Dispersive corrections to continuum elastic theory in cubic crystals

D. P. DiVincenzo*

*Laboratory of Atomic and Solid State Physics and Materials Science Center, Cornell University,
Ithaca, New York 14853-2501*

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Recent experiments using ballistic phonon imaging have provided very accurate pictures of acoustic-phonon dispersion for GaAs in the frequency range 0.1—1 THz. This work has been previously analyzed using microscopic lattice-dynamics models. An alternative description of this phenomenon which is natural, compact, and simple is provided by continuum elasticity theory with dispersive corrections included, as developed in the present paper. Previous models and experiments are successfully analyzed by the present theory. It is shown that the reason for the failure of many earlier models to match experiment is that they impart insufficient dispersion to the transverse acoustic branches. A single additional parameter contained in the dispersive elastic model appears to control the important features of the dispersion of these branches; six other parameters permitted by symmetry do not appear crucial. We show that the present dispersive elastic model can be derived as a rigorous limit of microscopic lattice-dynamical theory. The elastic model illuminates simple relationships between the phonon-imaging experiments and other physical phenomena, e.g., specific heat and ultrasound.

I. INTRODUCTION

The nature of phonon propagation in perfect crystals is a very old topic (Ref. 1, p. 68) but continues to stimulate fresh investigation. The interaction of phonons with defect centers and with electrons, and the perturbation of the thermal distribution of phonons in nonequilibrium transport problems, continue to play a role in several outstanding unsolved problems in solid-state physics. Thus, continuing attempts to construct new and simple ways to describe phonon energies and phonon propagation in crystals is worthwhile.

The present paper represents such an attempt. Here we perform a systematic study of the extension of continuum elasticity theory to relatively high-energy acoustic phonons. This is already the model of choice for long-wavelength, low-energy acoustic phonons; it has certain advantages of simplicity and economy of description compared with microscopic lattice-dynamical models of crystals.

Our study has been stimulated by the recent development of a new class of experiments, nonequilibrium phonon imaging,[2-6] which is exceptionally sensitive to features of acoustic phonons in about the central half of the Brillouin zone. A recent report[7] has used phonon imaging to study onset of dispersive effects (departure from linearity in the ω-vs-k relation) in the acoustic phonons of crystalline GaAs. In that paper, the phonon-imaging pattern for phonons with $\omega = 150$ GHz did not represent a significant departure from ordinary sound waves. However, an image taken in the range of $\omega \approx 650-750$ GHz (corresponding to phonon wavelengths on the order of 10—30 lattice constants) begins to show significant effects due to dispersion. The authors analyze their images in terms of various existing microscopic lattice-dynamical models. Their analysis illustrates the bewildering variety of con-

cepts that are used to construct microscopic lattice-vibrational pictures (rigid ions,[8] deformable ions,[8] polarizable bond charges,[9] rigid valence shells[10,11]), the formidable number of free parameters required in these models[12,13] (6—15), and, in the end, their remarkable lack of mutual agreement in the frequency range of interest (to be expected since neutron-scattering data, to which the lattice-dynamical models are fitted, show large relative error bars in this frequency regime). In this paper we will develop an alternative analytical procedure; it is expressed in a simple language, is economical of free parameters, and is appropriate for describing phonons with wavelengths in this mesoscopic (neither macroscopic nor microscopic) regime.

A short review of the mechanism of phonon imaging is in order.[3] It will permit us to introduce the concepts of standard sound theory, and to show the direction that we will extend it in the present work. In phonon imaging, a pulse of heat is generated at one point on the surface of a perfect crystal (see Fig. 1). Phonons radiate out in all directions; they are detected when they collide with the other surface of the crystal. The time of detection relative to excitation may be arranged such that only those phonons which propagate ballistically, i.e., those which suffer no scattering inside the crystal, are detected. Even finer time windowing can be arranged so that longitudinal- and transverse-phonon propagation can be distinguished. In short, the experiment is a laboratory-scale seismological experiment.

