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Theory of Nonlinear s-Polarized Phonon-Polaritons in Multilayered Structures

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Abstract

A theory is presented for the dispersion relations of the nonlinear phonon-polaritons arising when phonons are coupled to the electromagnetic waves in multilayered structures of nonlinear materials. The calculations are applied to a multilayered structure consisting of a thin film surrounded by semi-infinite bounding media where each layer may have a frequency dependent dielectric function and Kerr-type nonlinearity. At least one of the media is an ionic crystal supporting optical phonon modes. The resulting analytic and numerical solutions for the dispersion relations of phonon-polaritons with s-polarization are considered for several cases. We find that the presence of nonlinearities leads to multiple branches in the dispersion relation. The results are plotted as frequency versus wave vector and frequency versus nonlinearity for different phonon-polariton modes. The parameters that modify the modes correspond to the in-plane wave vector, the thickness of the film, the phonon frequencies and the nonlinearity of each layer.

Keywords: Nonlinear; Phonon-polaritons; Kerr type nonlinearity; Dispersion relations; Frequency; Media

1. Introduction

We present an analytic formulation for a specific type of collective excitations (optical phonons) that can propagate in a multilayer system composed of layer of different materials and are coupled to electromagnetic waves (EMW). It is known from studies of other geometries and materials that the characteristics of surface and interfaces give rise to localized and guided excitations [1-3]. The mixed modes constructed by coupling EMW and collective excitations, which are known generally as *polaritons*, have been studied over many years. For the case of coupling to the crystal vibrations that occurs in metals or semiconductors the resulting modes are called *phonon-polaritons*. These modes have both photon and optical phonon characteristics, depending on the wave vector and the properties of the multilayered system.

On the other hand, the properties of guided and/or surface EMW in optical structures that exhibit *nonlinear* effects of the *Kerr-type* have been the subject of both theoretical and experimental interest [4,5]. Now, with

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the inclusion of a characteristic frequency dependence (corresponding to crystal excitations) in the nonlinear dielectric function we may extend this work to nonlinear polaritons due to coupling of EMW to crystal excitations. In a recent work we studied the propagation of nonlinear plasmon-polariton modes [6] in a three-layered structure with planar interfaces, including the nonlinear dielectric constants, which was generally frequency dependent. In this present paper we consider the propagation of nonlinear phonon-polaritons, which presents another interesting case with distinctive properties. Specifically we investigate the TE (or s-polarized) surface phonon-polariton modes supported by a three-layered system (a thin film bounded symmetrically by semi-infinite bounding media) of the type often investigated experimentally (see, *e.g.*, Ref. 7). The dielectric functions of both materials are generally taken to be both frequency dependent and nonlinear (with a Kerr-type nonlinearity). The linear term in the dielectric function depends on the frequency in a way that represents an optical phonon in a metal or a doped semiconductor. The nonlinearity constant can, in general, be either positive or negative. For the positive case (or so-called self-focusing case) we consider the propagation of phonon-polariton modes in the general structure where either one or both of the constituent media are nonlinear.

This paper is outlined as follows. In section 2 we outline the theoretical procedure briefly, because the method is broadly similar to that of the plasmon-polariton case in Ref. 6. The important difference, however, is that in the phonon-polariton case there are additional frequency bands (compared with the plasmon-polariton case) in which the nonlinear polaritons may propagate. This is a consequence of the fact that, in a bulk linear medium, the phonon-polariton dispersion curve has two branches, whereas the plasmon-polariton curve consists of only one branch (see *e.g.* Ref. 6). This provides a motivation for extending our previous work on nonlinear plasmon-polaritons to the phonon case. In addition the frequency ranges can be different in the two cases, which may be important for practical applications. In section 3 we consider a special case corresponding to a nonlinear layer bounded by a medium with a linear frequency dependent dielectric function. We next present the general case in which both media are nonlinear and have frequency dependent dielectric functions in section 4. The calculations for the limiting case of a linear frequency dependent film bounded by nonlinear frequency dependent media are obtained as a special case when the nonlinearity of the film tends to zero. Numerical examples are given throughout the paper. Finally section 5 is devoted to conclusions, including the possibility of extending this method to other geometries and other type of dielectric media.

