R}') \quad (10)$$

$$U_0 = \frac{1}{2}N_A \sum_{\mathbf{R}}' W_{AA}(\mathbf{R}) + \frac{1}{2}N_B \sum_{\mathbf{R}}' W_{BB}(\mathbf{R}) \quad (11)$$

is the total internal energy of a pure A-atom system and pure B-atom system. Since only $P_A(\mathbf{R})$ occurs in equation (9), we simply use $P(\mathbf{R})$ to represent $P_A(\mathbf{R})$ in the following.

The crystal structure of the ground state can be determined by minimizing the expectation value of the energy when the atomic mean occupation probability at the lattice site is introduced [26]. The PWAC is represented by

$$P(\mathbf{R}) = \sum_{\mathbf{k}} Q(\mathbf{k}) \exp(i\mathbf{k} \cdot \mathbf{R}) \quad (12)$$

where \mathbf{k} is the wavevector which is taken in the first Brillouin zone; $Q(\mathbf{k})$ are the amplitudes of the probability wave with the wavevector \mathbf{k} and they are determined by the energy-minimum condition and the conservation of particle number. The

Fourier transform equation (12) converts the problem of determining $P(\mathbf{R})$ into that of determining N eigen-amplitudes $Q(\mathbf{k})$. Using equation (12), it gives

$$E = U_0 - \frac{1}{2}x(1-x)NV(0) + \frac{1}{2}N \sum_{\mathbf{k}}' V(\mathbf{k})|Q(\mathbf{k})|^2 \quad (13)$$

where

$$V(\mathbf{k}) = \sum_{\mathbf{R}}' W(\mathbf{R}) \exp(i\mathbf{k} \cdot \mathbf{R}) \quad (14)$$

$$x = N_A/N = Q(0). \quad (15)$$

This allows us to determine the lowest-energy state in \mathbf{k} -space and to determine the coefficients $Q(\mathbf{k})$ in equation (12) according to the minimum of energy. In [24], it is shown that if

$$[\partial V(\mathbf{k})/\partial \mathbf{k}]|_{\mathbf{k}=\mathbf{k}_0} = 0 \quad (16)$$

with $V(\mathbf{k}_0) \leq 0$, and \mathbf{k}_0 is n -fold degenerate with the degenerate states $V(\mathbf{k}_0^1)$, $V(\mathbf{k}_0^2)$, ..., $V(\mathbf{k}_0^n)$, the ground state is represented by the PWAC

$$P(\mathbf{R}) = x + \sum_i^n Q(\mathbf{k}_0^i) \exp(i\mathbf{k}_0^i \cdot \mathbf{R}) \quad (17)$$

and the energy of the ground state is

$$E = U_0 - \frac{1}{2}x(1-x)NV(0) + \frac{1}{2}N \sum_i^n V(\mathbf{k}_0^i)|Q(\mathbf{k}_0^i)|^2. \quad (18)$$

3. Ordered crystal structures

In ternary III-V semiconducting alloys $A_{1-x}^{III}B_x^{III}C^V$ or $A^{III}C_{1-x}^V D_x^V$, if we consider the situation of $A_{1-x}^{III}B_x^{III}C^V$, which is also applicable to $A^{III}C_{1-x}^V D_x^V$, there are two bases, i.e. $A^{III}C^V$ (simply called the A basis) and $B^{III}C^V$ (simply called the B basis). We regard a basis as a structure point so that it converts into the structure problem of a two-component solution. The corresponding structure is a FCC crystal structure, and Γ , X, K and W are the high-symmetry points in the Brillouin zone (BZ). The lattice sites are represented by

$$\mathbf{R} = \frac{1}{2}m_1\mathbf{a}_1 + \frac{1}{2}m_2\mathbf{a}_2 + \frac{1}{2}m_3\mathbf{a}_3 \quad (19)$$

where m_1 , m_2 and m_3 are integers and $m_1 + m_2 + m_3$ is even.

