

Electron-phonon interaction in ternary mixed crystals

Xu Wang

*Laboratory of Solid-State Physics, Department of Physics, Neimenggu University, Hohhot, Neimenggu 010021,
The People's Republic of China*

Xi Xia Liang

*Center of Theoretical Physics, Chinese Center of Advanced Science and Technology (World Laboratory),
P.O. Box 8730, Beijing 100080, China
and Laboratory of Solid-State Physics, Department of Physics, Neimenggu University, Hohhot,
Neimenggu 010021, The People's Republic of China*

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The interaction between an electron and a LO phonon in a ternary mixed crystal is studied. The Hamiltonian of the electron-phonon interaction is obtained by using a random-element-isodisplacement-type model and Born-Huang method. The eigenfrequencies of the optical vibration of the lattices and the Fröhlich electron-phonon coupling parameters are calculated for several mixed crystals. It is shown that the concentration dependence of the optical modes and the Fröhlich parameters exhibits one-mode or two-mode behavior. The interaction of the electron with two branches of the optical phonon is discussed.

INTRODUCTION

The ternary mixed crystal system $A_xB_{1-x}C$ is frequently used in heterostructures and superlattices. Many properties of the ternary mixed crystals are different from those of binary crystals and can be modulated by changing the mole fractions of the compounds. Some of the most interesting and most important effects on the physical properties of the mixed crystals arise from their lattice vibrations.

It is well known that the long-wavelength optical phonons in the ternary mixed crystal $A_xB_{1-x}C$ exhibit either a so-called one- or two-mode behavior.¹ There has been much theoretical and experimental work to try to understand the long-wavelength optical phonons in ternary mixed crystals.²⁻¹³ Some authors have discussed the one- and two-mode behavior in detail^{1,13} and have given some criteria for them, but have not dealt with the electron-phonon interaction. Adachi has considered the coupling between the electron and LO lattice vibrations and then discussed the electron effective mass.⁵ However, he only used a linear interpolation method, i.e., he simply considered the compositional dependence of the electron-phonon coupling parameter versus the mole fraction x of compound A as a linear relationship. The interactions of the electron with two optical-phonon modes have not been discussed.

The purpose of this paper is to investigate the electron-phonon interactions in the ternary mixed crystals. We start from the random-element-isodisplacement (REI) model of Chang *et al.*,¹¹ which has been successfully used to treat the optical behavior of a mixed crystal $A_xB_{1-x}C$. By solving the Born-Huang equations, we obtain the frequencies of the long-wavelength optical vibrations as functions of the mole fraction x of compound A

in Sec. I. The Hamiltonian operator for electron-phonon interaction is given in Sec. II. We also calculate the Fröhlich electron-phonon coupling parameters for several ternary mixed crystals in Sec. III. Finally, the conclusions obtained in this work are summarized briefly.

I. EIGENVALUES OF THE FREQUENCIES

Chang and Mitra have successfully derived the eigenfrequencies of the ternary mixed crystal.^{1,11} For further investigating the electron-phonon interaction, we retreat this problem by using a method analogous to the Born-Huang method.^{13,14}

On the REI model, we assume that a fraction x of the immediate neighbors of C ions are A ions, and the fraction $1-x$ are B ions. The A ions and B ions always have C ions as nearest neighbors. Let us use equations closely resembling the Born-Huang equations¹⁴ to describe the vibration of the lattice of a mixed crystal. Taking into account only nearest-neighbor interaction, the equations of motion for a mixed crystal $A_xB_{1-x}C$ of cubic symmetry can be written as

$$m_A \ddot{\mathbf{u}}_A = -\beta_1(\mathbf{u}_A - \mathbf{u}_C) + e_A \mathbf{E}^{\text{loc}}, \quad (1.1)$$

$$m_B \ddot{\mathbf{u}}_B = -\beta_2(\mathbf{u}_B - \mathbf{u}_C) + e_B \mathbf{E}^{\text{loc}}, \quad (1.2)$$

$$m_C \ddot{\mathbf{u}}_C = -x\beta_1(\mathbf{u}_C - \mathbf{u}_A) - (1-x)\beta_2(\mathbf{u}_C - \mathbf{u}_B) - e_C \mathbf{E}^{\text{loc}}, \quad (1.3)$$

In these equations, m , \mathbf{u} , and e are the masses, displacements, and effective charges of the ions A , B , and C . β_1 and β_2 are the nearest-neighbor force constants between A and C ions and B and C ions, respectively. \mathbf{E}^{loc} is the effective local field at the elementary cell location. This local field is related to the macroscopic field \mathbf{E} and the

polarization \mathbf{P} for simple cubic lattices by the well-known relation

$$\mathbf{E}^{\text{loc}} = \mathbf{E} + \mathbf{P}/3\epsilon_0, \quad (1.4)$$

where ϵ_0 is the vacuum dielectric constant.

The equations of motion for the relative displacements are given by

$$\ddot{\mathbf{s}}_1 = -\frac{\beta_1}{\mu_1} \frac{m_C + m_A x}{m_C + m_A} \mathbf{s}_1 - \frac{\beta_2}{\mu_2} \frac{m_A(1-x)}{m_C + m_B} \mathbf{s}_2 + \frac{e_1^*}{\mu_1} \mathbf{E}^{\text{loc}}, \quad (1.5)$$

$$\ddot{\mathbf{s}}_2 = -\frac{\beta_1}{\mu_1} \frac{m_B x}{m_C + m_A} \mathbf{s}_1 - \frac{\beta_2}{\mu_2} \frac{m_C + m_B(1-x)}{m_C + m_B} \mathbf{s}_2 + \frac{e_2^*}{\mu_2} \mathbf{E}^{\text{loc}}, \quad (1.6)$$

where $\mathbf{s}_1 = (\mathbf{u}_A - \mathbf{u}_C)$, $\mathbf{s}_2 = (\mathbf{u}_B - \mathbf{u}_C)$, and e_1^* and e_2^* are the effective charges of the pairs of A and C ions and B and C ions, respectively. μ_1 is the reduced mass of ions A and C and μ_2 is that of ions B and C .