The conditions of the experiment reported in Ref. 7 are such that the heat pulse, generated by a laser, creates a local distribution of phonons with a Planck distribution corresponding to $T \approx 50$ K (the ambient temperature is around 1 K), uniformly distributed in all propagation directions **k**. However this does *not* mean that sound waves are launched with uniform intensity in all direc-
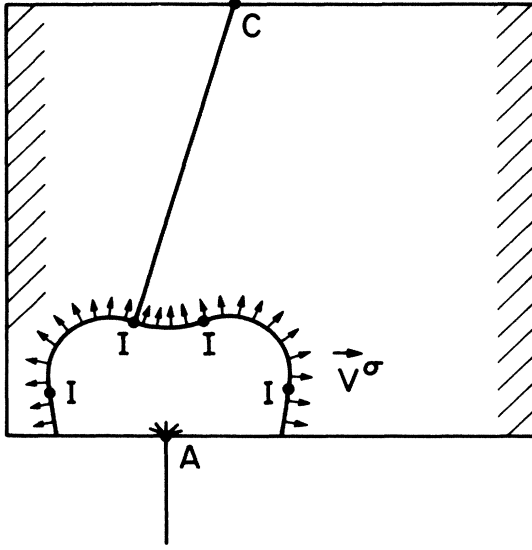
FIG. 1. An illustration of the ballistic-phonon transport process. A laser beam strikes the surface of a crystal at point A, creating a pulse of phonons. These phonons propagate along the group velocity directions $\mathbf{V}^\sigma$, which point normal to the phonon slowness (constant frequency) surface; the geometrical construction required to obtain the group velocities from the constant frequency surface is illustrated. Near points of inflection I on the slowness surface, the vectors $\mathbf{V}^\sigma$ all point in the same direction. Thus, a large concentration of acoustic energy propagates along the ray shown; a large phonon intensity arrives at C, creating an observable "caustic."

tions into the solid, because the GaAs of the experiment, unlike the mantle of the earth, is a crystal and is hence elastically anisotropic.

To understand how the cubic anisotropy of GaAs affects the energy density of emitted sound, we review the elastic description of homogeneous solids. Speaking loosely at this point, the basic hypothesis[14] is that there exists a linear relationship between stress $\sigma^{\alpha\beta}$ (an applied external force) and strain $e^{\alpha\beta}$ (a deformation):

$$\sigma^{\alpha\gamma} = c^{\alpha\beta\gamma\delta} e^{\beta\delta} \tag{1}$$

(repeated indices are summed over the three Cartesian directions). Equation (1) defines the elastic constants $c^{\alpha\beta\gamma\delta}$. If this relation is assumed to hold locally in every small volume of the solid, then the equations of motion of sound waves with propagation vector $\mathbf{k}$ may be derived:

$$\delta_{\alpha\gamma}\rho\omega_\alpha^2(\mathbf{k})v_\alpha = c^{\alpha\beta\gamma\delta}k_\beta k_\delta v_\gamma . \tag{2}$$

Here, v_α denotes the displacement eigenvector. That is, the three eigenvalues of the matrix $c^{\alpha\beta\gamma\delta}k_\beta k_\delta$ specify the three sound frequencies $\rho\omega_{1,2,3}^2$ (ρ is the mass density of the solid), and its eigenvectors specify the polarization vectors of the sound waves. In a solid with cubic symmetry, the character of the matrix $c^{\alpha\beta\gamma\delta}k_\beta k_\delta$ is such that, typically, sound waves with propagation vectors $\mathbf{k}$ of the same length but different directions have different frequencies. This means that the group velocity vector for mode σ, $V_\mu^\sigma = \partial\omega_\sigma/\partial k_\mu$, which points in the direction

$$V_\mu^\sigma = \frac{1}{2\rho}v_\alpha^\sigma\frac{\partial}{\partial k_\mu}(c^{\alpha\beta\gamma\delta}k_\beta k_\delta)v_\gamma^\sigma , \tag{3}$$

is not generally parallel to the propagation vector $\mathbf{k}$.

It is this difference between $\mathbf{V}^\sigma$ and $\mathbf{k}$ that allows phonon imaging to work, concentrating acoustic energy in particular directions in space. Consider Fig. 1. It shows a representative constant energy surface for sound (the locus of points $\mathbf{k}$ for which there are sound waves with frequency ω). The group velocity vector $\mathbf{V}$ is easily constructed as the vector normal to the constant energy surface; this vector specifies the direction in real space in which energy propagates for acoustic waves with wave vector $\mathbf{k}$. As advertised, this direction is not usually parallel to $\mathbf{k}$, as indicated in Fig. 1.

