# Free optical vibrations of an infinite plate of homogeneous isotropic elastic matter

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We adapt the standard theory of the free acoustic vibrations of an infinite plate of homogeneous isotropic elastic matter to the corresponding case of optical vibrations. Treating nonpolar material first, we show that the effect of the free surface is to couple LO and TO modes, and we demonstrate the existence of the optical analog of Rayleigh waves. Interface and guided modes are both present, and their respective mode patterns are derived. In polar materials the coupling between LO and TO is different because of the frequency splitting due to the ionic fields, but surface modes are still present. This result contradicts the conclusion of the hydrodynamic model that surface modes do not exist. The polar character also allows the existence of surface polaritons. It is shown that the standard description of these modes, which neglects the elastic properties of the material, is physically invalid. The effect of the free surface is to couple surface polaritons and LO modes, and a description is given of the mode patterns that may occur. General expressions for energy flux are given, and boundary conditions for the general case are suggested. This treatment goes some way towards reconciling the various theoretical models of phonon confinement that have been advanced recently.

## I. INTRODUCTION

A description of the free acoustic vibrations of an infinite plate of homogeneous isotopic elastic matter has been available for over a century,[1] but a corresponding description for optical vibrations has not been given, to the author's knowledge. Here we report such a description for nonpolar material, and subsequently for polar material. The motivation was to establish a consistent account of longitudinally polarized (LO) and transversely polarized (TO) optical modes in a thin layer treated as an isotopic elastic and dielectric continuum to act as a basis for calculating the electron-phonon and hole-phonon scattering rates in layered semiconductors. Here we limit attention to the free-standing plate since this is the simplest system that illustrates the basic physics. The latter emerges in the form of an unavoidable coupling of LO and TO modes brought about by the presence of a surface. Such a coupling between acoustic modes (LA and TA) is well known, but its significance for optical modes has not been widely appreciated hitherto.

Models of confined optical modes which are currently in the literature achieve differing levels of sophistication. The earliest and simplest was the dielectric-continuum (DC) model in which only electrical boundary conditions were used to determine LO mode patterns.[2,3] Such a model, clearly, could not describe confinement in nonpolar material, and it soon came into serious conflict with the predictions of linear chain models[4] through its lack of boundary conditions, which referred to mechanized stability. A more successful, but still overly simple, model described LO confinement using hydrodynamic (HD) boundary conditions,[5] in which it was argued that since an LO mode was characterized by zero electric displacement it contained no electromagnetic energy, and so only purely mechanical conditions had to be satisfied at a boundary. The HD model showed much closer agreement with linear chain models, though it was soon pointed out that HD boundary conditions were not strictly consistent with the actual ionic motion at the interface.[6] Nevertheless, the HD model displayed the twin virtues of conserving energy and establishing mechanical stability through its expansion of the Born-Huang model to include dispersion. On the other side of the balance sheet, it displayed the perceived defect of allowing tangential electric fields, and with them, the scalar potential, to be discontinuous at the interface.

As three-dimensional models of the lattice dynamics—usually referred to as microscopic ($M$) models—became more sophisticated, two aspects emerged. One was the primacy of mechanical conditions, and the other was the added complexity introduced by the elastic anisotropy of the crystals considered (mostly GaAs and AlAs). Huang and Zhu[7] attempted to reconcile the DC and $M$ models by *ad hoc* addition of scalar potentials so both electric and mechanical boundary conditions were satisfied, and by a reinterpretation of off-axis modes. Bechstedt and Gerecke[8] attempted a similar reconciliation. Both pairs of workers obtained a hybridization of LO guided modes with Fuchs-Kliewer[9] (FK) surface polaritons. The additions conceived by Huang and Zhu, besides being *ad hoc,* destroyed the orthogonality of the modes, but this has been rectified recently by Haupt and Wendler[10] who have gone on to calculate electron-phonon scattering rates. Some unsatisfactory elements in all of these approaches have been pointed out recently.[11]

A continuum theory of confined optical modes should be able to stand on its own and be internally self-consistent. It should also be applicable to nonpolar and polar materials. Only when these criteria are satisfied can a judgment be made concerning the validity of applying it to a real system composed of atoms. The HD model satisfies these criteria by limiting itself strictly to LO modes and seeing any discontinuity of the scalar potential that may arise as resolvable only on an atomic scale. The models mentioned above start from the basis of the DC model, which is clearly not self-consistent. In what follows we develop a continuum model that goes beyond the HD model in that it treats TO modes and the LO-TO interaction and which rediscovers the HD model as a special case. It bases itself on the firmly established theory of elasticity.

Our approach is straightforward. We first identify the dispersion of optical modes with the macroscopic elastic properties of the material. This immediately allows us to take over the theory established for acoustic modes in nonpolar material and apply it to optical modes. In doing so we discover the optical-mode analog of Rayleigh waves. The transition to polar material is effected simply by taking into account the splitting of LO and TO frequencies and introducing a new mode — the surface polariton. Throughout, we assume that the medium is elastically isotropic, which allows us to keep LO and TO modes distinct and allows us to orientate Cartesian axes irrespective of crystallographic direction. We take the $x$ axis parallel to the direction of propagation in the plane of the plate and the $z$ axis to be perpendicular to the plane of the plate. Modes with displacements in the plane of incidence ($xz$) are the LO mode and the $p$-polarized TO mode (or just $p$-TO); the mode with displacement at right angles to the plane of incidence (i.e., along the $y$ direction) is the $s$-polarized TO mode (or $s$-TO) (Fig. 1).

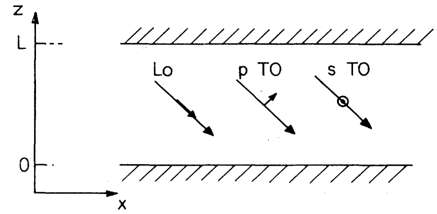


FIG. 1. Coordinate system and mode designation. Depicted are the propagation and polarization directions of the plane-wave components that together with their reflections make up waves with a standing component in the $z$-direction, which propagate along the $x$ axis.