The total polarization in the unit volume is

$$\mathbf{P} = x e_1^* \mathbf{s}_1 + (1+x) e_2^* \mathbf{s}_2 + [x\gamma_1 + (1+x)\gamma_2] \mathbf{E}^{\text{loc}}, \quad (1.7)$$

where γ_1 and γ_2 are the electronic polarizabilities of the pairs of A and C ions and B and C ions, respectively.

Inserting relation (1.4) into Eqs. (1.5)–(1.7) and replacing the vectors \mathbf{s}_1 and \mathbf{s}_2 by vectors $\mathbf{w}_1 = \mu_1^{1/2} \mathbf{s}_1$ and $\mathbf{w}_2 = \mu_2^{1/2} \mathbf{s}_2$, we can obtain the equations resembling the Born-Huang equations as follows:

$$\ddot{\mathbf{w}}_1 = b_{11} \mathbf{w}_1 + b_{12} \mathbf{w}_2 + b_{13} \mathbf{E}, \quad (1.8)$$

$$\ddot{\mathbf{w}}_2 = b_{21} \mathbf{w}_1 + b_{22} \mathbf{w}_2 + b_{23} \mathbf{E}, \quad (1.9)$$

$$\mathbf{P} = b_{31} \mathbf{w}_1 + b_{32} \mathbf{w}_2 + b_{33} \mathbf{E}. \quad (1.10)$$

We can now relate the coefficients entering in Eqs. (1.8)–(1.10) to macroscopic parameters according to the usual Born-Huang procedure. In fact, these equations and coefficients must degenerate into that for the binary crystal AC when $x = 1$ and that for the binary crystal BC when $x = 0$. Taking these conditions into our calculation, the following relations can be determined:

$$\gamma(x) = \frac{[\epsilon_1(\infty) - 1][\epsilon_2(\infty) + 2]x + [\epsilon_2(\infty) - 1][\epsilon_1(\infty) + 2](1-x)}{[\epsilon_2(\infty) + 2]x + [\epsilon_1(\infty) + 2](1-x)}. \quad (1.21)$$

Here, ω_{T1} and ω_{T2} are the TO-phonon frequency of the crystals AC and BC ; the subscripts 1 and 2 on the static and high-frequency dielectric constants $\epsilon(0)$ and $\epsilon(\infty)$ refer to the crystal AC and BC , respectively.

We now look for solutions of Eqs. (1.8)–(1.10) of the type $\exp[i(\mathbf{k} \cdot \mathbf{r} - \omega t)]$. External fields are not involved. One can divide \mathbf{w} into an irrotational and a divergence-

$$b_{11} = -\omega_{T1}^2 \left[\frac{\epsilon_1(0) + 2}{\epsilon_1(\infty) + 2} \frac{m_C + m_A x}{m_C + m_A} - \frac{\epsilon_1(0) - \epsilon_1(\infty)}{[\epsilon_1(\infty) + 2]^2} x a(x) \right], \quad (1.11)$$

$$b_{12} = -\omega_{T2}^2 (1-x) \left[\frac{\epsilon_2(0) + 2}{\epsilon_2(\infty) + 2} \frac{m_A}{m_C + m_B} \left(\frac{\mu_1}{\mu_2} \right)^{1/2} - \frac{\epsilon_2(0) - \epsilon_2(\infty)}{[\epsilon_2(\infty) + 2]^2} \left(\frac{\mu_2}{\mu_1} \right)^{1/2} a(x) \right], \quad (1.12)$$

$$b_{13} = \omega_{T1} \frac{\epsilon_0^{1/2} [\epsilon_1(0) - \epsilon_1(\infty)]^{1/2}}{\epsilon_1(\infty) + 2} [3 + \gamma(x)], \quad (1.13)$$

$$b_{21} = -\omega_{T1}^2 x \left[\frac{\epsilon_1(0) + 2}{\epsilon_1(\infty) + 2} \frac{m_B}{m_C + m_A} \left(\frac{\mu_2}{\mu_1} \right)^{1/2} - \frac{\epsilon_1(0) - \epsilon_1(\infty)}{[\epsilon_1(\infty) + 2]^2} \left(\frac{\mu_1}{\mu_2} \right)^{1/2} a(x) \right], \quad (1.14)$$

$$b_{22} = -\omega_{T2}^2 \left[\frac{\epsilon_2(0) + 2}{\epsilon_2(\infty) + 2} \frac{m_C + m_B(1-x)}{m_C + m_B} - \frac{\epsilon_2(0) - \epsilon_2(\infty)}{[\epsilon_2(\infty) + 2]^2} (1-x) a(x) \right], \quad (1.15)$$

$$b_{23} = \omega_{T2} \frac{\epsilon_0^{1/2} [\epsilon_2(0) - \epsilon_2(\infty)]^{1/2}}{\epsilon_2(\infty) + 2} [3 + \gamma(x)], \quad (1.16)$$

$$b_{31} = \omega_{T1} \frac{\epsilon_0^{1/2} [\epsilon_1(0) - \epsilon_1(\infty)]^{1/2}}{\epsilon_1(\infty) + 2} x a(x), \quad (1.17)$$

$$b_{32} = \omega_{T2} \frac{\epsilon_0^{1/2} [\epsilon_2(0) - \epsilon_2(\infty)]^{1/2}}{\epsilon_2(\infty) + 2} (1-x) a(x), \quad (1.18)$$

$$b_{33} = \epsilon_0 \gamma(x), \quad (1.19)$$

where

$$a(x) = \frac{[\epsilon_1(\infty) + 2][\epsilon_2(\infty) + 2]}{[\epsilon_2(\infty) + 2]x + [\epsilon_1(\infty) + 2](1-x)}, \quad (1.20)$$

and

free component ($\mathbf{w} = \mathbf{w}_L + \mathbf{w}_T$, $\nabla \times \mathbf{w}_L = 0$, $\nabla \cdot \mathbf{w}_T = 0$) and further take account of the condition

$$\nabla \cdot \mathbf{D} = \nabla \cdot (\epsilon_0 \mathbf{E} + \mathbf{P}) = 0. \quad (1.22)$$