In fact, where there is a point of inflection in the constant energy surface, the flux of acoustic energy is singular in the direction corresponding to the point of inflection, creating what is known as a *caustic* on the surface of the crystal. For a three-dimensional (3D) sample, these caustics form lines of intense phonon flux which are easily detected and constitute the most important qualitative feature of the phonon-imaging experiment.

In the low-frequency, nondispersive limit, where there is a linear relationship between ω and k, $\omega = v_s(\hat{\mathbf{k}})|\mathbf{k}|$, all constant energy surfaces have the same shape. In this regime, the relationships between the directions of $\mathbf{k}$ and $\mathbf{V}$ remain constant, and the caustic pattern is independent of frequency.

At frequencies for which sound becomes dispersive, the constant energy surfaces change shape, and the caustic lines move. This is what the authors of Ref. 7 observe in GaAs, and what they attempt to explain using lattice-dynamical theory. However, the caustic lines do not move a great deal with frequency in the experiment; therefore, it makes sense to introduce dispersion perturbatively within the elastic model rather than to use an entirely new language for the phenomenon. As we will show, the correct way to introduce dispersion is to permit the elastic tensor $c^{\alpha\beta\gamma\delta}$ to take on a small momentum dependence, as represented by the power series

$$c^{\alpha\beta\gamma\delta}(\mathbf{k}) = c^{\alpha\beta\gamma\delta}(\mathbf{k}=0) + d^{\alpha\beta\gamma\delta\rho}k_\rho + f^{\alpha\beta\gamma\delta\rho\sigma}k_\rho k_\sigma + \cdots . \tag{4}$$

We find that the first three terms in this series are sufficient to describe the observed phenomenon. It is clear that Eq. (4), when substituted into Eq. (3), will have the desired effect: it will slowly change the relative directions of $\mathbf{V}$ and $\mathbf{k}$ as $|\mathbf{k}|$ increases.

The remainder of this paper will be devoted to exploring the consequences of extending the elastic model in the way suggested by Eq. (4). Our hope is to demonstrate that because of its simplicity and suitability, this model will be one of the standard tools for understanding the properties of acoustic phonons.

The remainder of this paper is organized as follows. Section II is devoted to setting up the extension of elastic theory for cubic crystals: The basic Lagrangian of the solid extended into the dispersive regime is developed in Sec. II A. In Sec. II B we derive the equations of motion

and the dynamical matrix, confirming the hypothesis of Eq. (4). In Sec. II C we discuss the symmetry properties of the new elastic coefficients defined by Eq. (4). Section II D will be of central interest to practitioners of phonon imaging; it presents numerical comparisons of the present model with previous lattice-dynamical calculations and with experiment. It is shown that our extended elastic model is compatible both with previous theory and with phonon imaging. Section III takes a different approach to the dispersive elastic model, showing how it can be derived as a limit of microscopic lattice dynamics. The discussion and conclusions of Sec. IV include a consideration of various approximation schemes for the dispersive elastic model (e.g., the isotropic approximation), suggestions for cheap and dirty ways of fitting experimental results to the phenomenological model, and a consideration of the

application of this model to other physical properties (e.g., specific heat).

II. PHENOMENOLOGICAL THEORY

A. The elastic Lagrangian

As discussed above, the strategy of the present work is to describe dispersive effects in crystals using the same language as we normally use to describe the ordinary elastic response of solids. Many excellent discussions of the continuum theory of elasticity may be found in texts;[14,1,15-19] in this subsection we show how this theory can be extended to describe the dispersive sound regime. The form of the Lagrangian density for a solid (kinetic minus potential energy) within the continuum description[17,20] is

$$L = \tfrac{1}{2}\rho \dot{u}_\alpha \dot{u}_\alpha - \tfrac{1}{2}C^{\alpha\beta\gamma\delta}\frac{\partial u_\alpha}{\partial x_\beta}\frac{\partial u_\gamma}{\partial x_\delta} - D^{\alpha\beta\gamma\delta\rho}\frac{\partial u_\alpha}{\partial x_\beta}\frac{\partial^2 u_\gamma}{\partial x_\delta \partial x_\rho} - F_1^{\alpha\beta\gamma\delta\rho\sigma}\frac{\partial u_\alpha}{\partial x_\beta}\frac{\partial^3 u_\gamma}{\partial x_\delta \partial x_\rho \partial x_\sigma}$$