2. Theoretical Model

The EMW are assumed to be TE (or s-polarized) modes. We take the interfaces to be in the $x-y$ plane and choose the *x*-axis to represent the propagation direction of the coupled modes parallel to the interfaces. For phonons (in the absence of damping) we consider the dielectric function in layer *j* to have the form

$$\varepsilon_j(\omega)=\varepsilon_{0j}(\omega)+\alpha_j E^2 \tag{1}$$

where the nonlinearity coefficient α_j in each layer is assumed to be a positive constant and the linear term is given by the standard expression [1,9,10]

$$\varepsilon_{0j}(\omega)=\varepsilon_{\infty j}\left(1+\frac{\omega_{jL}^2-\omega_{jT}^2}{\omega_{jT}^2-\omega^2}\right) \tag{2}$$

The frequency ω_{jl} ($l=T,L$ where T stands for transverse optical phonons and L for longitudinal optical phonons) denotes the phonon frequency and index $j=1,2,3$ (with 2 labeling the film, while the bounding materials 1 and 3 are the same for a symmetric structure).

We now briefly outline the basic theory for nonlinear polaritons in a multilayered structure; details are to be found in Refs. 5 and 6. In a nonmagnetic medium, substituting the form of electric and magnetic fields for TE modes into Maxwell equations, we obtain a nonlinear wave equation for the electric field **E** $=(0,E_x,0)\exp[i(kx-\omega t)]$, where *k* is the in-plane wave vector and ω is the frequency. From this, we deduce the first integration of the differential equation for E_y in the form

$$\left(\frac{\partial E_j}{\partial z}\right)^2+\frac{\omega^2}{c^2}g_j\left(E_j^2\right)=C_j \tag{3}$$

where C_j is the constant of integration and the coordinate *z* (measured perpendicular to the layers) lies within the layer *j*. The function *g* is a quadratic function of the electric field amplitude, and it also contains the nonlinearity coefficient (see, *e.g.*, Ref. 6). It is given by $g=\left[\varepsilon_{oj}(\omega)-(ck/\omega)\right]E_j^2+\frac{1}{2}\alpha_j E_j^4$ The dispersion relations for the phonon-polariton modes can be obtained by rewriting Equation (3) as an expression for $\partial E / \partial z$ and then integrating it with respect to z. Following the procedure of Ref. 6 different cases may arise for the electric amplitude depending on the sign and magnitude of the constant C_2, leading to various possible types of Jacobian elliptic integrals [8]. The continuity of $\partial E / \partial z$ at the film boundaries necessitates a careful determination of the sign of the integral expression for $\partial E / \partial z$. Due to the multivalued nature of $\partial E / \partial z$ as a function of E it is convenient to sketch the phase trajectory graph for the mode under consideration before integrating Equation (3), as discussed in Ref. 6. For the case of nonlinear film (with $\alpha_2 > 0$) the phase trajectory is a closed loop and the solution $E(z)$ is a periodic function. Then the integration is performed segment by segment starting from the boundary of the film (layer 2). Since the integrand is an even function of amplitude E_j the integral is simplified.

3. Nonlinear Film Bounded by Linear Media

In this special case the three-layer structure consists of a nonlinear dielectric film of thickness d, chosen to occupy the region $|z| \le d/2$ and characterized by ε_2. The two linear semi-infinite media on each side are characterized by a frequency dependent dielectric function $\varepsilon_{01}(\omega)$ in the region $|z| > d/2$.