If we suppose that $P(\mathbf{R})$ is the occupation probability of the A basis at the site \mathbf{R} , according to the PWAC theory, it is described by equation (17). At the symmetry point in the first BZ, the wavevector in the same star $\{\mathbf{k}_s\}$ is denoted by \mathbf{k}_{j_s} . The amplitudes $Q(\mathbf{k})$ are written as a product of two parameters:

$$Q(\mathbf{k}_{j_s}) = \eta_s \gamma_s(j_s) \quad (20)$$

where η_s is only related to the $\{k_s\}$ star, while $\gamma_s(j_s)$ are related to the specific k_{j_s} contained in the $\{k_s\}$ star. In fact, η_s is the order parameter of the ordered phase. $\eta_s = 0$ corresponds to the disordered phase and $|\eta_s| = 1$ corresponds to the completely ordered phase:

$$P(\mathbf{R}) = x + \frac{1}{2} \sum_s \eta_s \sum_{j_s} [\gamma_s(j_s) \exp(i\mathbf{k}_{j_s} \cdot \mathbf{R}) + \gamma_s^*(j_s) \exp(-i\mathbf{k}_{j_s} \cdot \mathbf{R})]. \quad (21)$$

The key point in finding the energy value of the ground state in equation (9) and the corresponding PWAC is to determine the wavevector k_0 . When $V(k_0) > 0$, there is only a disordered state. When $V(k_0) \leq 0$ and the conditions of equation (16) are satisfied, phase decomposition will appear when $k_0 = 0$ while a superlattice will appear when $k_0 \neq 0$. Different k_0 will lead to different superlattices. In the first BZ of the crystal there are two kinds of $V(k_0)$ minimum point: firstly, arbitrary points in the BZ which depend on the atomic interaction parameters $W(\mathbf{R} - \mathbf{R}')$; secondly, high-symmetry points according to Lifshitz's [29] criterion, which are determined by the symmetry of the disordered state and lead to the most stable ordered structures. Therefore we shall consider only (X), (L) and (W) stars when we study the atomic configurations of the most stable ordered structures.

3.1. $V(L)$ minimum and $V(L) < 0$

There are two situations. Firstly

$$P(\mathbf{R}) = x + \eta_L \gamma_L \cos\left[\frac{1}{2}\pi(m_1 + m_2 + m_3)\right] \\ = \begin{cases} x + \eta_L \gamma_L & \text{when } \frac{1}{2}(m_1 + m_2 + m_3) \text{ is even} \\ x - \eta_L \gamma_L & \text{otherwise.} \end{cases} \quad (22)$$

Using the conditions of complete ordering $\eta_L = 1$, $P(\mathbf{R}) = 1$ or 0 , and the coefficient γ_L and the corresponding concentration of the completely ordered state are $\gamma_L = \frac{1}{2}$ and $x = \frac{1}{2}$, respectively. Figure 1(a) shows the superlattice determined by equation (22).

Secondly,

$$P(\mathbf{R}) = x + \eta_L \gamma_L \{-\cos\left[\frac{1}{2}\pi(m_1 + m_2 + m_3)\right] + \cos\left[\frac{1}{2}\pi(-m_1 + m_2 + m_3)\right] \\ + \cos\left[\frac{1}{2}\pi(m_1 - m_2 + m_3)\right] + \cos\left[\frac{1}{2}\pi(m_1 + m_2 - m_3)\right]\} \\ = \begin{cases} x + 2\eta_L \gamma_L & \text{when } \frac{1}{2}(m_1 + m_2 + m_3) \text{ and } m_i \text{ are even, or} \\ & \text{when } \frac{1}{2}(m_1 + m_2 + m_3) \text{ is odd and} \\ & \text{one of } m_i \text{ is even} \\ x - 2\eta_L \gamma_L & \text{otherwise.} \end{cases} \quad (23)$$

The coefficient γ_L and the concentration of the completely ordered state are $\gamma_L = \frac{1}{4}$ and $x = \frac{1}{2}$, respectively. Figure 1(h) shows the superlattice along the $\langle 111 \rangle$ direction.