From this we then have

$$\mathbf{E} = -(b_{31} \mathbf{w}_{1L} + b_{32} \mathbf{w}_{2L}) / (\epsilon_0 + b_{33}). \quad (1.23)$$

Equations (1.8) and (1.9) can be separated into irrotational and divergence-free components. They are

$$\ddot{\mathbf{w}}_{1T} = b_{11} \mathbf{w}_{1T} + b_{12} \mathbf{w}_{2T}, \quad (1.24)$$

$$\ddot{\mathbf{w}}_{2T} = b_{21} \mathbf{w}_{1T} + b_{22} \mathbf{w}_{2T}, \quad (1.25)$$

and

$$\ddot{\mathbf{w}}_{1L} = b'_{11} \mathbf{w}_{1L} + b'_{12} \mathbf{w}_{2L}, \quad (1.26)$$

$$\ddot{\mathbf{w}}_{2L} = b'_{21} \mathbf{w}_{1L} + b'_{22} \mathbf{w}_{2L}. \quad (1.27)$$

In these equations the following relations hold for the coefficients b :

$$b'_{11} = b_{11} - \frac{b_{13}b_{31}}{\epsilon_0 + b_{33}}, \quad (1.28)$$

$$b'_{12} = b_{12} - \frac{b_{13}b_{32}}{\epsilon_0 + b_{33}}, \quad (1.29)$$

$$b'_{21} = b_{21} - \frac{b_{23}b_{31}}{\epsilon_0 + b_{33}}, \quad (1.30)$$

$$b'_{22} = b_{22} - \frac{b_{23}b_{32}}{\epsilon_0 + b_{33}}. \quad (1.31)$$

If we call the frequencies of the transverse and longitudinal waves ω_T and ω_L , respectively (these are just the limiting frequencies of the corresponding optical branches as k goes to zero), we finally have

$$\begin{vmatrix} b_{11} + \omega_T^2 & b_{12} \\ b_{21} & b_{22} + \omega_T^2 \end{vmatrix} = 0 \quad (1.32)$$

and

$$\begin{vmatrix} b'_{11} + \omega_L^2 & b'_{12} \\ b'_{21} & b'_{22} + \omega_L^2 \end{vmatrix} = 0. \quad (1.33)$$

There are two solutions of Eq. (1.32), ω_{1T} and ω_{2T} , which are TO-phonon frequencies of the ternary mixed crystal, and ω_{1L} and ω_{2L} in (1.31) are LO-phonon frequencies.

Using Eqs. (1.32) and (1.33), we have calculated the TO- and LO-mode frequencies of the mixed crystals $\text{Se}_x\text{S}_{1-x}\text{Zn}$, $\text{Al}_x\text{Ga}_{1-x}\text{As}$, $\text{K}_{1-x}\text{Rb}_x\text{Cl}$, $\text{GaAs}_{1-x}\text{Sb}_x$, and $\text{InAs}_{1-x}\text{Sb}_x$. The results are shown in Fig. 1.

There are two distinct classes of behavior shown schematically in Fig. 1. The case of (a) and (b) in Fig. 1 is well-known two-mode behavior. There are two TO-LO-mode pairs and each TO-LO-mode pair for the end members degenerates to an impurity mode.

On the other hand, (c)–(e) in Fig. 1 belong to another case. In this case the above LO-TO-mode pair ω_{2T} and ω_{2L} , which has higher frequencies, is dominant. The TO- and LO-phonon frequencies shift continuously from ω_{T1} and ω_{L1} of crystal AC at one end to ω_{T2} and ω_{L2} of crystal BC at the other end for the main pair. Another LO-TO-mode pair ω_{1T} and ω_{1L} , whose frequencies are lower, is not important, because its oscillator strength is weak.¹³ So we call it the one-mode case.

It is interesting to point out the criterion for the one- and two-mode behavior. In our calculation, it can be given by