For the nonlinear film, using Equation (3) and introducing a dimensionless variable $\overline{E} = E / E_1$ where E_1 is the value of E at the interface $z = -\frac{1}{2}d$ (see Ref. 6), we get

$$\left(\frac{\lambda \partial \overline{E}}{\partial z}\right)^2 = (2\pi)^2 \left[\frac{c^2}{\omega^2} C_2 - g_2(|\overline{E}|^2)\right] \tag{4}$$

which for the linear (bounding) media we may write

$$\left(\frac{\lambda \partial \overline{E}}{\partial z}\right)^2 = (2\pi) \left[\frac{c^2 k^2}{\omega^2} - \varepsilon_{01}\right] \overline{E}^2 \tag{5}$$

Here $\lambda = 2\pi c / \omega$ represents the vacuum wavelength. A necessary condition $c^2 k^2 / \omega^2 > \varepsilon_{01}$ for decaying waves in the bounding layer as $|z| \to \infty$ is implied from Equation (6). For convenience we shall assume at this stage that $\varepsilon_{02} > \varepsilon_{01}$ and $\alpha_2 > 0$ in order to study the guided and surface waves in the film, depending on the value of ω. By analogy with Ref. 6 this is a sufficient condition for $C_2 > 0$, which simplifies the analysis. The phonon-polariton dispersion relation can then be obtained by integrating the expressions for $\partial E / \partial z$ from Equations. (4) and (5), taking care to deal correctly which the signs (as mentioned earlier) and applying the usual electromagnetic boundary conditions at $z = \pm \frac{1}{2} d$. The result leads to the following equation (by analogy with Ref. 6) for the phonon-polariton dispersion relation

$$b.\mathrm{cn}\left(\frac{qd}{2} - JK \middle| m\right) - 1 = 0 \tag{6}$$

where $K \equiv cn^{-1}(0|m)$ is one quarter at the period of the elliptic function $\mathrm{cn}(x|m)$ and the integer J takes the four values 0,1,2,3 for a complete description of the solutions. The argument m determines the shape, the period and the elliptic function of interest, the parameter q is a frequency dependent, effective wave number in the z direction, and b is an amplitude factor for the cn function. The quantities m, q and b depend on the parameters of the layered structure, and are defined by analogy with Refs. 5 and 6. We solve Equation (6) numerically to obtain the behavior of the nonlinear phonon-polariton modes in the geometry under consideration.

We express the above results in a convenient form for the numerical calculations by introducing a reduced frequency $\Omega = \omega / \omega_{1T}$ and a reduced wave vector $\kappa = ck / \omega_{1T}$. We may also define the ratio $\Omega_0 = \omega_{1L} / \omega_{1T} > 1$, which typically lies between 1 and 2 for alkali halides (see, *e.g.*, Ref. 8). It is convenient to employ a characteristic length parameter (define in terms of the bounding medium) given by $d_0 = 2\pi c / \omega_{1T}$, and we have made numerical calculations for values of the film thickness corresponding to $d = d_0$ and $d = \frac{1}{2} d_0$. For example, in the case of NaBr, we have $\omega_{1T} / 2\pi \approx 0.4 \times 10^{12}$ Hz and $\Omega_0 \approx 1.56$, implying that $d_0 = 75$ µm. For NaCl the corresponding values are $\omega_{1T} / 2\pi \approx 4.9 \times 10^{12}$ Hz, $\Omega_0 \approx 1.61$, and $d_0 \approx 60$ µm. The condition mentioned earlier for bounded solutions as $z \to \pm\infty$ is just $\kappa^2 / \Omega^2 > \varepsilon_{01}$, which implies that the physical solutions are restricted to the two regions of Ω characterized by $0 < \Omega < \Omega_{c1}$ and $1 < \Omega < \Omega_{c2}$ with

$$\Omega_{c1}(\kappa)=\sqrt{\frac{\kappa^2+\varepsilon_{1\infty}\Omega_0^2-\sqrt{\left(\kappa^2+\varepsilon_{1\infty}\Omega_0^2\right)^2-4\varepsilon_{1\infty}\kappa^2}}{2\varepsilon_{\infty 1}}} \quad (7a)$$

$$\Omega_{c2}(\kappa)=\sqrt{\frac{\kappa^2+\varepsilon_{1\infty}\Omega_0^2+\sqrt{\left(\kappa^2+\varepsilon_{1\infty}\Omega_0^2\right)^2-4\varepsilon_{1\infty}\kappa^2}}{2\varepsilon_{1\infty}}} \quad (7b)$$