## II. OPTICAL PHONONS

The dispersion relation for long-wavelength optical phonons in nonpolar material can be written as follows:

$$(\omega^2-\omega_0^2)\mathbf{u}(\mathbf{k})=-H(\mathbf{k})\mathbf{u}(\mathbf{k}) , \tag{1}$$

where

$$H(\mathbf{k})=\begin{bmatrix} Ak_x^2+B(k_y^2+k_z^2) & Ck_xk_y & Ck_xk_z \\ Ck_yk_x & Ak_y^2+B(k_z^2+k_x^2) & Ck_yk_z \\ Ck_zk_x & Ck_zk_y & Ak_z^2+B(k_x^2+k_y^2) \end{bmatrix} \tag{2}$$

is a $3\times3$ matrix, $\mathbf{u}(\mathbf{k})$ is the relative displacement, and $\omega_0$ is the frequency at $k=0$. We take $H(\mathbf{k})$ to be identical to the matrix for acoustic modes, from which it follows that

$$\begin{aligned} &A=v_L^2=c_{11}/\rho , \quad B=v_T^2=c_{44}/\rho , \\ &C=(c_{12}+c_{44})/\rho , \end{aligned} \tag{3}$$

where $v_L$, $v_T$ are the velocities of LA and TA modes, $c_{11}$, etc. are the elastic constants, and $\rho$ is the mass density. Thus the dispersion relations for LO and TO modes are

$$\begin{aligned} \omega^2&=\omega_0^2-v_L^2k^2 \text{ LO} \\ &=\omega_0^2-v_T^2k^2 \text{ TO } (s \text{ and } p) , \end{aligned} \tag{4}$$

where $k^2=k_x^2+k_y^2+k_z^2$ for all directions provided the material is elastically isotropic, viz.,

$$c_{11}-c_{12}-2c_{44}=0 . \tag{5}$$

We will assume this to be the case.

Dispersion arises as a consequence of the elastic stresses produced by a traveling optical wave. The equivalent strains are given by

$$\begin{aligned} &S_1=-\frac{\partial u_x}{\partial x} , \quad S_2=-\frac{\partial u_y}{\partial y} , \quad S_3=-\frac{\partial u_z}{\partial z} , \\ &S_4=-\frac{1}{2}\left[\frac{\partial u_z}{\partial y}+\frac{\partial u_y}{\partial z}\right] , \\ &S_5=-\frac{1}{2}\left[\frac{\partial u_x}{\partial z}+\frac{\partial u_z}{\partial x}\right] , \\ &S_6=-\frac{1}{2}\left[\frac{\partial u_y}{\partial x}+\frac{\partial u_x}{\partial y}\right] . \end{aligned} \tag{6}$$

The minus signs arise as a consequence of the out-of-phase vibration of the two atoms in the unit cell. The stresses are given by the usual set of equations:

$$
\begin{aligned}
T_1 &= c_{11}S_1 + c_{12}(S_2+S_3) , \\
T_2 &= c_{11}S_2 + c_{12}(S_3+S_1) , \\
T_3 &= c_{11}S_3 + c_{12}(S_1+S_2) , \\
T_4 &= 2c_{44}S_4 , \\
T_5 &= 2c_{44}S_5 , \\
T_6 &= 2c_{44}S_6 .
\end{aligned} \tag{7}
$$

Equations (6) and (7) allow us to calculate dilatational and shear stresses produced by a travelling optical wave.

The assumption of isotropy allows us to decompose any displacement $\mathbf{u}$ into a longitudinally polarized part $\mathbf{u}_L$ and a transversely polarized part $\mathbf{u}_T$

$$\mathbf{u} = \mathbf{u}_L + \mathbf{u}_T \tag{8}$$

with $\mathbf{u}_L$ and $\mathbf{u}_T$ defined by[12]

$$
\begin{aligned}
\nabla \times \mathbf{u}_L &= 0 , \\
\nabla \cdot \mathbf{u}_T &= 0 .
\end{aligned} \tag{9}
$$

Of course, $\mathbf{u}_L$ and $\mathbf{u}_T$ must have the same time dependence, i.e, the same frequency.

## III. NONPOLAR MATERIAL

The boundary condition which must be satisfied when the surface is free is that the dilational stress perpendicular to the surface and shear stress across the surface vanish. The situation is simplest for $s$-polarized TO modes. A solution is

$$u_y = e^{ik_x x}(Ae^{ik_z z} + Be^{-k_z z}) , \tag{10}$$

which satisfies $\nabla \cdot \mathbf{u} = 0$. It must also satisfy

$$T_3 = 0 , \quad T_4 = 0 , \quad T_5 = 0 \tag{11}$$

at $z=0$ and $z=L$. For this wave the stresses $T_3$ and $T_5$ are zero everywhere. To obtain $T_4=0$ we must have

$$u_y = e^{ik_x x}\cos k_z z , \quad k_z L = n\pi , \tag{12}$$

where $n$ is an integer. In this case no mixing with other modes is required.

The surface couples LO and $p$-polarized TO modes. Thus we take

$$
\begin{aligned}
u_x &= k_x e^{ik_x x}(Ae^{ik_L z} + Be^{-ik_L z}) \\
&\quad + k_T e^{ik_x x}(Ce^{ik_T z} + De^{-ik_T z}) , \\
u_z &= k_L e^{ik_x x}(Ae^{ik_L z} - Be^{ik_L z}) \\
&\quad - k_x e^{ik_T x}(Ce^{ik_T z} - De^{-k_t z}) .
\end{aligned} \tag{13}
$$

We distinguish the $z$ components of the LO and TO wave vectors $k_L$ and $k_T$, respectively. Equation (9) is satisfied for the individual LO and TO components, and it is clear that the condition $T_4=0$ holds everywhere. The wave-vector component along the surface must be common to both components. They must also have the same frequency, from which (Fig. 2)

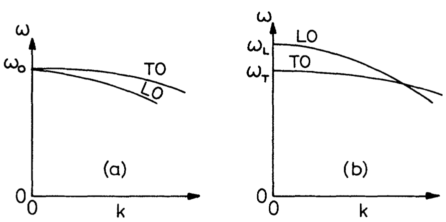


FIG. 2. Schematic dispersion relationship of (a) nonpolar and (b) polar material.

$$\omega^2 = \omega_0^2 - v_L^2(k_x^2 + k_L^2) = \omega_0^2 - v_T^2(k_x^2 + k_T^2) . \tag{14}$$

It is convenient to define a "plate" velocity $v$ and a parameter $s$ such that

$$\omega_0^2 - \omega^2 = v^2 k_x^2 \quad \text{and} \quad s = v^2/v_T^2 , \tag{15}$$

from which

$$k_L^2 = (\gamma s - 1)k_x^2, \quad k_T^2 = (s-1)k_x^2 , \tag{16}$$

where $\gamma = v_T^2/v_L^2 = c_{44}/c_{11}$.