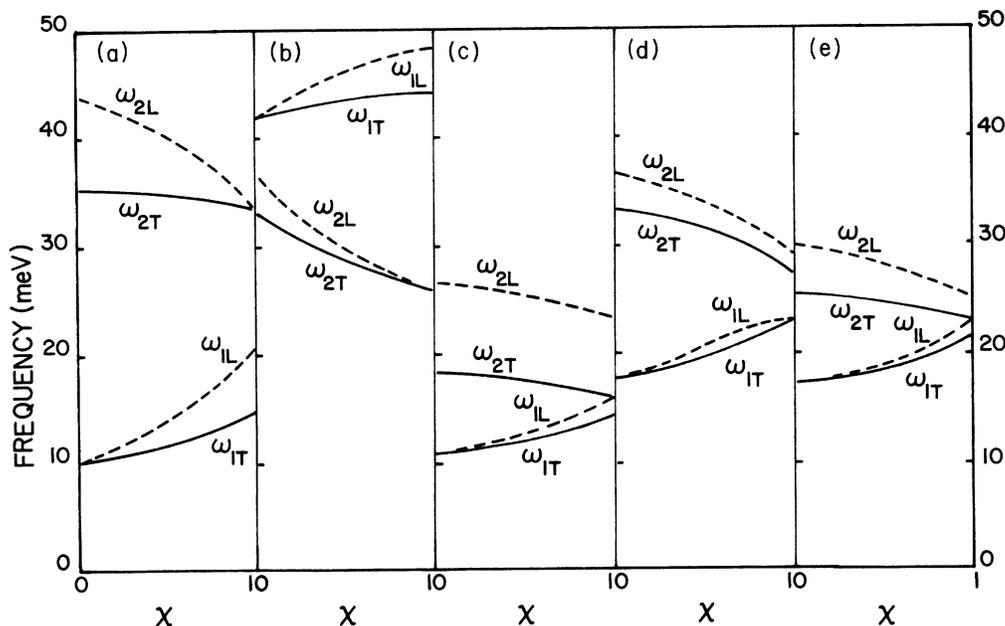


FIG. 1. Calculated eigenfrequencies of longitudinal (dashed line) and transverse (solid line) optical modes of several ternary mixed crystals: (a) $\text{Se}_x\text{S}_{1-x}\text{Zn}$, (b) $\text{Al}_x\text{Ga}_{1-x}\text{As}$, (c) $\text{K}_{1-x}\text{Rb}_x\text{Cl}$, (d) $\text{GaAs}_{1-x}\text{Sb}_x$, (e) $\text{ZnAs}_{1-x}\text{Sb}_x$. The parameters of the binary crystals are taken from Ref. 22.

$$\omega_{L2}^2 \frac{\epsilon_2(\infty)}{\epsilon_2(0)} \frac{\epsilon_2(0)+2}{\epsilon_2(\infty)+2} \frac{m_C}{m_C+m_B} > \omega_{L1}^2, \quad \text{for } \omega_{L2} > \omega_{L1} \quad (1.34)$$

$$\omega_{L1}^2 \frac{\epsilon_1(\infty)}{\epsilon_1(0)} \frac{\epsilon_2(0)+2}{\epsilon_1(\infty)+2} \frac{m_C}{m_C+m_B} > \omega_{L2}^2, \quad \text{for } \omega_{L1} > \omega_{L2}. \quad (1.35)$$

If relations (1.34) or (1.35) are satisfied, the ternary mixed crystal $A_x B_{1-x} C$ exhibits two-mode behavior, otherwise we get one-mode behavior.

An analogous criterion has also been given by Chang and Mitra.¹

II. ELECTRON-PHONON INTERACTION

First, we express the displacements w_{1L} and w_{2L} in the scalar form along the direction of the wave vector k as follows:

$$w_{1L} = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} (A_{1\mathbf{k}} + B_{1\mathbf{k}}) + \text{H.c.}, \quad (2.1)$$

$$w_{2L} = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} (A_{2\mathbf{k}} + B_{2\mathbf{k}}) + \text{H.c.}, \quad (2.2)$$

where

$$A_{j\mathbf{k}} = A'_{j\mathbf{k}} e^{-i\omega_{jL}t} \quad (j=1,2),$$

$$B_{j\mathbf{k}} = B'_{j\mathbf{k}} e^{-i\omega_{2L}t} \quad (j=1,2).$$

In Eqs. (2.1) and (2.2), \mathbf{r} is a continuously variable coordinate. $A_{i\mathbf{k}}$ and $B_{i\mathbf{k}}$ are amplitudes dependent on the wave vector \mathbf{k} corresponding to two branches of LO-vibration frequencies ω_{1L} and ω_{2L} .

Substituting (2.1) and (2.2) into (1.26) and (1.27), we can find that the amplitudes $A_{i\mathbf{k}}$ and $B_{i\mathbf{k}}$ must observe the relations as follows:

$$B_{1\mathbf{k}} = \frac{b'_{12}}{b'_{11} + \omega_{2L}^2} B_{2\mathbf{k}} = -C_1 B_{2\mathbf{k}}, \quad (2.3)$$

$$A_{2\mathbf{k}} = \frac{b'_{11} + \omega_{1L}^2}{b'_{12}} A_{1\mathbf{k}} = C_2 A_{1\mathbf{k}}, \quad (2.4)$$

and their complex-conjugate formulas. Using these relations and replacing amplitudes $A_{i\mathbf{k}}$ by $A_{\mathbf{k}}$ and $B_{2\mathbf{k}}$ by $B_{\mathbf{k}}$, Eqs. (2.1) and (2.2) can be written as

$$w_{1L} = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} (A_{\mathbf{k}} - C_1 B_{\mathbf{k}}) + \text{H.c.}, \quad (2.5)$$

$$w_{2L} = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} (C_2 A_{\mathbf{k}} + B_{\mathbf{k}}) + \text{H.c.} \quad (2.6)$$

Carrying out inverse Fourier transformations of (2.5) and (2.6), we have

$$A_{\mathbf{k}} + A_{-\mathbf{k}}^* = \frac{1}{1+C_1 C_2} \int \frac{d\tau}{\sqrt{V}} e^{-i\mathbf{k}\cdot\mathbf{r}} (w_{1L} + C_1 w_{2L}), \quad (2.7)$$

$$B_{\mathbf{k}} + B_{-\mathbf{k}}^* = \frac{1}{1+C_1 C_2} \int \frac{d\tau}{\sqrt{V}} e^{-i\mathbf{k}\cdot\mathbf{r}} (-C_2 w_{1L} + w_{2L}), \quad (2.8)$$

and then

$$A_{\mathbf{k}} = \frac{1}{2\omega_{1L}(1+C_1 C_2)} \times \int \frac{d\tau}{\sqrt{V}} e^{-i\mathbf{k}\cdot\mathbf{r}} [\omega_{1L}(w_{1L} + C_1 w_{2L}) + i(\dot{w}_{1L} + C_1 \dot{w}_{2L})], \quad (2.9)$$