$$- F_2^{\alpha\beta\gamma\delta\rho\sigma}\frac{\partial^2 u_\alpha}{\partial x_\beta \partial x_\delta}\frac{\partial^2 u_\gamma}{\partial x_\rho \partial x_\sigma} - \cdots - A^{\alpha\beta\gamma\delta\rho\sigma}\frac{\partial u_\alpha}{\partial x_\beta}\frac{\partial u_\gamma}{\partial x_\delta}\frac{\partial u_\rho}{\partial x_\sigma} - \cdots .$$

(5)

Here, the Greek indices denote the Cartesian directions (x,y,z); repeated indices are summed. The first term is the kinetic energy, where ρ is the mass density of the solid. The remaining terms constitute the potential energy of the solid. The potential energy depends only on the derivatives of u, not upon u itself. This is shown by requiring that the energy remain invariant under a uniform translation of the solid: $u(r) \rightarrow u(r)+u_0$. In addition, the potential energy has no terms linear in $\partial u/\partial x$; this is usually explained by saying that the solid is in equilibrium and under no external stress. We will see in Sec. III in our discussion of the relationship of this Lagrangian to the microscopic description of lattice vibrations that the absence of a term linear in $\partial u/\partial x$ is appropriate and justifiable. At the same time, we will see that it is also appropriate for terms linear in $\partial^2 u/\partial x^2, \partial^3 u/\partial x^3$, etc. to be absent. In the literature, this has been termed "lack of hyperstresses."[20]

Finally, let us discuss the terms which *do* appear. The second term dominates the potential energy; it describes the long-wavelength elastic excitations of the solid—the lowest-order description of sound. The coefficients $C^{\alpha\beta\gamma\delta}$ are what are normally called the elastic constants of the solid; the factor $\tfrac{1}{2}$ is conventional.[14] To distinguish them from the coefficients of the remaining terms in this potential energy, I will normally refer to them as the nondispersive elastic constants.

This dominant term is quadratic in the u's (i.e., harmonic), and it contains two spatial gradients. This term can be generalized in at least two ways which satisfy the above-mentioned requirement of locality in the potential

energy. The first, which is the main subject of this paper, increases the number of spatial derivatives without increasing the number of u's. The third through fifth terms in Eq. (5) are the most important which arise from this generalization. There is just one distinct type of term of third order in the spatial gradients, and there are two of fourth order. The coefficients in front of these terms, D, F_1, and F_2, we term "dispersive elastic constants," since as we show below, they describe changes in the sound velocities as the wavelength decreases.

There has been some previous discussion in the literature of the third-order term,[21,20,22] principally because it describes the phenomenon observed in some solids known as "acoustic activity," which, by analogy to optical activity, is the rotation of the plane of polarization of a transverse sound wave upon passage through a material. The fourth-order terms have been referred to in only the most general way[20,22,18,23,21] in the previous literature. For the present study, both the third- and fourth-order terms are essential for describing the observed dispersion in GaAs (see Sec. II D). Therefore we will discuss both the D's and F's in detail.

It is very important to recognize that this type of generalization to the elastic potential energy stays within the realm of the harmonic approximation, since the Lagrangian remains quadratic in the displacement field u. Thus, with these terms included, the eigenstates of the equation of motion (see below) are still the ordinary plane waves describing sound; the effect of these terms is only felt in the eigenvalues, which deviate from the linear ω-vs-k relationship. It is for this reason that it is appropriate to call

the D's and F's dispersive elastic constants.

Of course, it is possible to generalize the elastic potential energy beyond Hooke's law as well. The last term in Eq. (5) illustrates the simplest possible extension along these lines. This term contains three u's, and as a consequence the coefficients in front of them, the A's, may be called the "anharmonic elastic constants." They describe such phenomena as the pressure dependence of the elastic response and the decay rate of phonons;[24,25] since these phenomena are not important in the phonon-imaging experiments to be described here, we will drop these and all other higher-order terms.