As indicated in the Appendix, the boundary conditions can be satisfied provided the following equation is true:

$$
\begin{aligned}
&(qr+pt)^2 \sin k_T L \sin k_L L \\
&\qquad - 2qrpt[\cos(k_L - k_T)L - 1] = 0 ,
\end{aligned} \tag{17}
$$

where

$$
\begin{aligned}
p &= \gamma(s-2) , \quad q = 2\gamma\sqrt{s-1} , \\
r &= 2\sqrt{\gamma s - 1}, \quad t = s-2 .
\end{aligned} \tag{18}
$$

This admits of two waves. One is a guided mode consisting of coupled phased-matched LO and TO waves, the other is a surface mode consisting of coupled evanescent LO and TO waves. The latter is the optical analog of the Rayleigh wave in acoustics. Some mode patterns are described below.

### A. Guided modes

These waves have phase-matched components with the following wave vectors:

$$k_L L = n_L \pi , \quad k_T L = n_T \pi , \quad n_T - n_L = 2m , \tag{19}$$

where $n_L$, $n_T$, and $m$ are all integers. The displacement in the $z$ direction can be described by sine or cosine. The sine solution is

$$
\begin{aligned}
u_x &= 2Ae^{ik_x x}\left[ k_x \cos k_L z + k_T \frac{p}{q}\cos k_T z \right] , \\
u_z &= 2iAe^{ik_x x}\left[ k_L \sin k_L z - k_x \frac{p}{q}\sin k_T z \right] ,
\end{aligned} \tag{20}
$$

and the cosine solution is

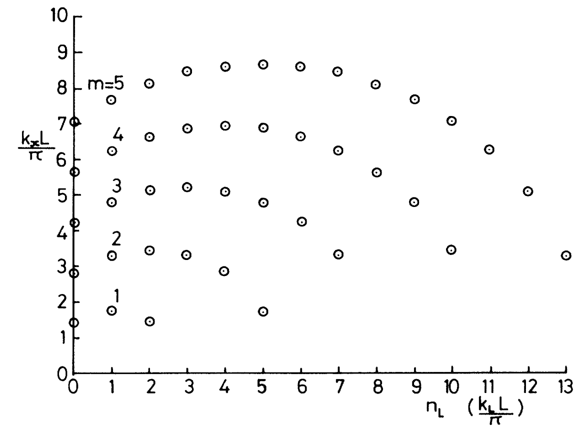


FIG. 3. Relation between $k_L$ and $k_x$ for guided modes in nonpolar material with $\gamma=\frac{1}{3}$. $L$ is the width of the plate and $2m=n_T-n_L$ ($n_L=k_L L/\pi$, $n_T=k_T L/\pi$).

$$u_x=2iAe^{ik_x x}\left[k_x \sin k_L z - k_T \frac{r}{t}\sin k_T z\right],$$
$$u_z=2Ae^{ik_x x}\left[k_L \cos k_L z + k_x \frac{r}{t}\cos k_T z\right]. \tag{21}$$

Figure 3 shows the relationship between $k_L$ and $k_x$ when $\gamma=\frac{1}{3}$. The spectrum can be seen as consisting of branches defined by the integer $m$. Figure 4 depicts the corresponding dispersion. All branches are associated with a velocity factor $s\approx 3$, corresponding to a velocity $v$ close to $v_L$. Note that the choice of $\gamma$ ($=c_{44}/c_{11}$) to be one-third in these examples is reasonable in view of the actual ratios of elastic constants found among semiconductors.

These guided waves are a profound hybridization of LO and TO modes, which arises because their frequencies lie close together. The frequency of the TO mode is the higher as a consequence of the smaller velocity of TA modes. One consequence of this is that the smallest TO vector component in the $z$ direction that is allowed is one where $n_L=1$ and $n_T=3$, to satisfy Eq. (19). Confinement eliminates $n_T=1$ and $n_T=2$.

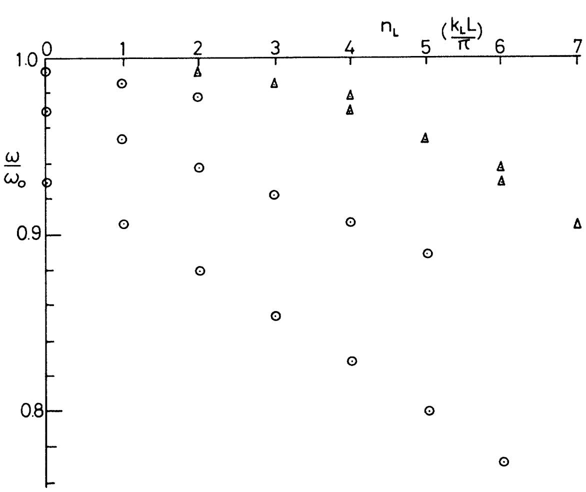


FIG. 4 Dispersion for guided modes in nonpolar material with $\gamma=\frac{1}{3}$ and $\omega_0 L/v_T\pi=20$. (⊙ LO modes; △ TO modes.) ($n_L=k_L L/\pi$).

### B. Surface modes

No surface-wave solutions exists for $s$-polarized TO modes, but Eq. (17) allows both $k_L$ and $k_T$ to be imaginary. For brevity we quote mode patterns only for $L\rightarrow\infty$ and $L\rightarrow 0$, since simple expressions are obtained in these limits.

