Effects of the alloy compositions on the phonon-polaritons in ternary mixed crystals*

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Composition dependence of bulk and surface phonon-polaritons in ternary mixed crystals are studied in the framework of the modified random-element-isodisplacement model and the Born-Huang approximation. The numerical results for several Ⅱ - Ⅵ and Ⅲ - Ⅴ compound systems are performed, and the polariton frequencies as functions of the compositions for ternary mixed crystals $Al_xGa_{1-x}As$, GaP_xAs_{1-x}, ZnS_xSe_{1-x}, $GaAs_xSb_{1-x}$, $Ga_xIn_{1-x}P$, and $Zn_xCd_{1-x}S$ as examples are given and discussed. The results show that the dependence of the energies of two branches of bulk phonon-polaritons which have phonon-like characteristics, and surface phonon-polaritons on the compositions of ternary mixed crystals are nonlinear and different from those of the corresponding binary systems.

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The study of phonon-polaritons is an interesting field both in experiments and theories [1-8]. The properties of ternary mixed crystal (TMC) systems have also been paid much attention [9-12], because its application in artificial layered materials enables one to modify the optical and electric properties of the systems by altering the compositions of the alloys. The novel properties of optical phonons in TMCs, such as the composition dependence of the longitudinal-transverse energies, contribute new characteristics of bulk phonon-polaritons. Besides, as the counterparts of bulk phonon-polaritons the surface phonon-polaritons in TMCs present some new features, which are different from those of correspongding binary crystals. The further investigations on the composition dependence of bulk and surface phonon-polaritons in TMC systems are therefore needed.

In this paper we study the composition effect on the phonon-polariton properties in polar TMCs in the framework of the modified random-element-isodisplacement model and the Born-Huang approximation. Both the frequencies of the bulk and surface phonon-polaritons are calculated numerically for some typical TMC systems. The nonlinear effect of the composition on the phonon-polariton energies is found out.

We first explore the properties of bulk phonon-polaritons in polar TMCs. An electromagnetic wave is considered to propagate in a polar TMC $A_xB_{1-x}C$ of cubic symmetry, which couples with long-wavelength transverse optical (TO) modes of lattice vibrations and forms phonon-polaritons. For simplicity, the dielectric function $\varepsilon(k, \omega)$ is considered as isotropic for the bulk TMC material. The propagation of phonon-polaritons coupled by the electromagnetic radiation with long wavelengths TO phonon field in the TMCs can be described by linking the Maxwell's equations with the Born-Huang-like equations,

$$\nabla \times \boldsymbol{E} = -\mu_0 \frac{\partial \boldsymbol{H}}{\partial t} , \tag{1}$$

$$\nabla \times \boldsymbol{H} = \frac{\partial}{\partial t}\left(\varepsilon_0 \boldsymbol{E} + \boldsymbol{P}\right), \tag{2}$$

$$\nabla \cdot \boldsymbol{D} = 0 , \tag{3}$$

$$\nabla \cdot \boldsymbol{H} = 0 , \tag{4}$$

$$\ddot{\boldsymbol{W}}_1 = b_{11}\boldsymbol{W}_1 + b_{12}\boldsymbol{W}_2 + b_{13}\boldsymbol{E} , \tag{5}$$

$$\ddot{\boldsymbol{W}}_2 = b_{21}\boldsymbol{W}_1 + b_{22}\boldsymbol{W}_2 + b_{23}\boldsymbol{E} , \tag{6}$$

$$\boldsymbol{P} = b_{31}\boldsymbol{W}_1 + b_{32}\boldsymbol{W}_2 + b_{33}\boldsymbol{E} . \tag{7}$$

here, $\boldsymbol{E}$, $\boldsymbol{H}$ are the macroscopic electric and magnetic fields, respectively; $\boldsymbol{P}$ is the polarization; μ_0 and ε_0 are the vacuum magnetic permeability and the dielectric constant, respectively; $\boldsymbol{W}_1 = \mu_1^{1/2}\boldsymbol{s}_1$ and $\boldsymbol{W}_2 = \mu_2^{1/2}\boldsymbol{s}_2$, where $\boldsymbol{s}_1 = \boldsymbol{u}_A - \boldsymbol{u}_C$ and $\boldsymbol{s}_2 = \boldsymbol{u}_B - \boldsymbol{u}_C$ are the relative displacements of the A-C and B-C ion