B. The elastic equations of motion and the dynamical matrix

In this subsection we review the manipulations which are required to obtain the equations of motion and the dynamical matrix from the continuum Lagrangian equation (5). While this simply involves an application of the standard prescriptions of classical mechanics, we will go through them here since they are important for understanding some of the symmetry properties of the D and F coefficients.

We wish the variations of the action to be stationary:

$$\delta A = \delta \left[\int L\, d\mathbf{x}\, dt \right] = 0 , \tag{6}$$

$$\delta A = \int d\mathbf{x} \int dt \{ \rho \delta \ddot{u}_\alpha \dot{u}_\alpha - \tfrac{1}{2} C^{\alpha\beta\gamma\delta}[\partial_\beta(\delta u_\alpha)\partial_\delta u_\gamma + \partial_\delta(\delta u_\gamma)\partial_\beta u_\alpha] - D^{\alpha\beta\gamma\delta\rho}[\partial_\beta(\delta u_\alpha)\partial_{\delta\rho} u_\gamma + \partial_\beta u_\alpha \partial_{\delta\rho}(\delta u_\gamma)]$$
$$- F_1^{\alpha\beta\gamma\delta\rho\sigma}[\partial_\beta(\delta u_\alpha)\partial_{\delta\rho\sigma} u_\gamma + \partial_{\delta\rho\sigma}(\delta u_\gamma)\partial_\beta u_\alpha] - F_2^{\alpha\beta\gamma\delta\rho\sigma}[\partial_{\beta\delta}(\delta u_\alpha)\partial_{\rho\sigma} u_\gamma + \partial_{\rho\sigma}(\delta u_\gamma)\partial_{\beta\delta} u_\alpha] \} . \tag{7}$$

Here,

$$\partial_\alpha \equiv \frac{\partial}{\partial x_\alpha}, \quad \partial_{\alpha\beta} \equiv \frac{\partial^2}{\partial x_\alpha\, \partial x_\beta}, \quad \text{and} \quad \partial_{\alpha\beta\gamma} \equiv \frac{\partial^3}{\partial x_\alpha\, \partial x_\beta\, \partial x_\gamma} .$$

As usual, we now integrate by parts[17] to obtain the elastic equations of motion:

$$\rho \ddot{u}_\alpha = c^{\alpha\beta\gamma\delta}\partial_{\beta\delta} u_\gamma + d^{\alpha\beta\gamma\delta\rho}\partial_{\beta\delta\rho} u_\gamma + f^{\alpha\beta\gamma\delta\rho\sigma}\partial_{\beta\delta\rho\sigma} u_\gamma . \tag{8}$$

Here we have introduced new "dynamic" elastic constants $c^{\alpha\beta\gamma\delta}$, $d^{\alpha\beta\gamma\delta\rho}$, and $f^{\alpha\beta\gamma\delta\rho\sigma}$. These are related to the static elastic constants by the appropriate symmetrizations or antisymmetrizations of indices:[14]

$$c^{\alpha\beta\gamma\delta} = \mathrm{sym}_{(\alpha,\gamma)}\mathrm{sym}_{(\beta,\delta)} C^{\alpha\beta\gamma\delta} , \tag{9}$$

$$d^{\alpha\beta\gamma\delta\rho} = \mathrm{asym}_{(\alpha,\gamma)}\mathrm{sym}_{(\beta,\delta,\rho)} D^{\alpha\beta\gamma\delta\rho} , \tag{10}$$

$$f_{\alpha\beta\gamma\delta\rho\sigma} = \mathrm{sym}_{(\alpha,\gamma)}\mathrm{sym}_{(\beta,\delta,\rho,\sigma)}(F_1^{\alpha\beta\gamma\delta\rho\sigma} - F_2^{\alpha\beta\gamma\delta\rho\sigma}) . \tag{11}$$

The symmetrization (antisymmetrization) with respect to the α and γ indices arises because of the different terms generated by integration by parts. Equation (10) contains an antisymmetrization because one term requires a single integration by parts, and the other a double integration by parts; these terms enter with different signs. The symmetrization with respect to the remaining indices is simply a reflection of the permutation symmetry of multiple derivatives, e.g., $\partial_{\beta\delta} = \partial_{\delta\beta}$.