For $L\rightarrow\infty$, the antisymmetric solution is, with $k_L=i\alpha_L$ and $k_T=i\alpha_T$,

$$u_x=2Ae^{ik_x x}\left[k_x e^{-\alpha_L L/2}\cosh\alpha_2(z-L/2) + i\alpha_T \frac{p}{q} e^{-\alpha_T L/2}\cosh\alpha_T(z-L/2)\right],$$
$$u_z=2Ae^{ik_x x}\left[-i\alpha_L e^{-\alpha_L L/2}\sinh\alpha_L(z-L/2) + k\frac{p}{q} e^{-\alpha_T L/2}\sinh\alpha_T(z-L/2)\right] \tag{22}$$

and the symmetric solution is

$$u_x=2Ae^{ik_x x}\left[-k_x e^{-\alpha_L L/2}\sinh\alpha_L(z-L/2) - i\alpha_T \frac{p}{q} e^{-\alpha_T L/2}\sinh\alpha_T(z-L/2)\right],$$
$$u_z=2Ae^{ik_x x}\left[i\alpha_2 e^{-\alpha_L L/2}\cosh\alpha_L(z-L/2) - k_x \frac{p}{q} e^{-i\alpha_T L/2}\cosh\alpha_T(z-L/2)\right]. \tag{23}$$

In this limit Eq. (17) reduces to

$$qr+pt=0\ , \tag{24}$$

which leads to a cubic equation for $s$, the velocity factor, viz.,

$$s^3-8s^2+8(3-2\gamma)s-16(1-\gamma)=0\ . \tag{25}$$

This equation is just that found in the theory of Rayleigh waves.[12]

With $\gamma=\frac{1}{3}$ this reduces to

$$(s-4)(3s^2-12s+8)=0\ . \tag{26}$$

The solutions are, therefore,

$$s=4,\quad s=2\left[1+\frac{1}{\sqrt{3}}\right],\quad s=2\left[1-\frac{1}{\sqrt{3}}\right]. \tag{27}$$

Only the last is consistent with having $k_L$ and $k_T$ imaginary. We thus obtain the relationship between $\alpha_L$, $\alpha_T$, and $k_x$, viz.,

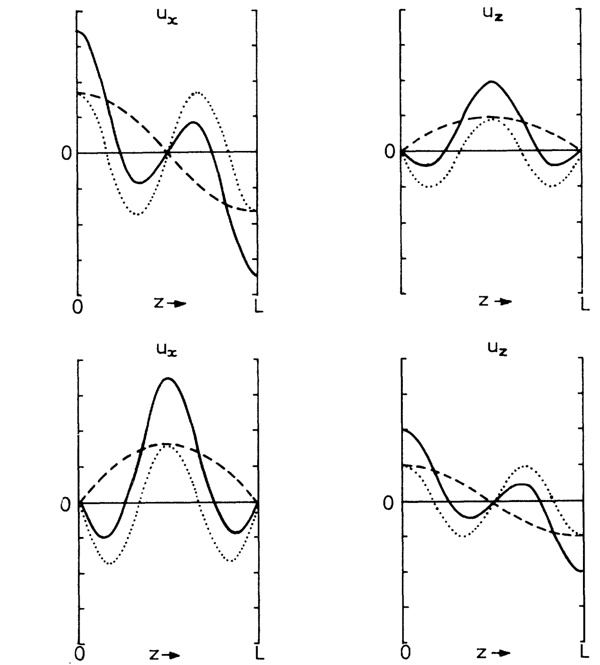


FIG. 5. Mode patterns for $n_L=1$, $n_T=3$ in nonpolar material. (Dashed line, LO; dotted line TO; continuous line, LO + TO.)

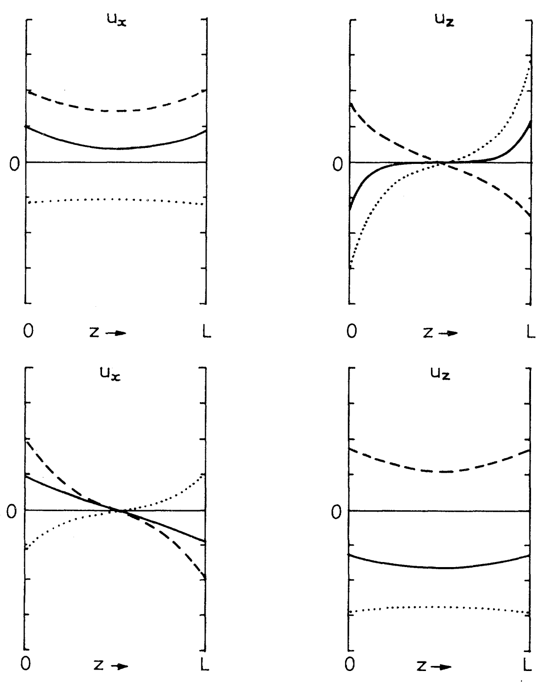


FIG. 6. Mode patterns for surface waves in nonpolar material (designations as in Fig. 5).

$$\alpha_L=\left[\frac{1}{3}\left[1+\frac{2}{\sqrt{3}}\right]\right]^{1/2} k_x \ ,$$
$$\alpha_T=\left[\frac{2}{\sqrt{3}}-1\right]^{1/2} k_x \ . \tag{28}$$

Note that the LO component drops off from the surface more rapidly than does the TO component.

For $L\rightarrow 0$ only the antisymmetric mode survives, the mode pattern being that of Eq. (22) with $\exp(-\alpha_T L/2)=1$. In this case we find

$$s=2(1-2\gamma) \ , \tag{29}$$

from which

$$\alpha_L=(1-2\gamma+4\gamma^2)^{1/2} \ , \quad \alpha_T=(4\gamma-1)^{1/2}k_x \ . \tag{30}$$

This completes our description of the allowed modes of a nonpolar plate. A depiction of mode patterns is given in Figs. 5 and 6.

