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INFLUENCES OF NONLINEAR INTERACTIONS ON POLARITONS

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In this paper, we have analysed theoretically the polarization and dielectric constant generated by the nonlinear interactions between ions in ionic crystals. The spectrum of polaritons (coupling modes of photons and optical phonons) under nonlinear interactions has been developed. A new branch of dispersion relations has emerged in the original frequency gap between $\omega_{\rm TO}$ and $\omega_{\rm LO}$.

Keywords: polariton, self-energy correction
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I. INTRODUCTION

In ionic crystals, the optical mode corresponds to a wave in which positive and negative ions move relative to each other. It interacts strongly with electromagnetic waves, so it is important for the study of the electric and optical properties of ionic crystals. Since the wavelength is very long, the relative displacements of positive and negative ions are nearly the same in different primitive cells. Long wavelength phonons and photons couple into mixed vibration modes, namely polaritons. Huang discussed polaritons early in 1951, and has given a correct theoretical description of polaritons in diatomic cubic ionic crystals.[1,2] The theory is based upon linear interactions between ions. However, the fact is that there are nonlinear components of interactions, and with the advancement of laser science and technology, the electromagnetic waves can be very intense; they can couple with the nonlinear components of interactions between ions and affect the polaritons. So exploring the influences of the nonlinear components of the interactions on the dielectric constant and polaritons may be valuable.

II. NONLINEAR DIELECTRIC CONSTANT OF IONIC CRYSTALS IN LONG-WAVELENGTH APPROXIMATION

We suppose thst there are only two oppositely charged ions in a primitive cell as, in the long wavelength limit, the relative displacements of positive and negative ions ($\boldsymbol{\mu}_+ - \boldsymbol{\mu}_-$) are nearly the same. So we can introduce a vector $\boldsymbol{W}$ to describe the optical wave:[3]

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

where ρ is the reduced mass density,

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

and $M_\pm$ is the mass of positive or negative ions, M is the reduced mass, and Ω is the volume of the primitive cell. For a centrosymmetric crystal, we expand the nonlinear component of the interactions between ions to the third order. We can generalize Huang's equations into

$$\ddot{\boldsymbol{W}} = b_{11}\boldsymbol{W} + b_{12}\boldsymbol{E} + g\boldsymbol{W}^3, \tag{3}$$

$$\boldsymbol{P} = b_{12}\boldsymbol{W} + b_{22}\boldsymbol{E}. \tag{4}$$

(a) In a static electric field, the relative displacements of the positive and negative ions have $\ddot{\boldsymbol{W}}$=0. Then we obtain

$$g\boldsymbol{W}^3 + b_{11}\boldsymbol{W} + b_{12}\boldsymbol{E} = 0, \tag{5}$$

$$\boldsymbol{W} \doteq -\frac{b_{12}}{b_{11}}\boldsymbol{E} + \frac{g b_{12}^3}{b_{11}^3}E^2\boldsymbol{E}. \tag{6}$$

Substituting Eq.(6) into Eq.(4), we have $\boldsymbol{P} \doteq \left(b_{22} - \frac{b_{12}^2}{b_{11}}\right)\boldsymbol{E} + \frac{g b_{12}^4}{b_{11}^3}E^2\boldsymbol{E}$. Then with $\boldsymbol{D} = \varepsilon_0\boldsymbol{E} + \boldsymbol{P} = \varepsilon_{\rm s}\varepsilon_0\boldsymbol{E}$, we obtain the dielectric constant $\varepsilon_{\rm s}$ including the nonlinear effect

$$\begin{aligned}\varepsilon_{\rm s} &= \left(b_{22} - \frac{b_{12}^2}{b_{11}} + g\frac{b_{12}^4}{b_{11}^3}E^2\right)/\varepsilon_0 + 1 \\ &= \varepsilon_{\rm s}^1 + g\frac{b_{12}^4}{\varepsilon_0 b_{11}^3}E^2,\end{aligned} \tag{7}$$

in which $\varepsilon_{\rm s}^1$ is the linear relative static dielectric constant. Equation (7) may generate self-focusing and Kerr effects and other nonlinear physical phenomena.