Other conditions may add further constraint on the range of Ω , *e.g.* the assumption of $\varepsilon_2 > \varepsilon_1$ and $p_2 > 0$ which ensures that $C_2 > 0$. By re-expressing Equation (6) in terms of the dimensionless quantities defined above, we have studied the nonlinear phonon-polariton dispersion relations.

As a numerical example we first choose NaCl to be the bounding medium (with $\varepsilon_{1\infty} = 2.25$, $\omega_{1T}/2\pi = 4.9\times10^{12}$ Hz, $\omega_{1L}/2\pi = 7.9\times10^{12}$ Hz) and the film to be a medium whose dielectric function has negligible frequency dependence in this range of frequencies, *e.g.*, Si where we take $\varepsilon_{02}(0) = 11.7$. The physical solutions are then constrained to lie within the ranges shown shaded in Figure 1 which corresponds to satisfying the necessary conditions $\varepsilon_{02}(0) > \varepsilon_1(\omega)$ and $\kappa^2/\Omega^2 > \varepsilon_{01}(\omega)$. In Figure 2a we have plotted the reduced frequency Ω *versus* wave vector κ in taking parameters $d = \frac{1}{2}d_0$, $p_2 = 0.05$ and $J = 0,1,2,3$. Also, the solutions for Ω *versus* $\sqrt{p_2}$ with a fixed wave-vector value $\kappa = 0.5$ are demonstrated in Figure 2b. For each value of J a sequence of curves representing the solutions associated with phonon-polariton modes are obtained which are related to the periodicity of $\mathrm{cn}(x|m)$ function. In order to illustrate the effect of the film thickness on the nonlinear phonon-polaritons, we show in Figures 3a and 3b the corresponding calculations for Ω *versus* κ and Ω *versus* $\sqrt{p_2}$, respectively, with $d = d_0$ and other parameters as before. The ranges of Ω are the same as before, but there are now more branches to the spectra, corresponding to the larger value of *d*.

Next it is assumed that ε_{02} (the linear part of dielectric function of the film) may also be frequency dependent. In this more general case we express $\varepsilon_1(\omega)$ and $\varepsilon_{02}(\omega)$ in terms of the dimensionless quantities introduced earlier, and also define the ratios $s = \omega_{2L}/\omega_{1L}$ and $t = \omega_{2T}/\omega_{1T}$. Depending on the values of parameters *t* and *s* (in particular, whether they are greater or less than 1) different cases may arise. For example, in the case where GaSb is the film material and InAs is the bounding medium we have $t = 0.95$, $s = 0.98$, $\Omega_0 = 1.09$, $\varepsilon_{1\infty} = 12.3$ and $\varepsilon_{2\infty} = 14.4$. In general, the physical modes correspond to those bands where Ω lies within the intervals defined by

$$0 < \Omega < \min(\Omega_{c1}, \Omega_{d1}) \text{ and}$$

$$\max(t,1) < \Omega < \min(\Omega_{c2}, \Omega_{d2}) \quad (8)$$

where the characteristic frequencies are defined as follows (generalizing Equation 7):

$$\Omega_{c1,c2}(\kappa)=\sqrt{\frac{\kappa^2+\varepsilon_{1\infty}\Omega_0^2\mp\sqrt{\left(\kappa^2+\varepsilon_{1\infty}\Omega_0^2\right)^2-4\varepsilon_{1\infty}\kappa^2}}{2\varepsilon_{1\infty}}} \quad (9a,b)$$

$$\Omega_{d1,d2}(\kappa,s,t)=\sqrt{\frac{\kappa^2+\varepsilon_{2\infty}s^2\Omega_0^2\mp\sqrt{\left(\kappa^2+\varepsilon_{2\infty}s^2\Omega_0^2\right)^2-4\varepsilon_{2\infty}\kappa^2t^2}}{2\varepsilon_{2\infty}}} \quad (9c,d)$$