## IV. POLAR MATERIAL

When the two atoms in the unit cell are oppositely charged their oscillations are accompanied by long-range electric fields that modify the elastic restoring forces and alter the frequencies of LO and TO modes. The dispersion relation for long-wavelength optical modes becomes modified along the following lines:

$$(\omega^2-\omega_T^2)\mathbf{u}(k)=[-H(k)+(\omega_L^2-\omega_T^2)I(k)]u(\mathbf{k}) \ , \tag{31}$$

where $\omega_L$, $\omega_T$ are the LO and TO zone-center frequencies, and

$$I(k)=\frac{1}{k^2}\begin{bmatrix} k_x^2 & k_xk_y & k_xk_z \\ k_yk_x & k_y^2 & k_yk_z \\ k_zk_x & k_zk_y & k_z^2 \end{bmatrix} \ . \tag{32}$$

The LO frequency is shifted upwards relative to $\omega_0$ of Sec. III. The main effect is this shift in frequency. For simplicity we will continue to regard the material as elastically isotropic with effective elastic constants modified by the polar fields. Consequently we can employ the analysis of the previous section, merely changing the definition of certain terms.

Thus we maintain the distinction between irrotational and divergenceless modes according to Eq. (9). It is convenient to modify the definition of the plate velocity $v$ as follows:

$$\omega_L^2-\omega^2=v^2k_x^2 \tag{33}$$

from which it follows that Eq. (16) becomes

$$k_L^2=(\gamma s-1)k_x^2 \ , \quad k_T^2=(s-1)k_x^2-k_0^2 \ , \tag{34}$$

where

$$k_0^2=\frac{\omega_L^2-\omega_T^2}{v_T^2} \ . \tag{35}$$

The secular equation, Eq. (17), remains valid, but now Eq. (18) becomes

$$p=\gamma(s-2), \quad q=2\gamma[(s-1)k_x^2-k_0^2]^{1/2}/k_x \ , \quad r=2\sqrt{\gamma s-1} \ , \quad t=(s-2)-(k_0^2/k_x^2) \ . \tag{36}$$

The splitting of the LO and TO frequencies profoundly modifies the ability of the surface to couple these modes together. Over the frequency range $\omega_L-\omega_T$ coupling of guided LO modes can only be accomplished with rapidly varying TO evanescent modes, and for $\omega \le \omega_T$, only short-wavelength LO modes can be involved. Because of these consequences, a description of coupled modes tends to stretch any continuum model to its limits of validity and perhaps beyond it. With the caveat we will proceed first to describe the guided modes.

### A. Guided modes

It may be noted that our results for $s$-polarized TO modes remain unaffected by the ionic nature of the material, and so we turn immediately to the coupling between guided LO and evanescent TO modes. Taking $k_T \approx ik_0$ and assuming for simplicity that $k_0$ is large, we obtain,

$$u_x=2Ae^{ik_xx}\left\{ik_x\sin k_Lx - ik_0\frac{r}{t}[e^{-k_0z}-\cos(k_LL)e^{-k_0L}e^{k_0z}]\right\},$$
$$u_z=2Ae^{ik_xx}\left\{k_L\cos k_Lz + k_x\frac{r}{t}[e^{-k_0z}+\cos(k_LL)e^{-k_0L}e^{k_0z}]\right\} \tag{37}$$

with $k_LL\approx n_L\pi$, $n_L$ an integer (Fig. 7). In this coupling the contribution of the TO mode to the dilational stress is negligible, but the presence of the TO mode is still vital to cancel out the shear stress. If shear is neglected, no TO mode need be involved, and this is exactly the implicit assumption underlying the HD model, whose boundary condition $\nabla\cdot\mathbf{u}=0$ at the surface is exactly the same as our condition $T_3=0$ when $\gamma=0$.

It is interesting to observe that the mode pattern of Eq. (37) satisfies the electromagnetic boundary conditions. The electric and magnetic fields associated with the TO mode are vanishingly small, so they do not enter. The LO mode has zero magnetic field and zero electric displacement $D$. Thus $D_z=0$, and since $k_L=n\pi$, the tangential electric field vanishes at each surface. The continuity of $D_z$ and $E_x$ is then satisfied by having no fields in the vacuum.

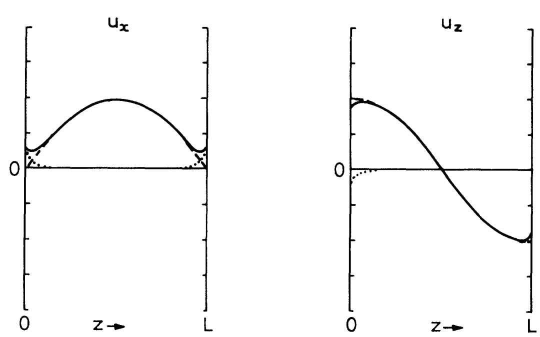


FIG. 7. Mode patterns for $n_L=1$ in polar material (with $k_x=k_L$) (designations as in Fig. 5). Mode patterns for surface waves are approximately those of Fig. 6 in the absence of the TO component.

The coupling between guided LO and guided TO for $\omega \le \omega_T$ gives the mode patterns already given in Eqs. (2) and (21) but now with $\omega_T$ replacing $\omega_L$ in Eq. (33), $k_TL=n_T\pi$, $k_LL=n_L\pi$ and $n_L-n_T=2m$,

$$k_L^2=\gamma k_0^2+k_x^2(\gamma s-1) \ , \quad k_T^2=(s-1)k_x^2 \tag{38}$$

and

$$p=\gamma[k_0^2+(s-2)k_x^2]/k_x^2 \ , \quad q=2\gamma\sqrt{s-1} \ , \quad r=(2/k_x)\sqrt{\gamma k_0^2+k_x^2(\gamma s-1)} \ , \quad t=(s-2) \ . \tag{39}$$