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pairs, respectively; μ_1 and μ_2 are the corresponding reduced masses; $\boldsymbol{u}_A$, $\boldsymbol{u}_B$, and $\boldsymbol{u}_C$ are the displacements of the ions A, B, and C, respectively. The dynamical coefficients b_{ij}(i, j=1,2, 3) in equations (5)-(7) have been determined by the previous work[13].

Let us assume the solutions of equations (1)-(7) as following form:

$$\boldsymbol{W}, \boldsymbol{P}, \boldsymbol{E}, \boldsymbol{H} \propto \exp[i(k \cdot r - \omega t)], \tag{8}$$

where ω and $\boldsymbol{k}$ are the frequency and wave-vector of the phonon-polaritons respectively.

Using the solutions of form (8) in Eqs. (1)-(7), one can obtain the relation between the macroscopic electric field $\boldsymbol{E}$ and the polarization $\boldsymbol{P}$:

$$\boldsymbol{P} = \left\{ b_{31}\left[\frac{b_{23}b_{12} - b_{13}\left(b_{22}+\omega^2\right)}{\left(b_{11}+\omega^2\right)\left(b_{22}+\omega^2\right) - b_{12}b_{21}} \right] + b_{32}\left[\frac{b_{13}b_{21} - b_{23}\left(b_{11}+\omega^2\right)}{\left(b_{11}+\omega^2\right)\left(b_{22}+\omega^2\right) - b_{12}b_{21}} \right] + b_{33} \right\} \boldsymbol{E} \ . \tag{9}$$

Then the dielectric function of the TMC can be written as,

$$\varepsilon(\omega) = 1 + \chi(\omega)/\varepsilon_0 \, , \tag{10}$$

where $\chi(\omega) = \left\{ b_{31}\left[\frac{b_{23}b_{12} - b_{13}\left(b_{22}+\omega^2\right)}{\left(b_{11}+\omega^2\right)\left(b_{22}+\omega^2\right) - b_{12}b_{21}} \right] + b_{32}\left[\frac{b_{13}b_{21} - b_{23}\left(b_{11}+\omega^2\right)}{\left(b_{11}+\omega^2\right)\left(b_{22}+\omega^2\right) - b_{12}b_{21}} \right] + b_{33} \right\}$. (11)

The implied dispersion relation of bulk phonon-polaritons is then given by

$$\frac{k^2c^2}{\omega^2} = 1 + \chi(\omega)/\varepsilon_0 \, . \tag{12}$$

Solving equation (12) with (11), one can obtain generally three frequencies of the phonon-polariton propagating in the TMC system as functions of the composition x.

The numerical computations of the frequencies of bulk phonon-polariton modes have been performed for several II-VI and III-V semiconductor TMC systems. For example, the energies of the bulk phonon-polaritons as functions of the composition x in the TMCs $Al_xGa_{1-x}As$, GaP_xAs_{1-x}, ZnS_xSe_{1-x}, $GaAs_xSb_{1-x}$, $Ga_xIn_{1-x}P$, and $Zn_xCd_{1-x}S$ are illustrated in Fig.1, where we have chosen k = 1 for definition. The parameters used in the calculations are listed in Tab.1.

Tab.1 Optical phonon energies, dielectric constants, effective masses of electrons, and the lattice constants for binary materials. Energy is measured in meV, *m* in the electron rest mass, and the lattice constants in nm.