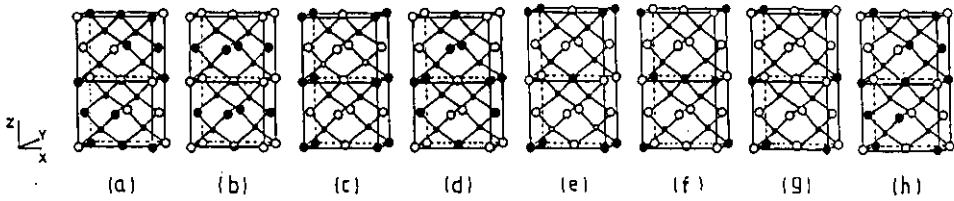


Figure 1. Ordered structures in ternary III-V semiconductor alloys: (a) the superlattice in the [111] direction; (b) the superlattice in the [100] direction; (c) the superlattice in the <100> direction; (d) the superlattice in the [10 $\frac{1}{2}$] direction; (e) the superlattice in the [001] and [10 $\frac{1}{2}$] directions; (f) the superlattice in the [001] and <111> directions; (g) the superlattice in the <100> and <111> directions; (h) the superlattice in the <111> direction.

3.2. $V(X)$ minimum and $V(X) < 0$

Equation (21) reduces to

$$P(\mathbf{R}) = x + \eta_X [\gamma_X(1) \cos(\pi m_1) + \gamma_X(2) \cos(\pi m_2) + \gamma_X(3) \cos(\pi m_3)]. \tag{24}$$

There are two situations. Firstly, for $\gamma_X(1) = \gamma_X \neq 0, \gamma_X(2) = \gamma_X(3) = 0,$

$$P(\mathbf{R}) = x + \eta_X \gamma_X \cos(\pi m_1) = \begin{cases} x + \eta_X \gamma_X & \text{when } m_1 \text{ is even} \\ x - \eta_X \gamma_X & \text{when } m_1 \text{ is odd.} \end{cases} \tag{25}$$

The coefficient γ_X and the concentration of the completely ordered state are $\gamma_X = \frac{1}{2}$ and $x = \frac{1}{2},$ respectively. Figure 1(b) shows the superlattice along the [100] direction determined by equation (25).

Secondly, for $\gamma_X(1) = \gamma_X(2) = \gamma_X(3) = \gamma_X,$

$$\begin{aligned} P(\mathbf{R}) &= x + \eta_X \gamma_X [\cos(\pi m_1) + \cos(\pi m_2) + \cos(\pi m_3)] \\ &= \begin{cases} x + 3\eta_X \gamma_X & \text{when } m_i \text{ are all even} \\ x - \eta_X \gamma_X & \text{otherwise.} \end{cases} \end{aligned} \tag{26}$$

The coefficient γ_X and the concentration of the completely ordered state are $\gamma_X = \frac{1}{4}$ and $x = \frac{1}{4},$ respectively. Figure 1(c) shows the superlattice along the <100> direction determined by equation (26).

3.3. $V(W)$ minimum and $V(W) < 0$

Only one structure is obtained in this situation:

$$\begin{aligned} P(\mathbf{R}) &= x + \eta_W \gamma_W \left\{ \cos\left[\pi\left(m_1 + \frac{1}{2}m_3\right)\right] + \sin\left[\pi\left(m_1 + \frac{1}{2}m_3\right)\right] \right\} \\ &= \begin{cases} x + \frac{1}{2}\eta_W \gamma_W & \text{when } m_1 + \frac{1}{2}m_3 \text{ or } m_1 + \frac{1}{2}m_3 - \frac{1}{2} \text{ is even} \\ x - \frac{1}{2}\eta_W \gamma_W & \text{otherwise.} \end{cases} \end{aligned} \tag{27}$$

The coefficient γ_W and the concentration of the completely ordered state are $\gamma_W = 1$ and $x = \frac{1}{2},$ respectively. Figure 1(d) shows the superlattice along the [10 $\frac{1}{2}$] direction.