(b) We suppose an external electric field $\boldsymbol{E} = \sum_{\omega} \boldsymbol{E}(\omega)\mathrm{e}^{-\mathrm{i}\omega t}$ is directed into the crystal, and let $\boldsymbol{W} = \sum_{\omega} \boldsymbol{W}(\omega)\mathrm{e}^{-\mathrm{i}\omega t}$. Using Fourier transformation for Eq.(3), we have

$$\begin{aligned}&(-\omega^2 - b_{11})\boldsymbol{W}(\omega)\\ =&b_{12}\boldsymbol{E}(\omega) + g\sum_{\omega',\omega''} W(\omega')W(\omega'')\boldsymbol{W}(\omega-\omega'-\omega'').\end{aligned} \tag{8}$$

Let

$$G(\omega) = \frac{1}{-\omega^2 - b_{11}}, \tag{9}$$

$$\begin{aligned}\boldsymbol{W}(\omega) =&b_{12}G(\omega)\boldsymbol{E}(\omega) + gG(\omega)\\ &\cdot \sum_{\omega',\omega''} W(\omega')W(\omega'')\boldsymbol{W}(\omega-\omega'-\omega'').\end{aligned} \tag{10}$$

Iterating $\boldsymbol{W}(\omega)$, we have

$$\begin{aligned}\boldsymbol{W}(\omega) =&b_{12}G(\omega)\boldsymbol{E}(\omega) + \sum_{\omega',\omega''} gb_{12}^3 G(\omega)G(\omega')E(\omega')\\ &\cdot G(\omega'')E(\omega'')G(\omega-\omega'-\omega'')\boldsymbol{E}(\omega-\omega'-\omega'').\end{aligned} \tag{11}$$

With $\omega' = \pm\omega, \omega'' = \mp\omega$, or $\omega' = \omega'' = \omega$, electric intensities in the second term of Eq.(11) are not zero, so we obtain the self-energy correction

$$\begin{aligned}\boldsymbol{W}(\omega) =&b_{12}G(\omega)\boldsymbol{E}(\omega)\\ &+ 3gb_{12}^3 G^3(\omega)G(-\omega)E^2(\omega)\boldsymbol{E}(-\omega).\end{aligned} \tag{12}$$

With $\boldsymbol{P} = [\varepsilon(\omega) - 1]\varepsilon_0\boldsymbol{E}$, we can obtain

$$\begin{aligned}\varepsilon^{\mathrm{n}}(\omega) =&[b_{12}^2 G(\omega) + b_{22}\\ &+ 3gb_{12}^3 G^3(\omega)G(-\omega)E^2(\omega)]/\varepsilon_0 + 1.\end{aligned} \tag{13}$$

The preceding discussions have not considered the damping effect. When there is damping, we should replace Eq.(3) with

$$\ddot{\boldsymbol{W}} = b_{11}\boldsymbol{W} + b_{12}\boldsymbol{E} + g\boldsymbol{W}^3 - r\dot{\boldsymbol{W}}, \tag{14}$$

where $r > 0$ is the damping factor. Letting $G(\omega) = \frac{1}{-\omega^2 - b_{11} - \mathrm{i}r\omega}$, we iterate W once, and have

$$\begin{aligned}\boldsymbol{W}(\omega) =&b_{12}G(\omega)\boldsymbol{E}(\omega)\\ &+ 3gb_{12}^3 G^3(\omega)G(-\omega)E(\omega)^2\boldsymbol{E}(-\omega).\end{aligned} \tag{15}$$

So the nonlinear dielectric constant with damping is

$$\begin{aligned}\varepsilon^{\mathrm{n}}(\omega) =&[b_{12}^2 G(\omega) + b_{22} + 3gb_{12}^4 G^3(\omega)\\ &\cdot G(-\omega)E(\omega)E(-\omega)]/\varepsilon_0 + 1.\end{aligned} \tag{16}$$

With this nonlinear dielectric constant, we can discuss the nonlinear optical properties of the ionic crystal.

III. INFLUENCES OF NONLINEAR INTERACTIONS ON POLARITONS

Now we consider the coupling of phonons and electromagnetic photons. Since photons are quanta of transverse electromagnetic field, when an ionic crystal is illuminated by a light, the transverse electromagnetic field is stimulated, and the transverse oscillation of the long-wavelength optical phonons will be influenced. Particularly, when the photon frequency $\omega = kc$ and transverse phonon frequency $\omega_{\mathrm{T}}(\approx 10^{13}\mathrm{s}^{-1})$ are close to each other, the coupling will be very strong. Then the spectra of photons and phonons will change drastically, and as a result polaritons will be generated. We must find the solutions of the Maxwell equations and Huang's equations:[3,4]

$$\begin{aligned}&\nabla\times\boldsymbol{E}_{\mathrm{T}} = -\mu_0\dot{\boldsymbol{H}},\\ &\nabla\times\boldsymbol{H} = -(\varepsilon_0\dot{\boldsymbol{E}}_{\mathrm{T}} + \dot{\boldsymbol{P}}_{\mathrm{T}}),\\ &\ddot{\boldsymbol{W}}_{\mathrm{T}} = b_{11}\boldsymbol{W}_{\mathrm{T}} + b_{12}\boldsymbol{E}_{\mathrm{T}} + g\boldsymbol{W}_{\mathrm{T}}^3,\\ &\boldsymbol{P}_{\mathrm{T}} = b_{12}\boldsymbol{W}_{\mathrm{T}} + b_{22}\boldsymbol{E}_{\mathrm{T}}.\end{aligned} \tag{17}$$

These equations only involve transverse field vectors.

