Vol 11 No 2, February 2002
1009-1963/2002/11(02)/0144-06

Chinese Physics

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Influences of nonlinear interactions of polaritons in uniaxial crystals

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(Received 20 July 2001; revised manuscript received 13 September 2001)

We have analysed theoretically the polarization and dielectric constant of uniaxial ionic crystals, including the effect of the nonlinear interactions between ions. The spectrum of polaritons (coupling modes of photons and optical phonons) under nonlinear interactions was developed. A new branch of dispersion relations emerges in the original frequency gap.

Keywords: uniaxial crystal, polariton, self-energy correction
PACC: 7136

1. Introduction

In ionic crystals, the optical mode corresponds to the lattice vibration in which positive and negative ions of a primitive cell move relative to each other. It interacts strongly with electromagnetic waves, so it is important in the study of the electric and optical properties of ionic crystals. As the wavelength is very long, the relative displacements of positive and negative ions of different primitive cells change slowly in the crystal. Long wavelength phonons and photons couple into mixed vibration modes, namely polaritons. In ionic crystals, the attractive Coulomb forces between the positive and the negative ions and the repulsive Coulomb forces and the exchange forces between electrons of the outer shells are nonlinear. When the effect of the nonlinear interactions between ions cannot be ignored, it is necessary to discuss their effects on the polaritons. We have recently discussed[1] nonlinear effects on the polaritons of isotropic crystals, and have given interesting results. The dispersion relations of polaritons are very complicated in uniaxial crystals. So exploring the influences of nonlinear components of the interactions on their dielectric constants and polaritons is valuable.

2. Nonlinear dielectric constant of uniaxial ionic crystals in long-wavelength approximation

We suppose there are only two oppositely charged ions in a primitive cell. In the long-wavelength limit (the wavelength of the optical mode of the lattice vibration λ is much greater than the lattice constant a: $\lambda \approx 10^{-6}\mathrm{m} \gg a \approx 10^{-9}\mathrm{m}$), the relative displacement of positive and negative ions $(\boldsymbol{\mu}_+ - \boldsymbol{\mu}_-)$ changes slowly in different primitive cells. Hence we can introduce a vector $\boldsymbol{W}$ to describe the optical mode of lattice vibration[2]

$$\boldsymbol{W} = \rho^{1/2}(\boldsymbol{\mu}_+ - \boldsymbol{\mu}_-). \tag{1}$$

Here ρ is the reduced mass density

$$\rho = \frac{M}{\Omega}, \quad M = \frac{M_+ M_-}{M_+ + M_-}, \tag{2}$$

$M_+(M_-)$ is the mass of a positive (negative) ion, M is the reduced mass, and Ω is the volume of the primitive cell. Due to the anisotropy of the uniaxial crystal, there are two types of optical modes. One type is the ordinary mode (denoted by "o") in which the displacements of ions are perpendicular to the optical axis. Its vibration frequency and dielectric constant are ω_{ot} and ε_{o} respectively. The other type is the extraordinary mode (denoted by "e") in which the displacements of ions are in the plane where the optical axis and the wave vector lie. Its resonance frequency and dielectric constant are ω_{et} and ε_{e}, respectively, when the displacements of ions are parallel to the optical axis. If we neglect the coupling between the o mode and e mode, we can write two couples of Huang's equations.

For a centrosymmetric uniaxial crystal (i.e. possessing a centre of inversion), we can generalize Huang's equations into

$$(\ddot{\boldsymbol{W}})_{\mathrm{o,e}} = (b_{11}\boldsymbol{W} + b_{12}\boldsymbol{E} + g\boldsymbol{W}^3)_{\mathrm{o,e}}, \tag{3}$$

$$(\boldsymbol{P})_{\mathrm{o,e}} = (b_{12}\boldsymbol{W} + b_{22}\boldsymbol{E})_{\mathrm{o,e}}. \tag{4}$$