In Figures 4a and 4b the curves for Ω *versus* κ and Ω *versus* $\sqrt{p_2}$, respectively, has been plotted for the assumed InAs/GaSb/InAs three-layer structure in the case of $d = d_0$.

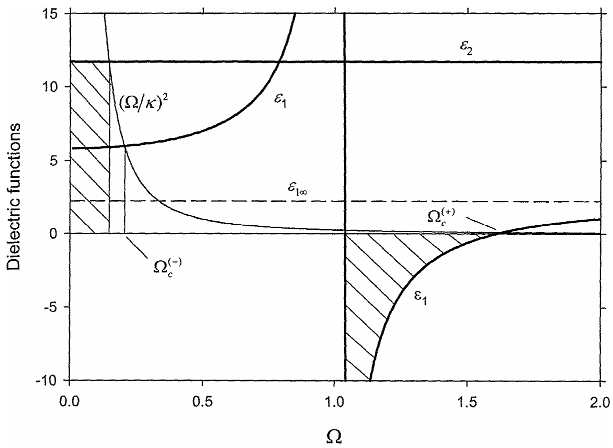

Figure 1. Plots of ε_{01} , ε_{02} and $(\kappa/\Omega)^2$ *versus* Ω for the special case where ε_{02} is independent of Ω . The shaded regions represent the bands of Ω where the phonon-polariton modes may exist. The parameters are for a three-layer structure such as NaCl/Si/NaCl (see the text) and $\kappa = 0.5$.

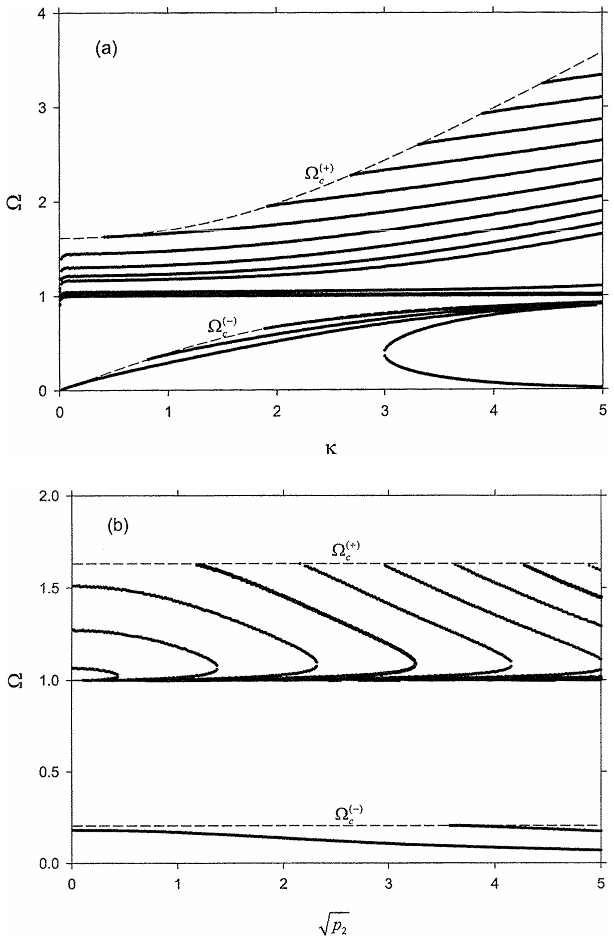

Figure 2. Dispersion relation curves, (a) Ω *versus* κ for constant p_2 and (b) Ω *versus* $\sqrt{p_2}$ for constant κ. The parameters are for a NaCl/Si/NaCl structure with $d = \frac{1}{2} d_0$ (see the text).