### B. Surface modes

There are still no surface $s$-polarized TO modes, but in general, surface waves consisting of mixed LO–$p$-polarized TO modes do exist, and their mode patterns for $L\to\infty$ are described by Eqs. (22) and (23) with $p$, $q$, $\alpha_L=ik_L$, $\alpha_T=ik_T$, given by Eqs. (34)–(36). Specific magnitudes must satisfy Eq. (24), which now leads to a quartic for the velocity factor $s$:

$$s^4-2(4+a)s^3+[8(3-2\gamma)+12a+a^2]s^2 -[16(1-\gamma)+8a(3-2\gamma)+4a^2]s+4a^2=0 \ , \tag{40}$$

where

$$a=k_0^2/k_x^2 \ .$$

Note that when $a=0$ Eq. (40) reduces to the nonpolar equation [Eq. (25)]. In the limit $a\to\infty$, the solution is

$$s=2 \ , \tag{41}$$

independent of $\gamma$, from which

$$\alpha_L^2=(1-2\gamma)k_x^2 \ , \quad \alpha_T^2=k_0^2-k_x^2 \ . \tag{42}$$

In this limit solutions exist provided $\gamma\le\frac{1}{2}$. Waves propagate with surface phase velocity equal to $2^{1/2}v_T$. The antisymmetric solution is

$$u_x=2Ak_xe^{ik_xx}\left\{e^{-\alpha_LL/2}\cosh\alpha_L(z-L/2) -2\frac{\alpha_L}{k_0}e^{-k_0L_2}\cosh k_0(z-L/2)\right\},$$
$$u_z=2iA\alpha_Le^{ik_xx}\left\{-e^{-\alpha_LL/2}\sinh\alpha_L(z-L/2) +2\frac{k_x^2}{k_0^2}e^{-k_0L/2}\sinh k_0(z-L/2)\right\} \tag{43}$$

and the symmetric solution is

$$u_x = 2Ak_x e^{ik_x x}\left[-e^{-\alpha_L L/2}\sinh_L(z-L/2) + 2\frac{\alpha_L}{k_0}e^{-k_0 L/2}\sinh k_0(z-L/2)\right]$$
$$u_z = 2iA\alpha_L r^{ik_x x}\left[e^{-\alpha_L L/2}\cosh\alpha_L(z-L/2) - 2\frac{k_x^2}{k_0^2}e^{-k_0 L/2}\cosh k_0(z-L/2)\right] . \quad (44)$$

The existence of this optical-mode analog to Rayleigh waves in polar material, as well as its appearance in nonpolar material, depends upon the shear modulus being nonzero. If $\gamma=0$, Eq. (42) shows that $\alpha_L^2=k_x^2$. In this case the LO component has the property $\nabla\times\mathbf{u}=0$ as well as $\nabla\times\mathbf{u}=0$. Such a mode must have zero amplitude, which is consistent with the prediction of the HD model[13] that no surface modes are possible in a free-standing plate. However, this conclusion derived from the HD model appears to be incorrect for real elastic solids, even though Eqs. (43) and (44) show that the amplitudes of the TO components are vanishingly small. We note that in this limit ($a\to\infty$), the solutions obtained above are valid for all plate thicknesses of interest. However, we also note that the tangential component of the electric field, which is proportional to $u_x$, is not zero at the surface, and so conventional electric boundary conditions are violated.

### C. Surface polaritons

One well-known consequence of polarity is that it allows electromagnetic waves to directly couple with TO modes to form surface polaritons, as described by Fuchs and Kliewer.[9] We will refer to these as FK modes. Their dispersion is depicted in Fig. 8. For large $k_x$ the mode patterns are of the following forms: antisymmetric,

$$u_x = 2Ae^{ik_x x}e^{-k_x L/2}k_x\cosh k_x(z-L/2) ,$$
$$u_z = -2iAe^{ik_x x}e^{-k_x L/2}k_x\sinh k_x(z-L/2) \quad (45)$$

and symmetric,

$$u_x = +2iAe^{ik_x x}e^{-k_x L/2}k_x\sinh k_x(z-L/2) ,$$
$$u_z = -2Ae^{ik_x x}e^{-k_x L/2}k_x\cosh k_x(z-L/2) . \quad (46)$$

When $k_x$ is large compared with the wave vector of light at the same frequency, these solutions, share the same frequency that lies between $\omega_T$ and $\omega_L$ and is given by

$$\omega_{\mathrm{FK}}^2 = \frac{\epsilon_s+\epsilon_0}{\epsilon_\infty+\epsilon_0}\omega_T^2 , \quad (47)$$

where $\epsilon_s$, $\epsilon_\infty$ are the static and high-frequency permittivities and $\epsilon_0$ is the permittivity of the vacuum. For simplicity in our discussion we will assume that this condition is met.

Two comments can be made. First, it would appear from Eqs. (45) and (46) that we are dealing with a null mode since both $\nabla\cdot\mathbf{u}$ and $\nabla\times\mathbf{u}$ are zero. This, however, is not really so since the decay constant in hyperbolic terms is only approximately equal to the wave vector $k_x$. In fact the modes are TO modes with finite, but in the present approximation, vanishingly small rotation, which describes the rate of change of magnetic field. The solutions quoted are those obtained neglecting retardation (i.e., assuming the velocity of light to be infinite). They match smoothly with an evanescent electromagnetic wave in the vacuum.

The second comment is that these solutions violate the condition that the elastic stress vanish at the surface. Fuchs-Kliewer modes, conventionally described, are therefore unphysical. This rather important defect can be rectified by coupling with an LO guided mode of the same frequency, as was done in Sec. IV A. For $L\to\infty$ only cosine waves couple, and the corresponding mode

$$u_x = 2Ae^{ik_x x}[ik_x\sin k_L z + ik_L(e^{-k_z z}-\cos k_L L e^{-k_x L}e^{k_x z})] ,$$
$$u_z = 2Ae^{ik_x x}[k_L\cos k_L z - k_L(e^{-k_x z}+\cos k_L L e^{-k_x L}e^{k_x z})] \quad (48)$$

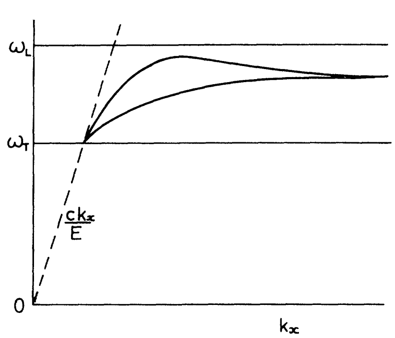


FIG. 8. Fuchs-Kliewer dispersion.