We now consider plane wave solutions. Supposing the wave vector $\boldsymbol{k}$ is along the z direction, $\boldsymbol{E}$, $\boldsymbol{P}$ and $\boldsymbol{W}$ are along the x direction and H is along the y direction, then we have

$$\begin{aligned}W_x/W_x(\omega) = P_x/P_x(\omega) = E_x/E_x(\omega)\\ = H_y/H_y(\omega) = \exp[\mathrm{i}(kz - \omega t)].\end{aligned} \tag{18}$$

Substituting Eq.(18) into Eq.(17) yields

$$\begin{aligned}&kE_x(\omega) - \mu_0\omega H_y(\omega) = 0,\\ &kH_y(\omega) - \omega\varepsilon_0 E_x(\omega) - P_x(\omega) = 0,\\ &-\omega^2 W_x(\omega) - b_{11}W_x(\omega) - b_{12}E_x(\omega) - gW_x^3(\omega) = 0,\\ &P_x(\omega) - b_{11}W_x(\omega) - b_{22}E_x(\omega) = 0.\end{aligned} \tag{19}$$

From Eqs.(19) and (9) we obtain the dispersion relations:

$$\begin{aligned}k^2 =&\frac{\omega^2}{c^2}\left[\varepsilon_\infty + \frac{\varepsilon_{\mathrm{s}} - \varepsilon_\infty}{\omega_{\mathrm{TO}}^2 - \omega^2}\omega_{\mathrm{TO}}^2\right.\\ &\left.+ 3\frac{b_{12}^4 gE_0^2(\omega)}{(b_{11} + \omega^2)^4\varepsilon_0}\right],\end{aligned} \tag{20}$$

where $c = 1/\sqrt{\varepsilon_0\mu_0}$ is the velocity of light in vacuum. Figure 1 shows the dispersion relation $(\omega - k)$ curve, in which there are three different ω at a given k. With

the increase of the nonlinear factor or electric field intensity, there emerges a new branch of coupled mode between $\omega_{\rm TO}$ and $\omega_{\rm LO}$, because of the nonlinearity. With $\omega \to \omega_{\rm TO}$, $k \to \infty$ becomes slower compared with the linear case of the lower branch. Figure 2 shows the relation of the dielectric constant and frequency, which is drawn according to Eq.(13). We can see that a part of $\varepsilon^{\rm n}(\omega) > 0$ lies between $\omega_{\rm TO}$ and $\omega_{\rm LO}$, corresponding to the new polariton modes in Fig.1.

Figure 3 displays the relation of the reflectance and frequency R-ω when the electromagnetic wave becomes perpendicular to the crystal surface

$$R(\omega) = \left| \frac{\sqrt{\varepsilon(\omega)} - 1}{\sqrt{\varepsilon(\omega)} + 1} \right|^2 . \tag{21}$$

It is shown that R <1 in some area between $\omega_{\rm TO}$ and $\omega_{\rm LO}$ and then the transmittance T >0. So there is a transmission peak corresponding to the new polariton modes in the original frequency gap in the linear case.

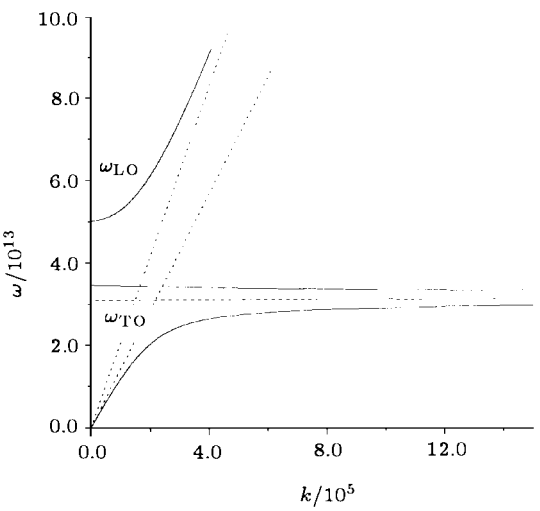


Fig.1. Dispersion relations.