From this section we conclude that the resulting dispersion relations show features that are different from those of nonlinear plasmon-polariton modes (*e.g.* there are typically two bands of frequencies) and are distinctive of the phonon frequencies. With other choices of materials for the phonon case (*e.g.*, such that $\varepsilon_{2\infty} < \varepsilon_{1\infty}$) the behavior of the phonon-polariton bands would be different. There is a variety of possible forms for the spectra, sensitively depending on the choice of materials. This topic would be attractive for experimental studies.

4. Nonlinear Film Bounded by Nonlinear Media

In this general case we allow both the film and bounding medium to have nonlinear, frequency-dependent dielectric functions, while restricting the calculations to self-focusing media (*i.e.* both α_1 and α_2 are positive). Without loss of generality, the functions $g_1 (= g_3)$ and g_2 may again be calculated by a procedure similar to that of Ref. 6. Different possible types of phase trajectories in the intermediate nonlinear layer may occur where the boundary conditions for the electric field amplitude lead to physical solutions for the phonon-polariton modes. In a similar way to the previous case of plasmon-polaritons, the dispersion relation is considered in the film while the properties of the symmetric bounding medium enter the result through the value of the integration constant C_2. Here we generalize section 3 further by distinguishing two different cases, namely $C_2 > 0$ and $C_2 < 0$, depending on the frequency and the parameters of the materials.

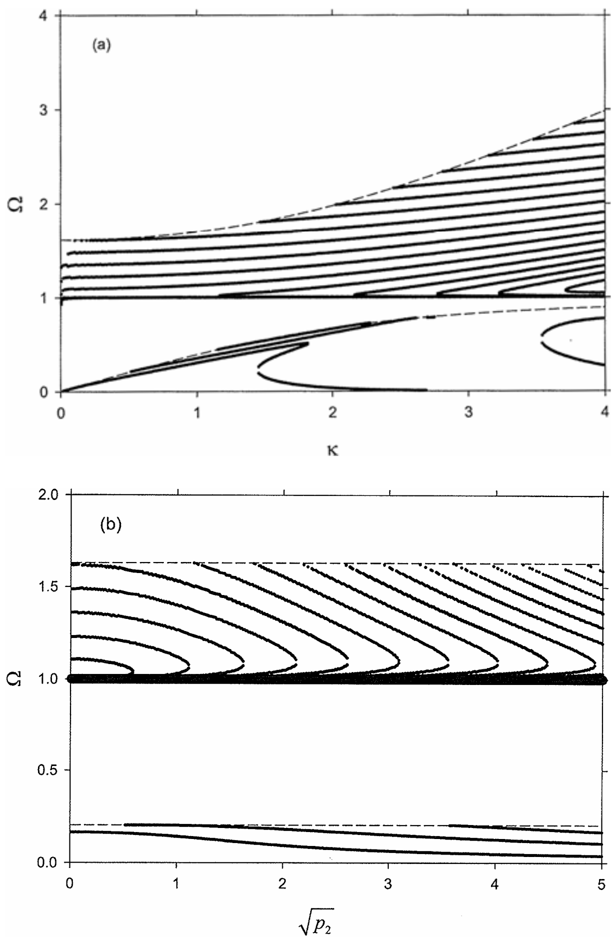

Figure 3. The same as for Figure 2, but for larger film thickness with $d = d_0$.