Materials	$\hbar\omega_{TO}$	$\hbar\omega_{LO}$	ε_0	ε_∞	m	M (a.u.)		a
AlAs[a]	44.88	50.09	10.06	8.16	0.150	26.99	74.92	0.5660[c]
GaAs[a]	33.29	36.25	13.18	10.89	0.067	69.72	74.92	0.5642[c]
GaP[b]	45.33	49.97	10.28	8.46	0.338	69.72	30.97	0.5451[c]
GaSb[b]	28.58	29.79	15.69	14.44	0.047	69.72	121.70	0.6096[c]
ZnSe[b]	25.67	30.50	8.33	5.90	0.171	65.38	78.90	0.5668[c]
ZnS[b]	34.65	44.00	8.00	5.14	0.280	65.38	32.06	0.5410[c]
CdS[b]	30.25	38.24	8.42	5.27	0.155	112.40	32.06	0.5825[c]

Notes: [a] reference [15], [b] reference [16], [c] reference [17].

Fig.1 shows clearly that there are three frequency branches of bulk phonon-polariton modes. The energies of the lowest branch, which has the photon-like characteristics for the k = 1 case, increase linearly with the increase of the composition of the Al-, P-, S-, As-, Ga-, Zn-components in the $Al_xGa_{1-x}As$, GaP_xAs_{1-x}, ZnS_xSe_{1-x}, $GaAs_xSb_{1-x}$, $Ga_xIn_{1-x}P$, and $Zn_xCd_{1-x}S$ TMC systems. Otherwise the energies of the two phonon-like branches of polaritons with the higher frequencies vary nonlinearly with the increase of the composition of the TMCs. The nonlinear property of polaritons originates mainly from the nonlinear behavior of the phonon frequencies on the composition.