If we look for the monochromatic wave solution, from Eq.(3), we have $-\omega^2\boldsymbol{W} = b_{11}\boldsymbol{W} + b_{12}\boldsymbol{E} + g\boldsymbol{W}^3$, where we have omitted the subscripts for simplicity. Introducing the Green function

$$G = \frac{1}{-\omega^2 - b_{11}}, \tag{5}$$

we have $W = b_{12}GE + gGW^3$. Taking the second term as a perturbation, we expand $\boldsymbol{W}$ by iteration and thus have

$$\begin{aligned}\boldsymbol{W} =& b_{12}G\boldsymbol{E} + gb_{12}^3G^4\boldsymbol{E}^3\\ =& b_{12}G\boldsymbol{E} + gG^4b_{12}^3E_0^2\frac{1+\cos 2(\omega t - \boldsymbol{k}\cdot\boldsymbol{r})}{2}\boldsymbol{E}.\end{aligned}$$

If we discuss only the self-energy correction, we can keep only the basic frequency terms and obtain

$$\begin{aligned}\boldsymbol{W} =& b_{12}G\boldsymbol{E} + \frac{3}{4}gG^4b_{12}^3E_0^2\boldsymbol{E}\\ =& \left(b_{12}G + \frac{3}{4}gG^4b_{12}^3E_0^2\right)\boldsymbol{E}.\end{aligned} \tag{6}$$

We suppose the optical axis of the uniaxial crystal is the z-axis in the Cartesian coordinate system. Substituting Eq.(6) into Eq.(4), we have

$$P_{x,y} = \left[b_{12} + b_{22}\left(b_{12}G + \frac{3}{4}gG^4b_{12}^3E_0^2\right)\right]_{\mathrm{o}}E_{x,y}, \tag{7}$$

$$P_{\mathrm{z}} = \left[b_{12} + b_{22}\left(b_{12}G + \frac{3}{4}gG^4b_{12}^3E_0^2\right)\right]_{\mathrm{e}}E_{\mathrm{z}}. \tag{8}$$

From Eqs.(7) and (8), we can obtain the dielectric constants for the o and e modes by considering the nonlinear effect

$$\varepsilon_{\mathrm{o}} = \left\{\left[b_{12} + b_{22}\left(b_{12}G + \frac{3}{4}gG^4b_{12}^3E_0^2\right)\right]_{\mathrm{o}} - 1\right\}\varepsilon_0, \tag{9}$$

$$\varepsilon_{\mathrm{e}} = \left\{\left[b_{12} + b_{22}\left(b_{12}G + \frac{3}{4}gG^4b_{12}^3E_0^2\right)\right]_{\mathrm{e}} - 1\right\}\varepsilon_0. \tag{10}$$

With these dielectric constants, we can discuss the nonlinear optical properties of the uniaxial ionic crystal. We can see that the two dielectric functions are similar, except that the values of the parameters $(b_{12})_{\mathrm{o}}$, $(b_{22})_{\mathrm{o}}$ and $(b_{12})_{\mathrm{e}}$, $(b_{22})_{\mathrm{e}}$ are different.

3. Influences of nonlinear interactions of polaritons

Now we consider the coupling of the phonons and electromagnetic photons. When the ion crystal is illuminated by light, a transverse electromagnetic field is stimulated, and transverse oscillation of long-wavelength optical phonons will be influenced. Particularly, when the photon frequency $\omega = kc$ and the transverse phonon frequency $\omega_{\mathrm{t}}(\sim 10^{13}\mathrm{s}^{-1})$ are close to each other, the coupling will be very strong, the spectra of photons and phonons will change drastically, and polaritons will be generated.

For a plane wave, we suppose ($\boldsymbol{E}$, $\boldsymbol{D}$, $\boldsymbol{H}$, $\boldsymbol{P}$, $\boldsymbol{W}$)=($\boldsymbol{E}_0$, $\boldsymbol{D}_0$, $\boldsymbol{H}_0$, $\boldsymbol{P}_0$, $\boldsymbol{W}_0$)cos($\omega t - \boldsymbol{k}\cdot\boldsymbol{r}$). From Maxwell equations,[3] we have

$$\boldsymbol{D} = \varepsilon_0\boldsymbol{E} + \boldsymbol{P} = \varepsilon_0\varepsilon[\boldsymbol{E} - \hat{\boldsymbol{k}}(\hat{\boldsymbol{k}}\cdot\boldsymbol{E})]. \tag{11}$$