If we apply the first case, $C_2 > 0$, then the restricted regions for the physical solutions would be those where the reduced frequency Ω is confined to have the values given formally as in Equation 8, but with the four characteristics frequencies now re-defined as

$$\Omega_{c1,c2}(\kappa,r) = \sqrt{\frac{\kappa^2+\varepsilon_{\infty 1}\Omega_0^2+rp_2 \mp \sqrt{\left(\kappa^2+\varepsilon_{\infty 1}\Omega_0^2+rp_2\right)^2-4\kappa^2\left(\varepsilon_{\infty 1}+rp_2\right)}}{2\left(\varepsilon_{\infty 1}+rp_2\right)}} \quad (10a)$$

$$\Omega_{d1,d2}(\kappa,r,s,t) = \sqrt{\frac{\kappa^2+\varepsilon_{\infty 2}s^2\Omega_0^2+rp_2 \mp \sqrt{\left(\kappa^2+\varepsilon_{\infty 2}s^2\Omega_0^2+rp_2\right)^2-4\kappa^2\left(\varepsilon_{\infty 2}t^2+rp_2\right)}}{2\left(\varepsilon_{\infty 2}+rp_2\right)}} \quad (10b)$$

where we denote $r = p_1/p_2$. The formalism as described earlier for the special case of a nonlinear film bounded by a linear medium carries over to the case of a nonlinear bounding medium with the exception that the definition of the effective wave vector q (along with the other quantities b and m in Equation 6) must be generalized. The dispersion relations for the symmetric and antisymmetric modes can be derived following the same procedure as developed in section 3 (see also Ref. 6). We additionally have modes that are analogous to those found in section 3, but which have modified properties. Therefore, the electric field amplitude is obtained by integrating Equation (3) following the path of the phase trajectory under consideration. This again leads to the generalization of all possible symmetric and antisymmetric phonon-polariton modes and the full set of dispersion relation is given by the same expression as in Ref. 6, but with new parameters appropriate to the phonon case. In Figure 5 the dispersion curve, Ω versus κ, has been plotted for InSb representing the bounding medium and GaSb denoting the film, taking $p_2 = 0.05$ and $p_1 = 0.025$ (implying $r = 0.5$), where the dielectric functions of both media are nonlinear and frequency dependent. In this particular case we have solved for Ω subject to the conditions that $(\kappa/\Omega)^2 > \varepsilon_1(\omega) + p_1$, $\varepsilon_2(\omega) > \varepsilon_1(\omega)$, $p_2 > p_1$ and $(\kappa/\Omega)^2 > \varepsilon_2(\omega)$.

We may next comment briefly on the other case, namely when $C_2 < 0$. Following the phase trajectory scheme to determine the field amplitude in the layer we obtain the periodic solution expressible in terms of the elliptic function $\mathrm{dn}(x|m)$ which is an even function of x. The dispersion relations can be derived in a similar way as discussed in the previous case and the results are formally the same as Ref. 6. Examples of dispersion curves for this general case have the same features as those in section 3.

Finally we note that the special case in which the film can be characterized by a linear dielectric function (which also may be frequency dependent) while the bounding media is characterized by a nonlinear frequency dependent dielectric function of the Kerr-type can readily be obtained from the results of this section by formally applying the limit $p_2 \to 0$ (with $p_1 \neq 0$) to the previous expressions. In the analogous case of plasmon-polariton modes a detailed discussion can be found in Ref. 6.