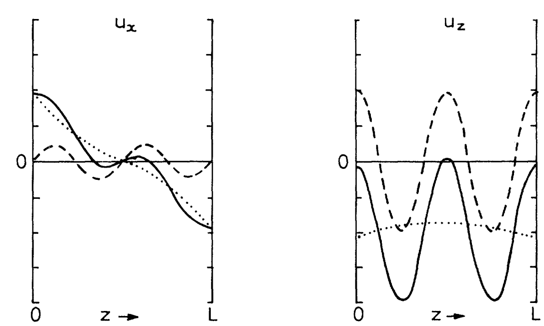


FIG. 9. Mode patterns for hybridized FK modes in polar material (with $KL=4\pi$, $k_xL=\pi$). (Dashed line, LO; dotted line, FK; continuous line, LO + FK.) Note that this combination of FK and LO is possible only if the two modes have the same frequency. In general only certain values of $k_x$ are allowed.

with $k_L L = n\pi$, satisfying

$$\omega_{\rm FK}^2 = \omega_L^2 - v_L^2(k_x^2 + k_L^2) \ . \tag{49}$$

This mode pattern (Fig. (9) satisfies all elastic and electromagnetic boundary conditions. Note that when $n$ is an odd integer, corresponding to an asymmetric guided mode, the coupling is to the asymmetric FK ode, and when $n$ is an even integer both components are symmetric. In other words, like parity hybridizes with like parity.

A further discussion of hybridization of FK and LO modes taking into account FK dispersion will be reported soon in the future.

## V. DISCUSSION

Our extension of classical elasticity theory to include optical modes has allowed us to described the mixing of LO and TO modes at a free surface and to obtain the confined phonon spectrum. In particular, it has allowed us to demonstrate the existence of surface optical modes, which are analogous to Rayleigh waves. Hitherto only HD theory has predicted interface modes, but only at internal surfaces and only for LO modes. DC theory and the more sophisticated versions mentioned in the Introduction do not predict interface modes other than FK polaritons, so our conclusions here are quite new. Deciding the ontology of surface waves is of some importance for the electron-phonon interaction, since under some circumstances surface modes can interact strongly with electrons.[14] For the same reason it is important to ascertain the strength of the interaction with FK polaritons. It has been argued elsewhere[11] that such an interaction has the nature of a magnetic ($\mathbf{A}\cdot\mathbf{p}$) rather than of an electrical ($e\phi$) interaction, but clearly an even greater reappraisal of the FK interaction is now necessary as a result of our demonstration of hybridization of FK and LO modes.

The foregoing treatment has included nothing but the simplest cases and has merely sketched a rough outline of the topic. An extension to the quantum-well situation is underway, where continuity of stress replaces the vanishing of stress, and continuity of energy flow is entailed. It is clear that a self-consistent picture of confined modes can emerge from this program, so it is of interest to ask how valid such a picture can be as a description of optical waves in real matter. There are at least four problems, and there may be more. These have to do with the following: (1) The spatial variation of the mode pattern, (2) the relation of relative displacement ($\mathbf{u}$) to ionic displacement, (3) the elastic anisotropy of real crystals, and (4) The electrical boundary conditions. We will comment on each in turn.

(1) It has already been remarked in Sec. IV that the polarity of the medium typically forced one of the components of a LO-TO hybridization to be rapidly varying, the more so the stronger the polarity, i.e., the more disparate the LO and TO zone-center frequencies. When the spatial variation of the mode function becomes significant over a primitive unit cell, the atomic nature of the medium cannot be ignored. The important parameters are the wave vector $k_0$, Eq. (35), and the dimension of the unit cell $a_0$. So for a continuum theory to be valid we expect

$$|k_0 a_0| < 1 \ . \tag{50}$$

For GaAs, $k_0 a_0 \approx 1$.

(2) The relation between HD boundary conditions and those arising from the linear-chain model was illuminated by Akero and Ando.[6] When the force constants on either side of the interface are the same, and when the interface is taken to lie midway between atom $A$ and atom $B$, the condition for mechanical stability simply entails the continuity of the displacements $u_A$ and $u_B$, viz.,

$$\left[u_A + \frac{a_0}{2}\frac{\partial u_A}{\partial z}\right]_1 = \left[u_A + \frac{a_0}{2}\frac{\partial u_A}{\partial z}\right]_2 , \quad \left[u_B - \frac{a_0}{2}\frac{\partial u_B}{\partial z}\right]_1 = \left[u_B - \frac{a_0}{2}\frac{\partial u_B}{\partial z}\right]_2 . \tag{51}$$

(Note there are no shear stresses here.) The parameter $a_0$ is now the interatomic spacing, assumed to be identical in both materials, and it is also assumed that $u_A$ and $u_B$ are describable by slowly varying envelope functions. Optical-mode relative displacement $u$ for long wavelengths is related to $u_A$ and $u_B$ as follows:

$$u_A = -\frac{u}{1+(M_A/M_B)}, \quad u_B = \frac{u}{1+(M_B/M_A)} , \tag{52}$$

where $M_A$, $M_B$ are the atomic masses. Substitution into Eq. (51) yields the connection rule

$$\begin{vmatrix} u \\ a_0\dfrac{\partial u}{\partial z} \end{vmatrix}_1 = T_{12} \begin{vmatrix} u \\ a_0\dfrac{\partial u}{\partial z} \end{vmatrix}_2 , \tag{53}$$

where

$$T_{12} = \frac{r_1}{r_2}\begin{bmatrix} \frac{1}{2}(R+R^{-1}) & \frac{1}{4}(R-R^{-1}) \\ R-R^{-1} & \frac{1}{2}(R+R^{-1}) \end{bmatrix} . \tag{54}$$

Here $r = r_m + r_m^{-1}$, $r_m = (M_A/M_B)^{1/2}$, $R = r_{m1}/r_{m2}$. This connection rule satisfies continuity of energy flow. The corresponding connection rule for the continuum theory is

$$T_{12} = \begin{bmatrix} 1 & 0 \\ 0 & \left[\dfrac{r_1}{r_2}\right]^2 \end{bmatrix} , \tag{55}$$

which agrees with microscopic theory only when $R = 1$, i.e., the mass ratios are common. In reality, the interface is never as precisely located as conventional microscopic theory assumes, and the position and properties of the interface affect the off-diagonal elements in Eq. (54). Consequently, boundary conditions relating relative displacement and elastic stresses in a continuum theory may not be a major source of error.