We select the z-axis as the optical axis. In uniaxial crystals, there are two independent linear-polarized light waves, which are ordinary and extraordinary light waves respectively. Without losing generality, we suppose $\boldsymbol{k}$ is in the yz plane, and its angle with the z-axis is θ, therefore k_x=0, k_y=sinθ and k_z=cosθ. From Eq.(11), we have

$$\begin{cases} D_x = \varepsilon_0\varepsilon E_x,\\ D_y = \varepsilon_0\varepsilon(E_y - \hat{k}_y(E_y\hat{k}_y + E_z\hat{k}_z),\\ D_z = \varepsilon_0\varepsilon(E_z - \hat{k}_z(E_y\hat{k}_y + E_z\hat{k}_z).\end{cases} \tag{12}$$

As we have adopted the principal axial coordinate system, for uniaxial crystals, we have $D_{x,y} = \varepsilon_0\varepsilon_{\mathrm{o}}E_{x,y}$, $D_z = \varepsilon_0\varepsilon_{\mathrm{e}}E_z$. Compared to Eq.(12), we obtain

$$(\varepsilon - \varepsilon_{\mathrm{o}})E_x = 0, \tag{13}$$

$$\begin{cases} E_y(\varepsilon\cos^2\theta - \varepsilon_{\mathrm{o}}) - E_z\varepsilon\sin\theta\cos\theta = 0,\\ -E_y\varepsilon\sin\theta\cos\theta + E_z(\varepsilon\sin^2\theta - \varepsilon_{\mathrm{e}}) = 0.\end{cases} \tag{14}$$

For the ordinary light there is $\boldsymbol{E} = E_x\boldsymbol{i}$ and, from Eq.(13), we have

$$\varepsilon = \varepsilon_{\mathrm{o}}. \tag{15}$$

We can see that the dielectric constant of the ordinary light is independent of the direction of the light.

For the extraordinary light there is $\boldsymbol{E} = E_y\boldsymbol{j} + E_z\boldsymbol{k}$. From Eq.(14), we obtain

$$\varepsilon_\theta = \frac{\varepsilon_{\mathrm{o}}\varepsilon_{\mathrm{e}}}{\varepsilon_{\mathrm{o}}\sin^2\theta + \varepsilon_{\mathrm{e}}\cos^2\theta}. \tag{16}$$

We can see that when $\theta = 0°$, we have $\varepsilon_\theta = \varepsilon_{\mathrm{o}}$ and when θ=90°, we have $\varepsilon_\theta = \varepsilon_{\mathrm{e}}$. So the dielectric constant of the extraordinary light varies between ε_{o} and ε_{e}.

With Eqs.(15), (16) and $\omega = kc/\sqrt{\varepsilon}$, we obtain the dispersion relations of o and e light respectively. For the ordinary light, we have

$$k^2 = \frac{\omega^2}{c^2}\left[\varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{\omega_{to}^2 - \omega^2}\omega_{to}^2 + 3\frac{b_{12}^4 g E_0^2}{(b_{11} + \omega^2)^4 \varepsilon_0}\right]_o . \quad (17)$$

Here c is the velocity of light in vacuum. In Fig.1(a), the k dependence of ω shows that a new branch of coupling modes emerges between $(\omega_{to})_o$ and $(\omega_{lo})_o$. Here $(\omega_{lo})_o$ is the resonance frequency of the longitudinal ordinary optical mode.

For the extraordinary mode, we have

$$k^2 = \frac{\omega^2}{c^2}\frac{\varepsilon_o \varepsilon_e}{\varepsilon_o \sin^2\theta + \varepsilon_e \cos^2\theta}. \quad (18)$$

The k dependence of ω is shown in Fig.1(b) (θ=60°) and Fig.1(c) (θ=90°). When θ=0°, the curve is the same as Fig.1(a). Figures 1(a), 1(b) and 1(c) show e modes when θ equals 0°, 60° or 90°, and the o modes are the same as the e modes when θ=0°. When θ is not 0° or 90°, we can see that the curve of the e mode is quite different to that of the o mode. Figures 2(a), 2(b) and 2(c) show the relations between the dielectric constant and frequency, drawn according to Eqs.(15) and (16). We can see a part of $\varepsilon > 0$ between $(\omega_{to} - \omega_{lo})_{o,e}$ corresponding to the polariton modes in Fig.1.