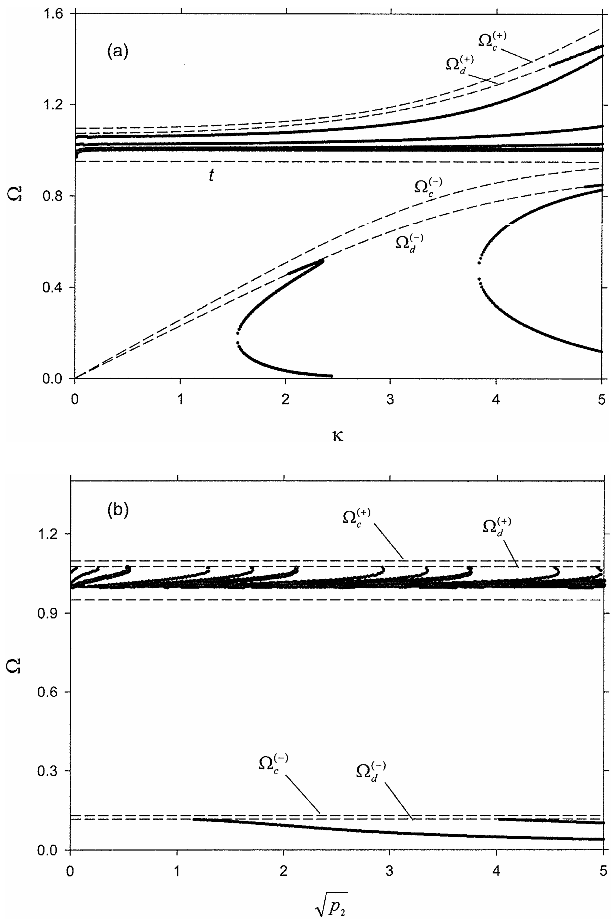

Figure 4. Dispersion relation curves, (a) Ω *versus* κ for $d = d_0$ constant p_2 and (b) Ω *versus* $\sqrt{p_2}$ for constant κ. The parameters are for a InAs/GaSb/InAs structure with $d = \frac{1}{2}d_0$ (see the text).

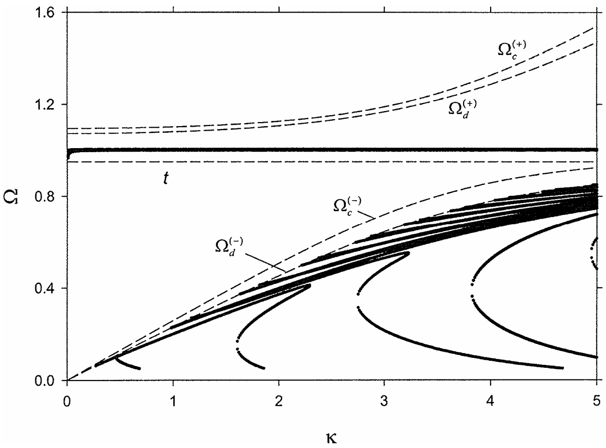

Figure 5. Dispersion relation curve, Ω *versus* κ, for the general case where each layer of the three-layer system, InAs/GaSb/InAs, are both frequency dependent and have Kerr-type nonlinearity for $d = \frac{1}{2} d_0$ (see the text).

6. Conclusions

The propagation of the phonon-polariton modes has been investigated for a three layered structure where the layers generally may have a Kerr-type nonlinearity characterized by $\varepsilon_1 (= \varepsilon_3)$ for the bounding medium and ε_2 for the bounded slab (film). The calculations for dispersion relations of the frequency versus wave vector and frequency versus nonlinearity have been carried out for different cases. First, a special case (a film with Kerr-type nonlinearity bounded by a linear medium) was considered. The electric field amplitude $E(z)$ was calculated using a phase trajectory scheme leading to Jacobian elliptic integrals. The dispersion curves show a sequence of branches labeled by *J*, in accordance with the periodicity of $\mathrm{cn}(x|m)$ function, for each value of film thickness *d*. The nonlinear phonon-polariton modes were found to have distinct branches characteristic of the optical phonons and showing features that are different from those of plasmon-polaritons [6]. Another special case which is a linear film bounded on each side by a nonlinear medium, was also considered. In illustrating the dispersion curves we also made applications of the most general structure where both media have Kerr-type nonlinearity.

For the most interesting situation in which both media are nonlinear we distinguished two different cases with respect to the integration constant C_2, namely $C_2 > 0$ or $C_2 < 0$. The numerical calculations show how the nonlinearity of the bounding media affects the resulting dispersion relations leading to new modes. This theory may also be extended to apply to the superlattice case in which two different materials (at least one being nonlinear and at least one having a frequency dependent dielectric function) could be considered as the constituent layers stacked periodically.

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