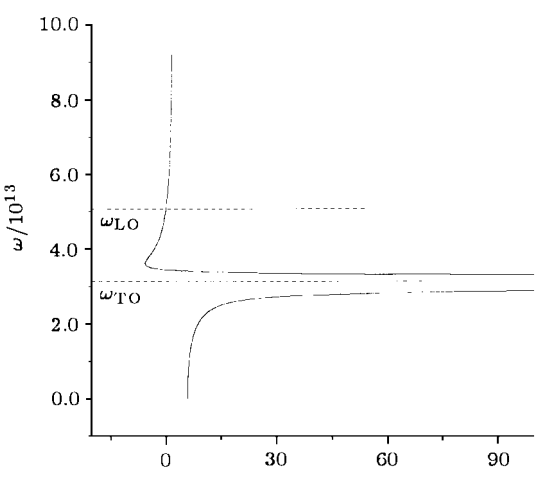


Fig.2. ε-ω relations.

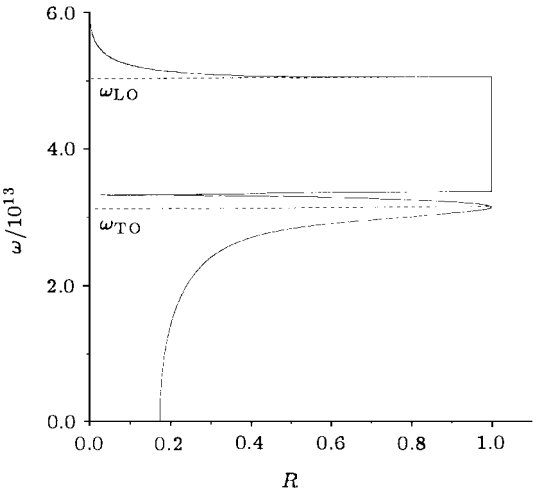


Fig.3. R-ω relations.

The quadratic term of nonlinear interactions of non-centrosymmetric crystals is not zero. Thus, for these materials, we can replace Eq.(3) with $\ddot{\boldsymbol{W}} = b_{11}\boldsymbol{W} + b_{12}\boldsymbol{E} + f\boldsymbol{W}^2$, where f is the nonlinear coefficient. Then we make the Fourier transformation and iterate twice, and obtain

$$\begin{aligned} \boldsymbol{W}(\omega) =& b_{12}G(\omega)\boldsymbol{E}(\omega) + 2\sum_{\omega',\omega''} f^2 b_{12}^3 \\ & \cdot G(\omega)G(\omega')G(\omega'')G(\omega'-\omega'')G(\omega-\omega') \\ & \cdot E(\omega'')E(\omega-\omega')\boldsymbol{E}(\omega'-\omega''). \end{aligned} \tag{22}$$

When the electric intensities in the second term of Eq.(21) are not zero, i.e. $\omega' = 0$, $\omega'' = \pm\omega$ we obtain the self-energy correction

$$\boldsymbol{W}(\omega) = b_{12}G(\omega)\boldsymbol{E}(\omega) + 4f^2 b_{12}^3 G^4(\omega)E^2(\omega)\boldsymbol{E}(-\omega) \tag{23}$$

and dispersion relations

$$\begin{aligned} k^2 =& \frac{\omega^2}{c^2}\left\{ \varepsilon_\infty + \frac{\varepsilon_{\rm s} - \varepsilon_\infty}{\omega_{\rm TO}^2 - \omega^2}\omega_{\rm TO}^2 \right. \\ & \left. + \frac{4f^2 b_{12}^4 E_0^2(\omega)}{(\omega^2 - \omega_{\rm TO}^2)^4 \omega_{\rm TO}^2 \varepsilon_0} \right\}. \end{aligned} \tag{24}$$

With diagrams for k-ω, we can also find that new coupling modes will appear in the original frequency gap.

IV. DISCUSSION AND CONCLUSION

We have added the nonlinear component into Huang's equations when discussing the coupling of electromagnetic waves and long-wavelength optical waves in polar crystals. With the iteration method (equivalent to the perturbation expansion), we found

the self-energy correction to the dielectric constant. By combining Huang's equations with the Maxwell equations of the electromagnetic field, we found the influences of nonlinear components of interactions on the spectrum of polaritons. A new branch of polariton mode will emerge in the original forbidden gap between ω_{TO} and ω_{LO}. The electromagnetic waves corresponding to this branch of oscillation can transmit through the crystal. This is a valuable result which could be useful, and experiments to test this are suggested.

It is necessary to point 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 generate other nonlinear optical effects such as second harmonic generating, parametric amplification etc.

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