$$A_{-\mathbf{k}}^* = \frac{1}{2\omega_{1L}(1+C_1 C_2)} \times \int \frac{d\tau}{\sqrt{V}} e^{-i\mathbf{k}\cdot\mathbf{r}} [\omega_{1L}(w_{1L} + C_1 w_{2L}) - i(\dot{w}_{1L} + C_1 \dot{w}_{2L})], \quad (2.10)$$

$$B_{\mathbf{k}} = \frac{1}{2\omega_{2L}(1+C_1 C_2)} \times \int \frac{d\tau}{\sqrt{V}} e^{-i\mathbf{k}\cdot\mathbf{r}} [\omega_{2L}(-C_2 w_{1L} + w_{2L}) + i(-C_2 \dot{w}_{1L} + \dot{w}_{2L})], \quad (2.11)$$

$$B_{-\mathbf{k}}^* = \frac{1}{2\omega_{2L}(1+C_1 C_2)} \times \int \frac{d\tau}{\sqrt{V}} e^{-i\mathbf{k}\cdot\mathbf{r}} [\omega_{2L}(-C_2 w_{1L} + w_{2L}) - i(-C_2 \dot{w}_{1L} + \dot{w}_{2L})]. \quad (2.12)$$

Now let us perform quantization for the system. If we consider the coordinates s_1 and s_2 and the corresponding canonically conjugate momenta p_1 and p_2 as operators, they then satisfy the commutation relations

$$[p_n, s_1] = \frac{\hbar}{i} \delta_{nl} \delta(\mathbf{r} - \mathbf{r}'), \quad (2.13)$$

$$[s_n, s_l] = 0, \quad (2.14)$$

$$[p_n, p_l] = 0, \quad (2.15)$$

with $n, l = 1, 2$. Using these relations, one can verify that the following commutation relations also hold:

$$\dot{w}_n(\mathbf{r}) w_l(\mathbf{r}') - w_l(\mathbf{r}') \dot{w}_n(\mathbf{r}) = \frac{\hbar}{i} \delta_{nl} \delta(\mathbf{r} - \mathbf{r}') \quad (n, l = 1, 2), \quad (2.16)$$

$$w_n(\mathbf{r}) w_l(\mathbf{r}') - w_l(\mathbf{r}') w_n(\mathbf{r}) = 0 \quad (n, l = 1, 2), \quad (2.17)$$

$$\dot{w}_n(\mathbf{r}) \dot{w}_l(\mathbf{r}') - \dot{w}_l(\mathbf{r}') \dot{w}_n(\mathbf{r}) = 0 \quad (n, l = 1, 2). \quad (2.18)$$

Using relations (2.16)–(2.18) together with expressions (2.9)–(2.12), it is easy to verify that

$$[A_{\mathbf{k}}, A_{\mathbf{k}'}] = \frac{\hbar}{2\omega_{1L}} \frac{1+c_1^2}{(1+C_1 C_2)^2} \delta_{\mathbf{k}\mathbf{k}'}, \quad (2.19)$$

$$[B_{\mathbf{k}}, B_{\mathbf{k}'}] = \frac{\hbar}{2\omega_{2L}} \frac{1+C_2^2}{(1+C_1C_2)^2} \delta_{\mathbf{k}\mathbf{k}'}, \quad (2.20)$$

$$[A_{\mathbf{k}}, A_{\mathbf{k}'}] = [A_{\mathbf{k}}^*, A_{\mathbf{k}'}^*] = [B_{\mathbf{k}}, B_{\mathbf{k}'}] = [B_{\mathbf{k}}^*, B_{\mathbf{k}'}^*] = 0.$$

We next introduce new dimensionless amplitudes $a_{\mathbf{k}}$ and $b_{\mathbf{k}}$ in place of the amplitudes $A_{\mathbf{k}}$ and $B_{\mathbf{k}}$:

$$A_{\mathbf{k}} = \frac{\hbar}{2\omega_{1L}} \frac{1+C_1^2}{(1+C_1C_2)^2} a_{\mathbf{k}} = d_1 a_{\mathbf{k}}, \quad (2.21)$$

$$B_{\mathbf{k}} = \frac{\hbar}{2\omega_{2L}} \frac{1+C_2^2}{(1+C_1C_2)^2} b_{\mathbf{k}} = d_2 b_{\mathbf{k}}. \quad (2.22)$$

Starting with the commutations (2.19) and (2.20) we then obtain

$$[a_{\mathbf{k}}, a_{\mathbf{k}'}^\dagger] = \delta_{\mathbf{k}\mathbf{k}'}, \quad (2.23)$$

$$[b_{\mathbf{k}}, b_{\mathbf{k}'}^\dagger] = \delta_{\mathbf{k}\mathbf{k}'}, \quad (2.24)$$

and

$$[a_{\mathbf{k}}, a_{\mathbf{k}'}] = [a_{\mathbf{k}}^\dagger, a_{\mathbf{k}'}^\dagger] = 0, \quad (2.25)$$

$$[b_{\mathbf{k}}, b_{\mathbf{k}'}] = [b_{\mathbf{k}}^\dagger, b_{\mathbf{k}'}^\dagger] = 0, \quad (2.26)$$

$$[a_{\mathbf{k}}, b_{\mathbf{k}'}] = [a_{\mathbf{k}}, b_{\mathbf{k}'}^\dagger] = [a_{\mathbf{k}}^\dagger, b_{\mathbf{k}'}] = [a_{\mathbf{k}}^\dagger, b_{\mathbf{k}'}^\dagger] = 0. \quad (2.27)$$