Figure 3(a), 3(b), and 3(c) show the reflectance dependence of $\omega(R - \omega)$ when the incident electromagnetic wave is perpendicular to the crystal surface, and

$$R = \left|\frac{\sqrt{\varepsilon} - 1}{\sqrt{\varepsilon} + 1}\right|^2. \quad (19)$$

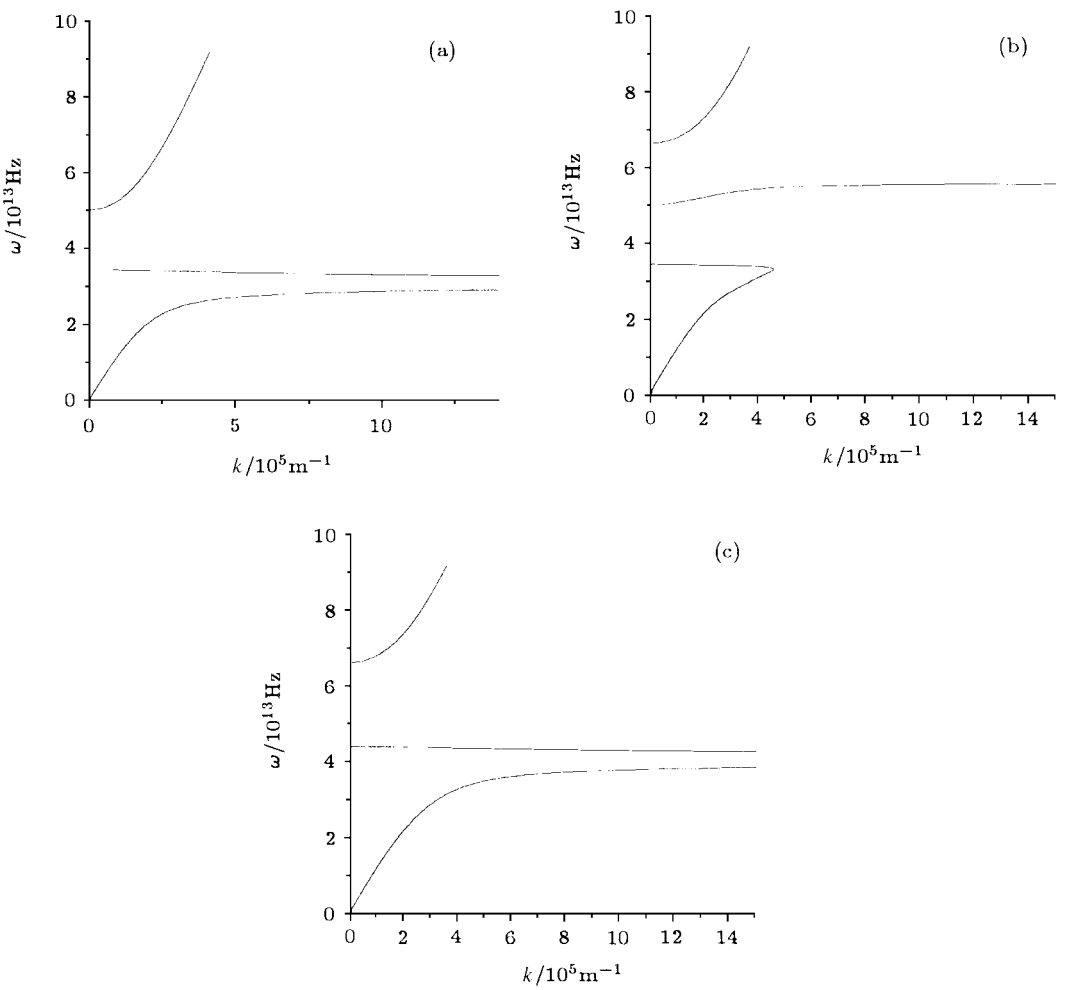


Fig.1. Dispersion relations.