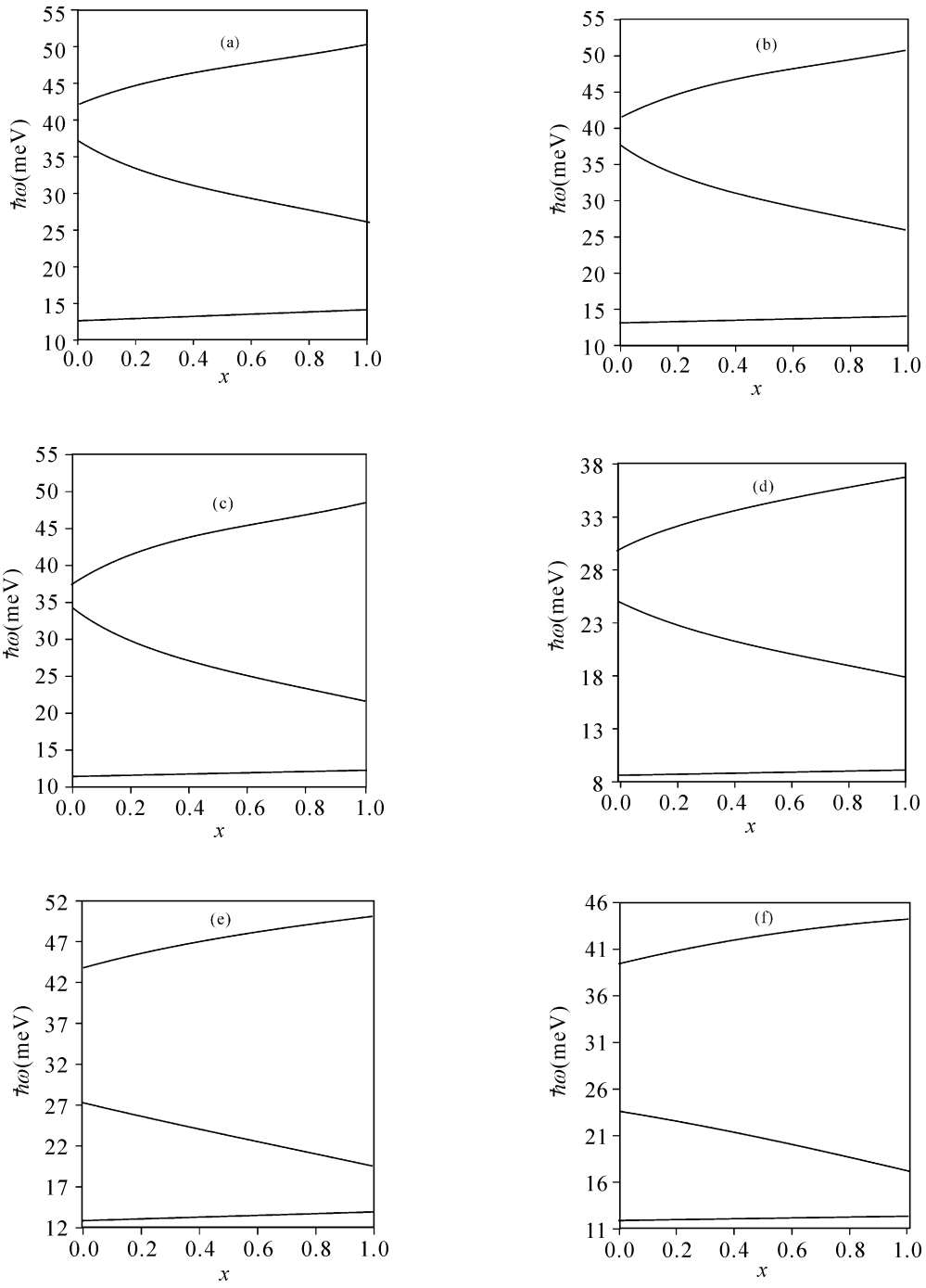

Fig.1 Energies of the bulk phonon-polaritons as functions of the compositions *x* in the TMC systems. (a) $Al_xGa_{1-x}As$, (b) GaP_xAs_{1-x}, (c) ZnS_xSe_{1-x}, (d) $GaAs_xSb_{1-x}$, (e) $Ga_xIn_{1-x}P$, and (f) $Zn_xCd_{1-x}S$ for *k*=1. The wave-vector *k* is measured in ω_{TGaAs}/c, ω_{TGaAs}/c, ω_{TZnSe}/c, ω_{TGaSb}/c, ω_{TInP}/c, and ω_{TCdS}/c, respectively. ω_{TGaAs}, ω_{TZnSe}, ω_{TGaSb}, ω_{TInP}, and ω_{TCdS} are respectively the frequencies of TO phonons of the binary crystals of GaAs, ZnSe, GaSb, InP, and CdS.

One can also see that the nonlinear property of the polariton energies on the composition in the "one-mode" behavior TMCs $GaAs_xSb_{1-x}$, $Ga_xIn_{1-x}P$, and $Zn_xCd_{1-x}S$ is much weaker than that in the "two-mode" behavior systems $Al_xGa_{1-x}As$, GaP_xAs_{1-x}, and ZnS_xSe_{1-x}.

Next we turn to consider surface phonon-polariton modes coupling with the surface phonon modes in the system. In this case we can treat the system as a semi-infinite material occupying the half space of $z < 0$ and the surface phonon-polaritons with the two-dimensional wave-vector $k_{\parallel}$ propa-

gating along the x direction, i.e. $k_x = k_{||}$ and $k_y = 0$. In the present work, we focus our attention on the surface phonon-polaritons of transverse magnetic(TM) character. The electric field $\boldsymbol{E}$ lies in the x-z plane and the magnetic field $\boldsymbol{H}$ is along the y-axis. By using a method similar to that used by Mills and Maradudin for polaritons in the films of binary crystal [14], we have obtained the dielectric function of the phonon-polaritons at the surface between semi-infinite TMC and the vacuum as follows:

$$\varepsilon(\omega)/\kappa_2 = -1/\kappa_1 , \tag{13}$$

where

$$\kappa_1^2 = k_{\|}^2 - \omega^2 / c^2 , \tag{14a}$$

$$\kappa_2^2 = k_{\|}^2 - \varepsilon(\omega)\omega^2 / c^2. \tag{14b}$$

Substituting Eq. (14) into (13) one can obtain the implied dispersion equation for the surface phonon-polaritons

$$k_{\|}^2 c^2 / \omega^2 = \varepsilon(\omega)/[1+\varepsilon(\omega)] , \tag{15}$$

where $\varepsilon(\boldsymbol{\omega})$ is the dielectric function of the TMC depending on the polariton frequency $\boldsymbol{\omega}$. It is evident from Eq. (15) that the surface polariton waves exist only if $\varepsilon(\boldsymbol{\omega}) < 0$.