(3) Elastic anisotropy adds significant complexity. In directions other than the major crystallographic ones there is no clean distinction between LO and TO, and the degree of surface-induced hybridization depends upon direction following the directional dependence of dispersion. However, orientational effects of this sort are not likely to add anything qualitatively new to the picture obtained on the basis of an isotropic model. In any case, a straightforward application to the problem of the electron-phonon interaction will require some angular averaging. Anisotropy is therefore not likely to lead to significant problems.

(4) There is no question but that a transversly polarized wave of electromagnetic character must satisfy electromagnetic boundary conditions. The same is true of static fields. The question however is open concerning the applicability of electric boundary conditions to LO fields. We noticed that the surface optical modes described in Sec. IV B did not satisfy these conditions. Nor do linear-chain models in certain situations. Indeed, it was this problem that brought the DC model into clear conflict with microscopic models. The same question arises in the context of plasma waves. The special property of LO waves is that they have no electrical energy because the permittivity is zero. Electrical fields under this circumstance cannot exist in the absence of ionic polarization. At the surface of an ionic solid the electric field must therefore drop to zero over a distance of the order of atomic dimensions. If this is true, there is as much need for a continuum theory to address the issue as it would be for it to address the problem of continuity of interatomic forces, or indeed the continuity of matter. A surface or an interface is already a discontinuity of matter plus its properties, including polarization and associated fields, which suggests that it is unnecessary to add nonmechanical boundary conditions. If this view is adopted, it becomes clear that the DC model, insofar as it refers to the confinement of LO modes, is fundamentally flawed, and discontinuities of LO fields are to be expected. On the other hand, if this is denied, then one of the things that follows is that the surface modes of Sec. IV B cannot survive. This implies that surface modes exist in nonpolar material but not in polar material irrespective of the strength of polarity, provided the latter is nonzero. We appear to have exchanged one type of discontinuity for another. Note one final comment. The necessity for any boundary condition must be justified by arguments based on the physics of the situation. Hitherto there appears to be no such argument specifically directed at $\epsilon(\omega)=0$ LO modes for ensuring the continuity of the tangential component of the electric field.

It turns out, however, that both elastic and electromagnetic boundary conditions can be satisfied by invoking a scheme of triple hybridization involving LO, TO, and FK modes and distinguishing carefully between scalar and vector potentials.

We conclude with some comments on energy flow. General expressions for the energy flow in the $z$-direction may be written as follows:

$$S_z=-\frac{\bar{\rho}\omega v_L^2}{2i}(u_z^*\nabla\cdot\mathbf{u}-u_z\nabla\cdot\mathbf{u}^*)\ \ \text{LO}\ ,$$
$$=-\frac{\bar{\rho}\omega v_T^2}{2i}[u_x^*(\nabla\times\mathbf{u})_y-u_x(\nabla\times\mathbf{u}^*)_y]\ \ \text{TO}\ , \tag{56}$$

where $\bar{\rho}$ is the reduced density. The solutions we have derived all obey $S_z=0$. In the general case a basic boundary condition must be the continuity of $S_z$. Similarly we must have continuity of the stress components. These considerations suggest that if the $z$ axis is perpendicular to the interface, the general boundary conditions to be satisfied for LO and TO modes are

$$S_{z'},T_{3'},T_{4'},T_5 \text{ continuous}\ . \tag{57}$$

In general, we must add the usual electromagnetic boundary conditions.

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## APPENDIX

The displacement of Eq. (13) must satisfy the following boundary conditions at $z=0$ and $L$:

$$T_3=-c_{11}\left[(1-2\gamma)\frac{\partial u_x}{\partial x}+\frac{\partial u_z}{\partial z}\right]=0\ , \tag{A1}$$

$$T_5=-c_{44}\left[\frac{\partial u_x}{\partial z}+\frac{\partial u_z}{\partial x}\right]=0\ . \tag{A2}$$

This entails the vanishing of the determinant

$$\begin{vmatrix} p & p & -q & -q \\ r & -r & t & -t \\ pf_L & pg_L & -qf_T & -qg_T \\ rf_L & -rg_L & tf_T & -tg_T \end{vmatrix}=0\ , \tag{A3}$$

where

$$\begin{aligned} &p=1-2\gamma+(k_L^2/k_x^2)\ ,\quad q=2\gamma k_T/k_x\ ,\\ &r=2k_L/k_x\ ,\quad t=(k_T^2/k_x^2)-1\ ,\\ &f_L=e^{ik_LL}\ ,\quad g_L=e^{-ik_LL}\ ,\\ &f_T=e^{ik_TL}\ ,\quad g_T=e^{-ik_TL}\ . \end{aligned} \tag{A4}$$

This leads to the secular equation

$$(qr+pt)^2\sin k_TL\ \sin k_LL-2qrpt[\cos(k_L-k_T)L-1]=0 \tag{A5}$$

with the constraint on the frequency, namely,

$$\omega^2=\omega_0^2-v_L^2(k_x^2+k_L^2)=\omega_0^2-v_T^2(k_x^2+k_T^2) . \quad \text{(A6)}$$

The amplitudes are as follows:

$$B=\frac{[qr(f_T-g_T)+pt(2f_L-f_T-g_T)]}{\Delta}A , \quad \text{(A7)}$$

$$C=\frac{qr(f_L+g_L-2g_T)+pt(f_L-g_L)}{\Delta}\cdot\frac{p}{q}A , \quad \text{(A8)}$$

$$D=\frac{qr(2f_T-f_L-g_L)+pt(f_L-g_L)}{\Delta}\cdot\frac{p}{q}A , \quad \text{(A9)}$$

$$\Delta=qr(f_T-g_T)+pt(f_T+g_T-2g_L) . \quad \text{(A10)}$$

Sometimes it is useful to express the secular equation (A5) as follows:

$$(qr\sin\theta_L\cos\theta_T+pt\cos\theta_L\sin\theta_T)\times(qr\cos\theta_L\sin\theta_T+pt\sin\theta_L\cos\theta_T)=0 , \quad \text{(A11)}$$

where

$$\theta_L=k_LL/2 \text{ and } \theta_T=k_TL/2 .$$

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