It is shown that we have $R < 1$ (so the transmittance $T > 0$) in some area between $(\omega_{\rm to} - \varepsilon_{\rm lo})_{\rm o,e}$. There is a transmission peak corresponding to the new polariton modes in the original frequency gap in the linear case. The peak will vary with θ in the extraordinary modes.

Figures 4(a), 4(b), and 4(c) show the relations of reflectance and frequency $(R - \omega)$ when the nonlinear coefficient is negative. It is shown that the transmission peak is shifted to a frequency lower than $\omega_{\rm to}$ and the frequency gap is expanded for the ordinary mode. There is another peak in the gap for the extraordinary mode.

The quadratic term of nonlinear interactions of noncentrosymmetric crystals is not zero, thus for these materials we can replace Eq.(3) with $\ddot{\boldsymbol{W}} = b_{11}\boldsymbol{W} + b_{12}\boldsymbol{E} + f\boldsymbol{W}^2$, f being the nonlinear coefficient. We make a Fourier transformation and iterate twice, and obtain the self-energy correction

$$\boldsymbol{W} = \left(b_{12} G \boldsymbol{E} + \frac{3}{2} f^2 b_{12}^3 G^4 E_0^2 \boldsymbol{E} \right)_{\rm o,e}, \tag{20}$$

and dispersion relations for o and e light

$$k_{\rm o}^2 = \left\{ \frac{\omega^2}{c^2} \left[\varepsilon_\infty + \frac{\varepsilon_{\rm s} - \varepsilon_\infty}{\omega_{\rm to}^2 - \omega^2} \omega_{\rm to}^2 + \frac{3 f^2 b_{12}^4 E_0^2}{2(\omega^2 - \omega_{\rm to}^2)^4 \omega_{\rm to}^2 \varepsilon_0} \right] \right\}_{\rm o}, \tag{21}$$

$$k_{\rm e}^2 = \frac{\omega^2}{c^2} \frac{\varepsilon_{\rm o}\varepsilon_{\rm e}}{\varepsilon_{\rm o} \sin^2\theta + \varepsilon_{\rm e} \cos^2\theta}. \tag{22}$$

With diagrams of $k - \omega$, we can also find that new coupling modes will appear in the original frequency gap.