Substituting (2.21) and (2.22) into (2.5) and (2.6), we can write the dimensionless displacements w_{1L} and w_{2L} in the form

$$w_{1L} = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} [d_1(a_{\mathbf{k}} + a_{-\mathbf{k}}^\dagger) - C_1 d_2(b_{\mathbf{k}} + b_{-\mathbf{k}}^\dagger)], \quad (2.28)$$

$$w_{2L} = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} [C_2 d_1(a_{\mathbf{k}} + a_{-\mathbf{k}}^\dagger) + d_2(b_{\mathbf{k}} + b_{-\mathbf{k}}^\dagger)]. \quad (2.29)$$

Now we consider the total polarization P described by Eq. (1.10). From the above two equations together with (1.10) and (1.23) we obtain

$$P = \frac{\epsilon_0}{\epsilon_0 + b_{33}} \sum_{\mathbf{k}} \frac{1}{\sqrt{V}} e^{i\mathbf{k}\cdot\mathbf{r}} [d_1(b_{31} + C_2 b_{32})(a_{\mathbf{k}} + a_{-\mathbf{k}}^\dagger) + d_2(-C_1 b_{31} + b_{32}) \times (b_{\mathbf{k}} + b_{-\mathbf{k}}^\dagger)]. \quad (2.30)$$

The interaction energy of an electron with the polarized medium is

$$H_I = -\frac{e}{4\pi\epsilon_0} \int d\tau \mathbf{P} \cdot \frac{(\mathbf{r} - \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|^3}, \quad (2.31)$$

where \mathbf{r}' is the electron coordinate. The integral in (2.31) is easy to carry out. since

$$\int e^{\pm i\mathbf{k}\cdot\mathbf{r}} \frac{\mathbf{r} - \mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|^3} d\tau = \mp 4\pi i \frac{\mathbf{k}}{k^2} e^{\mp i\mathbf{k}\cdot\mathbf{r}'}, \quad (2.32)$$

we then have for the electron-phonon interaction by inserting (2.30) in (2.31),

$$H_I = \sum_{\mathbf{k}} \frac{g_1}{k\sqrt{V}} (a_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} - a_{\mathbf{k}}^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}}) + \sum_{\mathbf{k}} \frac{g_2}{k\sqrt{V}} (b_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} - b_{\mathbf{k}}^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}}). \quad (2.33)$$

Here,

$$g_1 = \frac{ie}{(\epsilon_0 + b_{33})} \left[\frac{\hbar}{2\omega_{1L}} \frac{1+C_1^2}{(1+C_1C_2)^2} \right]^{1/2} (b_{31} + C_2 b_{32}), \quad (2.34)$$

$$g_2 = \frac{ie}{(\epsilon_0 + b_{33})} \left[\frac{\hbar}{2\omega_{2L}} \frac{1+C_2^2}{(1+C_1C_2)^2} \right]^{1/2} (-C_1 b_{31} + b_{32}). \quad (2.35)$$

In Eq. (2.33) we have already replaced the symbol of the electron coordinate \mathbf{r}' by \mathbf{r} for convenience.

It is natural and easy to verify that the electron-phonon-interaction Hamiltonian H_I of $A_x B_{1-x} C$ in (2.33) will degenerate into that of the binary crystal BC when $x=0$ and that of the crystal AC when $x=1$.

Finally, the total Hamiltonian operator for an electron-phonon system takes the form

$$H = H_0 + H_I, \quad (2.36)$$

$$H_0 = \frac{\hbar^2}{2m} \nabla^2 + \sum_{\mathbf{k}} \hbar\omega_{1L} (a_{\mathbf{k}} a_{\mathbf{k}}^\dagger + \frac{1}{2}) + \sum_{\mathbf{k}} \hbar\omega_{2L} (b_{\mathbf{k}} b_{\mathbf{k}}^\dagger + \frac{1}{2}), \quad (2.37)$$

$$H_I = \sum_{\mathbf{k}} g_1 (a_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} - a_{\mathbf{k}}^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}}) + \sum_{\mathbf{k}} g_2 (b_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} - b_{\mathbf{k}}^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}}). \quad (2.38)$$

Here, m is the band mass of the electron.

III. FRÖHLICH-COUPPING PARAMETER

Now let us apply the Schrödinger perturbation theory to the electron-phonon system in a ternary mixed crystal. We consider that the eigenenergies of a system described by the Hamiltonian H_0 are changed by the introduction of a perturbation H_I and then calculate the approximation of the energy to second-order correction.

Let $|\mathbf{q}, 0\rangle$ denote the zero-order wave function which is the plane wave $|\mathbf{q}\rangle$, extended by the vacuum state $|0\rangle$ of the phonon system. The corresponding zero-order energy is $E^0(\mathbf{q}) = \hbar^2 q^2 / 2m$; here m is the conduction-band mass. The approximation of the energy of the state of wave number \mathbf{q} can be written as

$$E(\mathbf{q}) = E^0(\mathbf{q}) + \langle \mathbf{q}, 0 | H_I | \mathbf{q}, 0 \rangle + \sum_{\mathbf{k}, l} \frac{|\langle \mathbf{q} - \mathbf{k}, 1_{\mathbf{k}} | H_I | \mathbf{q}, 0 \rangle|^2}{E^0(\mathbf{q}) - E^0(\mathbf{q} - \mathbf{k}) - \hbar\omega_{lL}}. \quad (3.1)$$

In Eq. (3.1), the states over which the additional terms are summed are states in which an optical phonon of energy $\hbar\omega_{lL}$ and wave number \mathbf{k} is emitted. Here, ω_{lL} is the

frequency of one of two branches of LO phonons ($i = 1, 2$), which is defined by Eq. (1.33).