There are another three stable ordered structures resulting from the degeneration of $V(\mathbf{k}).$

3.4. $V(X) = V(W) = V_{\min}(\mathbf{k}) < 0$

Then,

$$P(\mathbf{R}) = x + \eta_X \gamma_X \cos(\pi m_3) + \eta_W \gamma_W \cos[\pi(m_1 + \frac{1}{2}m_3)]$$

$$= \begin{cases} x + \eta_X \gamma_X + \eta_W \gamma_W & \text{when } m_3 \text{ and } m_1 + \frac{1}{2}m_3 \text{ are even} \\ x + \eta_X \gamma_X - \eta_W \gamma_W & \text{when } m_3 \text{ is even and } m_1 + \frac{1}{2}m_3 \text{ is odd} \\ x - \eta_X \gamma_X & \text{otherwise.} \end{cases} \quad (28)$$

The coefficients γ_X and γ_W and the concentration of the completely ordered state are $\gamma_X = \frac{1}{4}$, $\gamma_W = \frac{1}{2}$ and $x = \frac{1}{4}$, respectively. Figure 1(e) shows the superlattices in the [001] and $[10\frac{1}{2}]$ directions determined by equation (28).

3.5. $V(X) = V(L) = V_{\min}(\mathbf{k}) \leq 0$

There are two situations. Firstly,

$$P(\mathbf{R}) = x + \eta_X \gamma_X \cos(\pi m_3)$$

$$+ \eta_L \gamma_L \{ \cos[\frac{1}{2}\pi(m_1 + m_2 + m_3)] + \cos[\frac{1}{2}\pi(m_1 + m_2 - m_3)] \}$$

$$= \begin{cases} x + \eta_X \gamma_X + 2\eta_L \gamma_L & \text{when } m_3 \text{ and } \frac{1}{2}(m_1 + m_2 + m_3) \text{ are even} \\ x + \eta_X \gamma_X - 2\eta_L \gamma_L & \text{when } m_3 \text{ is even and } \frac{1}{2}(m_1 + m_2 + m_3) \\ & \text{is odd} \\ x - \eta_X \gamma_X & \text{otherwise.} \end{cases} \quad (29)$$

The coefficients γ_X and γ_L and the concentration of the completely ordered state are $\gamma_X = \frac{1}{4}$, $\gamma_L = \frac{1}{4}$ and $x = \frac{1}{4}$, respectively. Figure 1(f) shows the superlattice determined by equation (29).

Secondly,

$$P(\mathbf{R}) = x + \eta_X \gamma_X [\cos(\pi m_1) + \cos(\pi m_2) + \cos(\pi m_3)]$$

$$+ \eta_L \gamma_L \{ \cos[\frac{1}{2}\pi(m_1 + m_2 + m_3)] + \cos[\frac{1}{2}\pi(-m_1 + m_2 + m_3)]$$

$$+ \cos[\frac{1}{2}\pi(m_1 - m_2 + m_3)] + \cos[\frac{1}{2}\pi(m_1 + m_2 - m_3)] \}$$

$$= \begin{cases} x + 3\eta_X \gamma_X + 4\eta_L \gamma_L & \text{when } m_i \text{ and } \frac{1}{2}(m_1 + m_2 + m_3) \text{ are even} \\ x + 3\eta_X \gamma_X - 4\eta_L \gamma_L & \text{when } m_i \text{ is even and } \frac{1}{2}(m_1 + m_2 + m_3) \\ & \text{is odd} \\ x - \eta_X \gamma_X & \text{otherwise.} \end{cases} \quad (30)$$

The coefficients γ_X and γ_L and the concentration of the completely ordered state are $\gamma_X = \frac{1}{8}$, $\gamma_L = \frac{1}{8}$ and $x = \frac{1}{8}$, respectively. Figure 1(g) shows the superlattices determined from equation (30).

4. Discussion

We have used the PWAC to study the ground states in ternary III-V semiconductor alloys. The most stable ordered structures are determined.

There are many cases of ordering found in III-V ternary semiconductor alloys. Most of them are (111) ordering. The 'common-anion' alloy $\text{Ga}_{1-x}\text{In}_x\text{P}$ [4-7] has (111) ordering and the 'common-cation' alloy $\text{GaAs}_{1-x}\text{Sb}_x$ [16-18] also shows (111) ordering. Some other ordered structures have also been found. For some alloys, e.g. $\text{Ga}_{1-x}\text{In}_x\text{As}$ [9, 10], more than one special-point direction has been reported. It should be noted that the experimental conditions for long-range ordering are complicated. Some ordered phases of the alloy exist as metastable phases and the phenomenon of ordering also depends on the kinetics of growth.

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