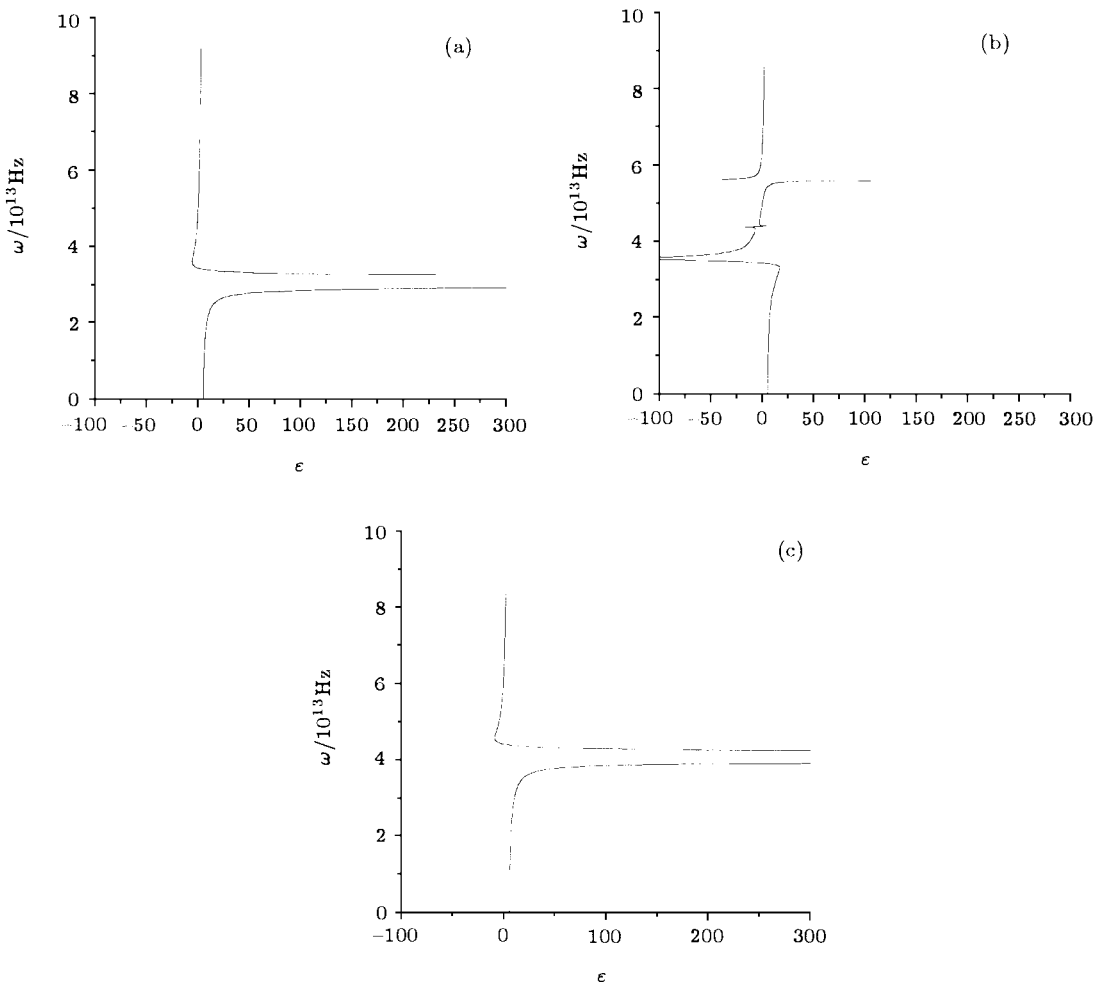


Fig.2. $\varepsilon \sim \omega$ relations.

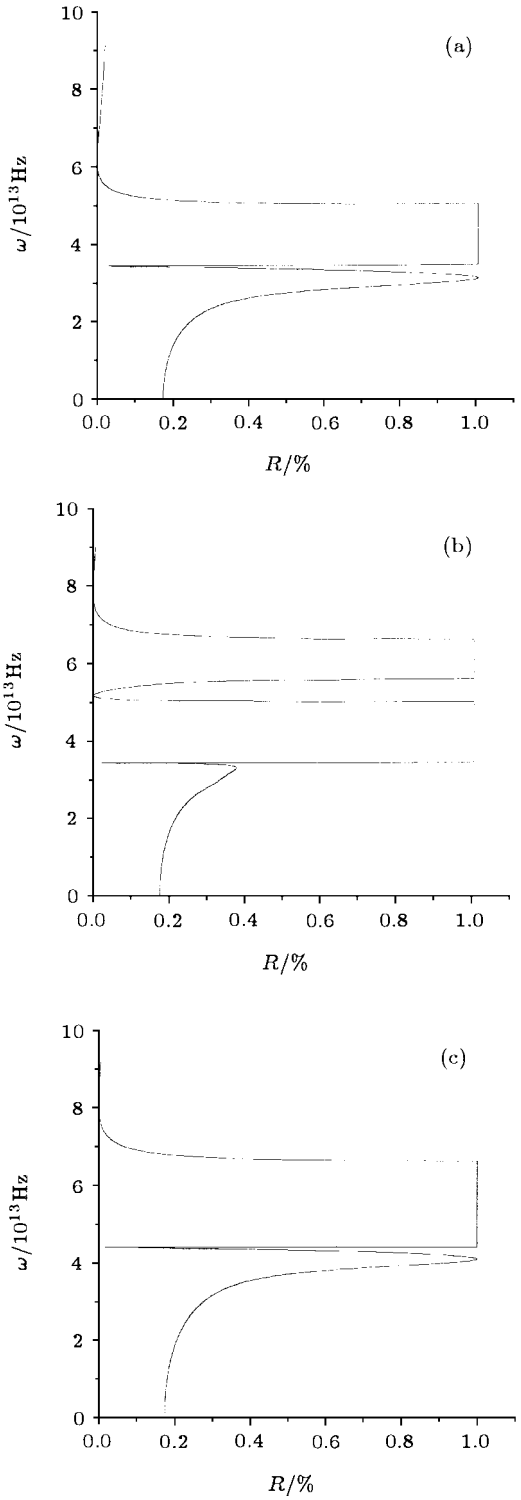


Fig.3. $R \sim \omega$ relations.