Equations (9)–(11) imply that information is lost upon going from the static potential energy to the dynamic equations of motion; in other words, that dynamic measurements of the elastic response cannot extract as much information as a static-stress experiment. This point has been considered previously by Lax,[14,18] who shows that despite the symmetrization in Eq. (9) no information is lost in the ordinary elastic constants, and that Eq. (5) can be inverted for $C^{\alpha\beta\gamma\delta}$:

$$C^{\alpha\beta\gamma\delta} = c^{\alpha\beta\gamma\delta} + c^{\beta\alpha\gamma\delta} - c^{\beta\gamma\alpha\delta} . \tag{12}$$

However, his analysis does not apply to the dispersive elastic constants, and Eqs. (10) and (11) are genuinely not invertible. [This is obvious for Eq. (11), where F_1 and F_2, which are the coefficients of two distinct static derivatives, are combined.] Since we are concerned with describing phonon propagation, which is a dynamic experiment, we will only consider the elastic constants $d^{\alpha\beta\gamma\delta\rho}$ and $f^{\alpha\beta\gamma\delta\rho\sigma}$. It should be kept in mind, however, that a static phenomenon which shows dispersive effects (corrections to the elastic energy of a dislocation, or domain wall shape[26,27]) will be described by the D's, F_1's, and F_2's of Eq. (5) rather than the d's and f's.

We now complete the derivation of the dynamical equations for sound in a perfect crystal. Since the system has translational invariance, Eq. (8) is solved by plane waves: $u_\alpha = \epsilon_\alpha e^{i\omega t} e^{i\mathbf{k}\cdot\mathbf{r}}$. The secular equation determining the dispersion law $\omega(\mathbf{k})$ and the polarization vectors ϵ_α is then

$$\rho\omega^2 \epsilon_\alpha = (c^{\alpha\beta\gamma\delta} k_\beta k_\delta + id^{\alpha\beta\gamma\delta\rho} k_\beta k_\delta k_\rho$$
$$+ f^{\alpha\beta\gamma\delta\rho\sigma} k_\beta k_\delta k_\rho k_\sigma)\epsilon_\gamma . \tag{13}$$

To conclude this subsection, we show that in order for Eq. (13) to represent a consistent lowest-order dispersive elastic theory, both the third and fourth-order terms in the dynamical matrix must be included; both terms contribute to the eigenfrequencies to the same order. We first imagine diagonalizing the dynamical matrix equation (13) to order k^2. It is known[14] that this can be accomplished by a real orthogonal transformation R_{ij}, since the matrix $c^{\alpha\beta\gamma\delta} k_\beta k_\delta$ is real and symmetric, and that R_{ij} depends on the direction but not the magnitude of $\mathbf{k}$. If we apply this orthogonal transformation to the full dynamical matrix in Eq. (13), we obtain

$$\rho\omega^2 \epsilon_\alpha^R = (\rho v_\alpha^2 k^2 \delta_{\alpha\gamma} + id_R^{\alpha\beta\gamma\delta\rho} k_\beta k_\delta k_\rho$$
$$+ f_R^{\alpha\beta\gamma\delta\rho\sigma} k_\beta k_\delta k_\rho k_\sigma)\epsilon_\alpha^R . \tag{14}$$

We have denoted the rotated eigenvectors as $\epsilon^R_\gamma \equiv R_{\gamma j} \epsilon_j$, the rotated third- and fourth-order terms as

$$id^{\alpha\beta\gamma\delta\rho}_R k_\beta k_\delta k_\rho \equiv iR_{\alpha i} d^{i\beta j\delta\rho}_R R_{\alpha j} k_\beta k_\delta k_\rho$$

and

$$f^{\alpha\beta\gamma\delta\rho\sigma}_R k_\beta k_\delta k_\rho k_\sigma \equiv R_{\alpha i} f^{i\beta j\delta\rho\sigma} R_{\alpha j} k_\beta k_\delta k_\rho k_\sigma \; ,$$