Eq. (15) can be solved numerically to obtain the frequencies of the surface phonon-polaritons as functions of the composition x by using Eqs. (10) and (11) for the dielectric function $\varepsilon(\boldsymbol{\omega})$ in the TMC. The numerical results for the energies of the surface phonon-polaritons as functions of the composition x in the TMCs $Al_xGa_{1-x}As$, GaP_xAs_{1-x}, ZnS_xSe_{1-x}, $GaAs_xSb_{1-x}$, $Ga_xIn_{1-x}P$, and $Zn_xCd_{1-x}S$ for $k_{||}$=2 are obtained and plotted in Fig.2.

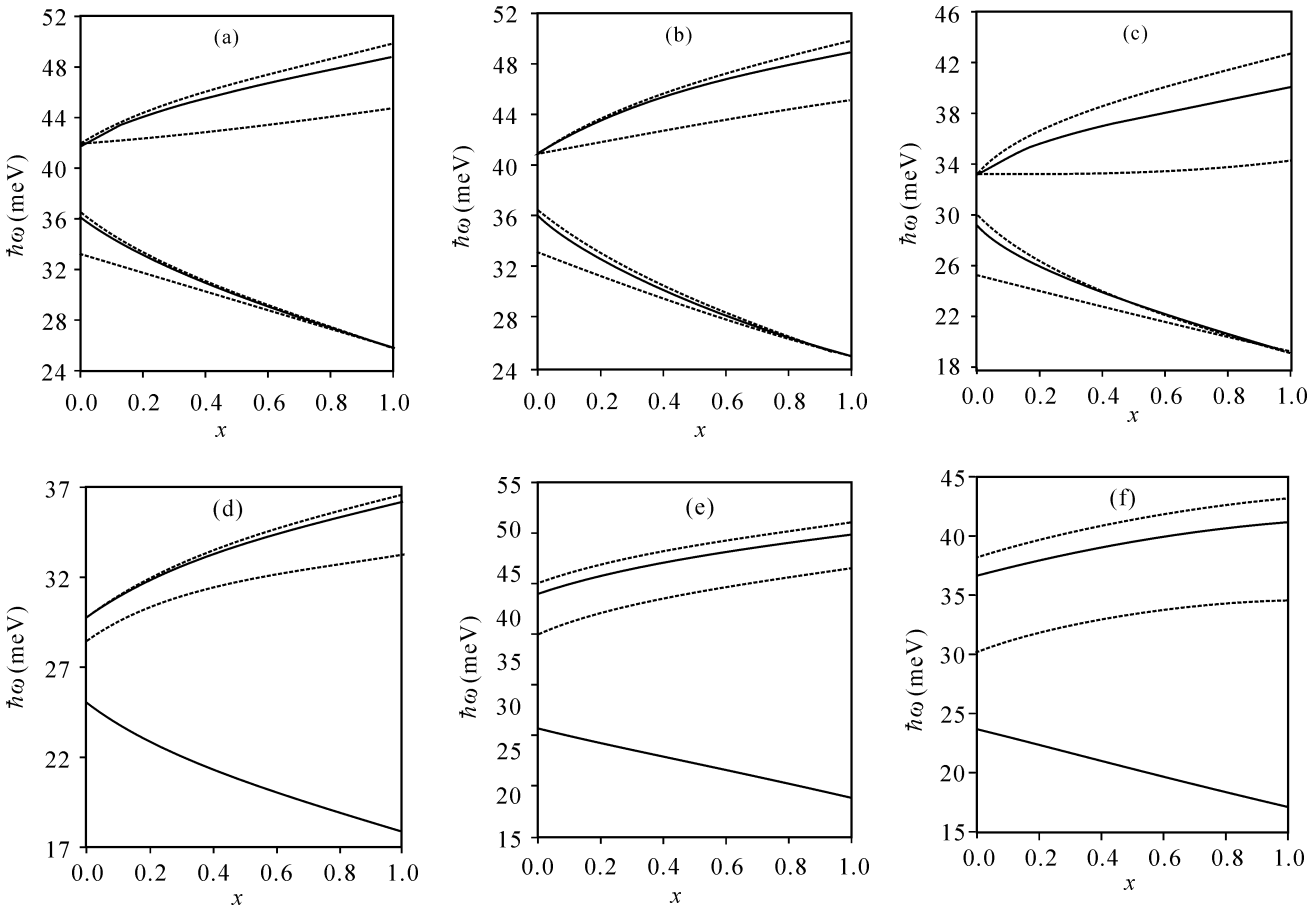

Fig.2 Energies of the surface phonon-polaritons (solid) as functions of the compositions *x*. (a) $Al_xGa_{1-x}As$, (b)GaP_xAs_{1-x}, (c) ZnS_xSe_{1-x}, (d) $GaAs_xSb_{1-x}$, (e) $Ga_xIn_{1-x}P$, and (f) $Zn_xCd_{1-x}S$ for $k_{||}$=2. The dashed lines are the energies of the long-wavelength TO and LO phonons of bulk optical phonons. The wave-vector $k_{||}$ is measured in ω_{TGaAs}/c, ω_{TGaAs}/c, ω_{TZnSe}/c, ω_{TGaSb}/c, ω_{TInP}/c, and ω_{TCdS}/c, respectively. ω_{TGaAs}, ω_{TZnSe}, ω_{TGaSb}, ω_{TInP}, and ω_{TCdS} are respectively the frequencies of TO phonons of the binary crystals of GaAs, ZnSe, GaSb, InP, and CdS.

It is found that there are two frequency branches of surface phonon-polariton modes lying respectively in the two forbidden bands of bulk phonon-polariton modes between the LO and TO frequencies of TMCs. The mode of surface phonon-polaritons with higher frequencies lies in the upper forbidden bands and we call it the "upper-branch", and another called the "lower-branch" is in the lower forbidden bands. The one- and two- mode behaviors of surface phonon-polaritons can be clearly seen in Fig.2. For the one mode behavior