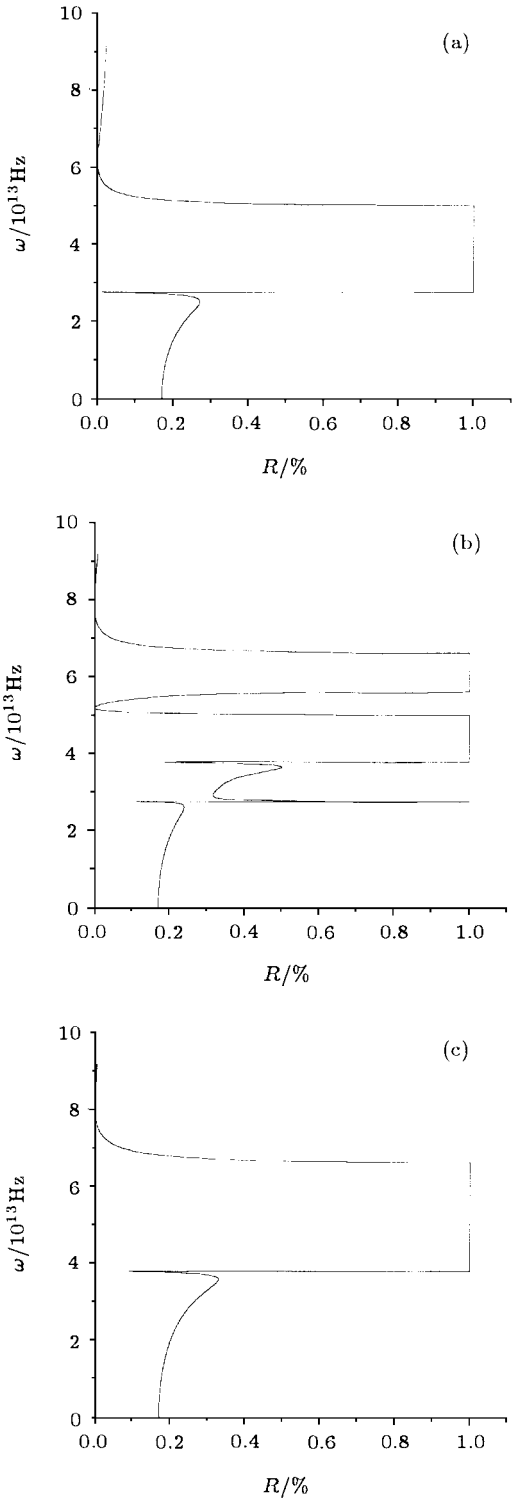


Fig.4. $R \sim \omega$ relations.

4. Discussions and conclusion

We have added the nonlinear component to Huangkun's equations when discussing the coupling of electromagnetic waves and long-wavelength optical waves of lattice vibration in uniaxial polar crystals. Using iteration with Green's function (equivalent to perturbation expansion), we obtained the self-energy correction to the dielectric constant. By combining Huangkun's equations with Maxwell equations of the

electromagnetic field, we have investigated the influences of nonlinear components of interactions on the spectrum of polaritons. A new branch of polariton modes emerges in the original forbidden gap. The new branch of o mode polaritons is independent of θ and that of e mode polaritons is dependent on θ. The electromagnetic waves corresponding to this branch of polariton modes can transmit through the crystal. This is a valuable result, and experiments to test this are suggested.

It should be pointed out that the nonlinear components of interactions between ions can result in corrections in the dielectric constant, which can not only make corrections to the self-energy compared to linear ones, but can also generate other nonlinear optical effects such as second harmonic generation, parametric amplification, etc.

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