It is obvious that the first-order correction in (3.1) vanishes, i.e.,

$$\langle \mathbf{q}, 0 | H_I | \mathbf{q}, 0 \rangle = 0. \quad (3.2)$$

The matrix element appearing in the third term of Eq. (3.1) is

$$\langle \mathbf{q} - \mathbf{k}, 1_{\mathbf{k}} | H_I | \mathbf{q}, 0 \rangle = g_i, \quad i = 1 \text{ or } 2. \quad (3.3)$$

Replacing the sum over \mathbf{k} by an integral over the Brillouin zone in \mathbf{k} space, we have

$$\begin{aligned} E(\mathbf{q}) &= \frac{\hbar^2 q^2}{2m} \\ &+ \frac{V}{(2\pi)^3} \int d\tau |g_1|^2 \left[\frac{\hbar^2}{2m} (2\mathbf{q} \cdot \mathbf{k} - k^2) - \hbar\omega_{1L} \right]^{-1} \\ &+ \frac{V}{(2\pi)^3} \int d\tau |g_2|^2 \left[\frac{\hbar^2}{2m} (2\mathbf{q} \cdot \mathbf{k} - k^2) - \hbar\omega_{2L} \right]^{-1}. \end{aligned} \quad (3.4)$$

We restrict ourselves in the following to the energy range $\hbar^2 k^2 / 2m < \hbar\omega_L$. Thereby the integrand in (3.4) can be expanded for small q .¹⁵ The result is

$$\begin{aligned} E(\mathbf{q}) &= \frac{\hbar^2 q^2}{2m} - \alpha_1 \left[\hbar\omega_{1L} + \frac{\hbar^2 q^2}{12m} \right] - \alpha_2 \left[\hbar\omega_{2L} + \frac{\hbar^2 q^2}{12m} \right] \\ &= -\alpha_1 \hbar\omega_{1L} - \alpha_2 \hbar\omega_{2L} + \frac{\hbar^2 q^2}{2m^*}, \end{aligned} \quad (3.5)$$

where

$$\alpha_1 = \frac{1}{4\pi\hbar^2\omega_{1L}^2} \left[\frac{2m\omega_{1L}}{\hbar} \right]^{1/2} |g_1|^2, \quad (3.6)$$

$$\alpha_2 = \frac{1}{4\pi\hbar^2\omega_{2L}^2} \left[\frac{2m\omega_{2L}}{\hbar} \right]^{1/2} |g_2|^2, \quad (3.7)$$

and

$$m^* = m \left[1 - \frac{\alpha_1 + \alpha_2}{6} \right]^{-1}. \quad (3.8)$$

The energy given by (3.4) and (3.5) is the energy of a polaron. Compared with the corresponding values for a free electron, its energy is reduced by $\alpha_1 \hbar\omega_{1L} + \alpha_2 \hbar\omega_{2L}$ and its mass by a factor $[1 - (\alpha_1 + \alpha_2)/6]^{-1}$. They are contributed by the interaction between the electron and the two branches of LO phonons of frequencies ω_{1L} and ω_{2L} , respectively. It will be discussed later.

It is necessary to point out that the band mass m is known to be connected with the composition x of the mixed crystal. There are many investigations on the compositional dependence of the electron effective mass in the mixed crystals.¹⁶⁻²⁰ Nichlas *et al.*²¹ have measured cyclotron resonance and infrared absorption in the complete range of alloys $\text{InAs}_x\text{P}_{1-x}$ in the region of the direct gap, and have found that the effective mass is an

almost linear function of the composition x . Adachi,⁵ in a recent publication on $\text{Al}_x\text{Ga}_{1-x}\text{As}$ mixed crystals, has also pointed out that the generally acceptable m value has a linear relationship with the composition x and can be derived from corresponding binary values. Therefore, at this time we use the following linear interpolation formula to calculate the electron band mass appearing in (3.5)–(3.8):

$$m_{ABC} = xm_{AC} + (1-x)m_{BC}, \quad (3.9)$$

where m_{ABC} , m_{AC} , and m_{BC} are the band masses in the ternary mixed crystal $A_xB_{1-x}C$, the binary crystal AC , and BC , respectively.

The parameters α_1 and α_2 defined by (3.6) and (3.7) are the Fröhlich electron-phonon coupling parameters related to two varied branches of LO-phonon modes, respectively. We have calculated the coupling parameters for the ternary mixed crystals $\text{Se}_x\text{S}_{1-x}\text{Zn}$, $\text{Al}_x\text{Ga}_{1-x}\text{As}$, $\text{K}_{1-x}\text{Rb}_x\text{Cl}$, $\text{GaAs}_{1-x}\text{Sb}_x$, and $\text{InAs}_{1-x}\text{Sb}_x$ as functions of the composition x . The results are plotted in Fig. 2.

Similar to the behavior of phonon modes, there are two kinds of coupling-parameter behavior shown in Fig. 2. $\text{Se}_x\text{S}_{1-x}\text{Zn}$ and $\text{Al}_x\text{Ga}_{1-x}\text{As}$, Figs. 2(a) and 2(b), belong to the first case, which should be called the two-mode case. In this case, α_1 and α_2 can be called the AC - and BC -like mode coupling parameters, respectively. When $x = 0$, the mixed crystal $A_xB_{1-x}C$ degenerates into the binary crystal BC and the electron-phonon coupling parameter, of course, becomes that of the binary crystal BC , i.e., α_{BC} . On the other hand, when $x = 1$ the coupling parameter becomes α_{AC} . One can see this from Figs. 2(a) and 2(b). With increasing the composition x of the element A , the AC -like parameter α_1 increases from zero monotonically and then up to the end value α_{AC} at $x = 1$. On the contrary, the BC -like parameter decreases from α_{BC} with the composition $1 - x$ of the element B and then down to zero. $\alpha_1 + \alpha_2$ gets the value of α_{AC} or α_{BC} at an end value of x and gets its minimum at some middle value of x because the contributions of A and B ions to the polarization field cancel each other out.