TMCs $GaAs_xSb_{1-x}$, $Ga_xIn_{1-x}P$, and $Zn_xCd_{1-x}S$, the two branches of "lower-frequency" bulk phonon-polariton energies are very close to each other, so that the related forbidden bands are rather narrow and near to vanish. Therefore it will be difficult for the "lower-branch" surface phonon-polariton modes to be observed. Otherwise the two-mode behavior is clearly seen for the systems $Al_xGa_{1-x}As$, GaP_xAs_{1-x} and ZnS_xSe_{1-x}, because the lower forbidden bands of bulk phonon-polariton modes is wider and the related surface modes are also observable.

It is seen that all the energies of the upper-branches of surface phonon-polariton modes increase with the increase of the Al-, P-, S-, As-, Ga-, Zn-compositions of the TMCs, but those of the lower-branches decrease with the increase of the components of the TMC systems. The dependences of the energies of surface polaritons on the compositon are all nonlinear.

In summary we have theoretically investigated the bulk and surface phonon-polaritons of ternary mixed crystals in the framework of the modified random-element-isodisplacement model and the Born-Huang approximation. The energies of the polaritons as functions of the compositions of TMC systems have been calculated. The numerical results for TMCs $Al_xGa_{1-x}As$, GaP_xAs_{1-x}, ZnS_xSe_{1-x}, $GaAs_xSb_{1-x}$, $Ga_xIn_{1-x}P$, and $Zn_xCd_{1-x}S$ show that the dispersion characteristic of the bulk and surface phonon-polaritons in TMC systems is sensitive to the compositions of TMCs. The dependences of the energies of bulk and surface phonon-polaritons on the compositions of TMCs are nonlinear.

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