The second case, for $\text{K}_{1-x}\text{Rb}_x\text{Cl}$, $\text{GaAs}_{1-x}\text{Sb}_x$, and $\text{InAs}_{1-x}\text{Sb}_x$ [see Figs. 2(c)–2(e)], is the one-mode behavior. In this case, the contributions of the two branches of LO-phonon modes to the electron-phonon interaction are very different. The parameter α_2 , corresponding to the higher frequency ω_{2L} , shifts continuously from α_{AC} at one end of $x = 0$ to α_{BC} at the other end of $x = 1$ and has a minimum at some middle composition. α_1 of lower frequency ω_{1L} is zero for either $x = 0$ or 1 and has a maximum at some middle composition. It is found from (c)–(e) of Figs. 1 and 2 that α_2 and $\hbar\omega_{2L}$ are much larger than α_1 and $\hbar\omega_{1L}$, respectively, so that the self-trapping energy $\alpha_2 \hbar\omega_{2L}$ is one order of magnitude larger than $\alpha_1 \hbar\omega_{1L}$. This shows that α_2 is dominant for the electron-phonon interaction but α_1 is not important. Therefore, we call the branch of higher frequency the main branch and that of lower frequency the weak branch. We can consider that ω_{2L} is due to the vibration of the A and the B ions against the C ions but ω_{1L} due to

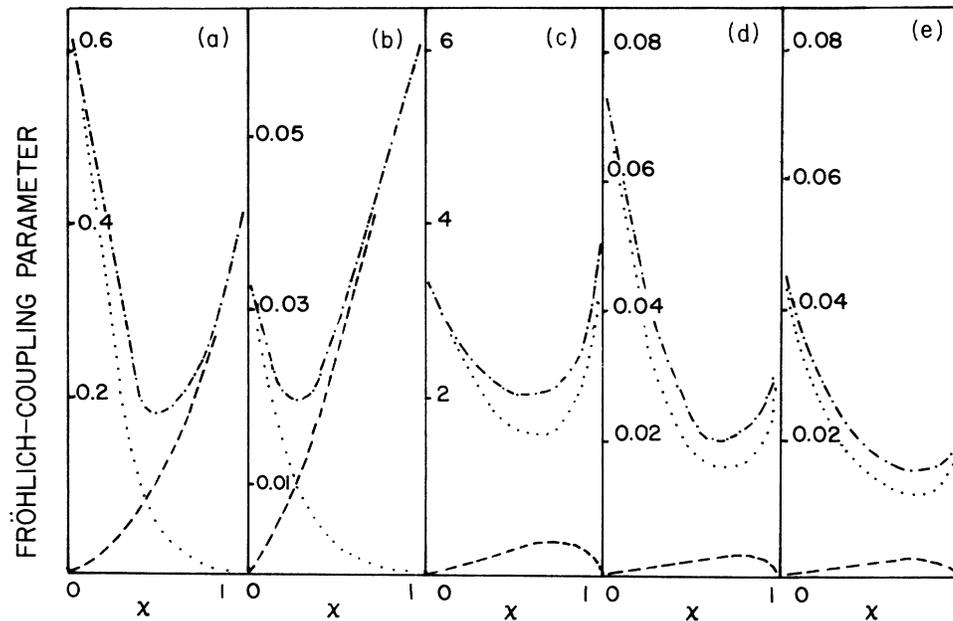


FIG. 2. Fröhlich-coupling parameters α_1 (dotted line), α_2 (dashed line), and $\alpha_1 + \alpha_2$ (dot-dashed line) as functions of composition x for several ternary mixed crystals: (a) $\text{Se}_x\text{S}_{1-x}\text{Zn}$, (b) $\text{Al}_x\text{Ga}_{1-x}\text{As}$, (c) $\text{K}_{1-x}\text{Rb}_x\text{Cl}$, (d) $\text{GaAs}_{1-x}\text{Sb}_x$, (e) $\text{ZnAs}_{1-x}\text{Sb}_x$. The parameters of the binary crystals are taken from Ref. 22.

the A ions against the B ions. As a result, α_2 is dominant but α_1 is negligible. As for the total contribution to the electron-phonon interaction, $\alpha_1 + \alpha_2$ has a minimum at some middle composition.

SUMMARY

We have used the REI-like model and the Born-Huang theory to investigate the modes of lattice vibration and the electron-phonon interaction for a ternary mixed crystal $A_xB_{1-x}C$. The eigenfrequencies of the long-wavelength optical vibration for several ternary mixed crystals are calculated. We have also derived the Hamiltonian operator describing the electron-phonon interaction in a mixed crystal and calculated the Fröhlich-coupling parameters for some mixed-crystal materials.

Our results show that there are two types of phonon-mode and electron-phonon-interaction behavior—the two- and the one-mode behavior. In the two-mode case, the two branches of LO phonons, and thereby Fröhlich-coupling parameters, can be called AC - and BC -like, respectively. The Fröhlich-coupling parameter for each of two branches is the binary value at an end (e.g., α_{AC} , for

$x = 1$) and goes down to zero at the other end. For the one-mode case, on the other hand, one of two branches is dominant but another is negligible. The coupling parameter of the main branch shifts continuously from α_{BC} at an end to α_{AC} at the other end. The weak-branch parameter is zero at the two ends.

It also is pointed out that either phonon frequencies or electron-phonon interaction in the ternary mixed crystals derived here is equivalent to that in corresponding binary crystals at $x = 0$ and 1.

Our results for phonon modes qualitatively agree with previous theoretical and experimental work. The results for the electron-phonon interaction are new results. We have not found any experimental results to be compared.

Finally, it must be pointed out that the continuum approximation is being made in discussing the Fröhlich parameter analogous to that which is done in standard treatments of the electron-phonon interaction in the diatomic case, and also it is assumed that there is no dispersion of the optical mode frequencies (despite the fact that in the case of a mixed crystal, lattice translational symmetry is destroyed, and it is not very meaningful to discuss the dispersion).

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