and the eigenvalues of the ordinary elastic dynamical matrix are denoted $\rho v^2_\alpha k^2$, where v_α is the sound velocity of the αth branch. We will now compute the corrections to these elastic eigenvalues by perturbation theory. Consider first the case where $\mathbf{k}$ points in a general direction, so that the v_α's are all different; in this case we can apply ordinary nondegenerate perturbation theory. The diagonal elements of the $f^{\alpha\beta\gamma\delta\rho\sigma} k_\beta k_\delta k_\rho k_\sigma$ matrix give a first-order contribution, changing the eigenvalue to order k^4. The matrix $id^{\alpha\beta\gamma\delta\rho}_R k_\beta k_\delta k_\rho$ has zero diagonal elements, since it is a pure imaginary Hermitian matrix; it makes its contribution in second-order perturbation theory. A typical term is

$$\frac{\mid d^{\alpha\beta\gamma\delta\rho}_R k_\beta k_\delta k_\rho \mid^2}{\rho(v^2_\alpha - v^2_\beta)k^2} \; , \tag{15}$$

which also is of order k^4. The d and f contributions are therefore equally important. This perturbative result can only be incorrect when $v_\alpha = v_\beta$, which will occur along high symmetry axes (the [100] and [111] axes in cubic crystals). In this case Eq. (15) diverges, signaling that the d contribution is lower order in k. This will be precisely the case in which the solid exhibits acoustic activity (rotation of the plane of polarization of transverse sound) and linear dispersion of the TO phonons near $k = 0$.[18,23] However, in crystals such as GaAs with T_d symmetry, the numerator of Eq. (15) also vanishes along the high-symmetry axes, so that acoustic activity does not occur and the contributions of the d tensor remains of order k^4. Reference 21 gives a detailed discussion of the group-theoretic origin of this result.

C. Symmetries of the continuum elastic theory

In the present elastic theory, the c, d, and f elastic constants alone completely determine acoustic dispersion, except near the Brillouin-zone boundaries. Therefore, a great deal of good can be done by using symmetry to reduce the number of independent components of the d and f tensors to their fewest number. This will be accomplished in the present subsection.

Time-reversal invariance applies to any paramagnetic material in the absence of an applied magnetic field. It requires that the Hamiltonian of a system be real in the space representation (Ref. 14, p. 276); this also implies that the potential and the Lagrangian must be real in the space representation. We conclude from Eq. (5) that all the elastic constants C, D, F_1, and F_2 are real, so that c, d, and f of Eq. (13) are also real. This, along with the symmetries of c, d, and f with respect to interchange of α and γ, guarantees that the eigenvalue problem equation (13) is Hermitian and the eigenvalues ω^2 are real.

The only other general symmetry constraint which we can impose is that the action [Eq. (6)] be invariant under rotations of the coordinate system. This simply requires that $c^{\alpha\beta\gamma\delta}$, $d^{\alpha\beta\gamma\delta\rho}$, and $f^{\alpha\beta\gamma\delta\rho\sigma}$ transform under rotation as tensors of ranks four, five, and six, respectively.[28] In more group-theoretic language, it says that they transform like direct products of the vector representation of the rotation group.

To make further progress we must invoke the specific point-group symmetry of the solid under study. In the present case we are interested in GaAs, which has a zinc-blende crystal structure and therefore the point-group symmetry T_d ($\overline{4}3m$). We wish to know how many independent components the elastic tensors contain if they are required to remain invariant under all the operations of the group T_d. The reader may recall the answer for the ordinary elastic constant tensor in solids with cubic symmetry:[15] there are three independent elastic constants, which may be taken as c^{1111}, c^{1212}, and c^{1122} (Ref. 29). It is worth reviewing how this statement is proved using group theory. As mentioned above, c transforms according to a representation involving the direct product of four vector representations. When the tensor index permutation symmetries are taken into account, the representation to which c belongs is

$$R_c = [[V^2]^2] \; , \tag{16}$$

using a notation of Refs. 30 and 31 in which V stands for the vector representation and the square brackets denote the symmetric part. The innermost symmetrization in Eq. (16) results from the fact that $c^{\alpha\beta\gamma\delta}$ is symmetric with respect to the interchange of α and γ or β and δ. The outer symmetrization in Eq. (16) results from the symmetry of $c^{\alpha\beta\gamma\delta}$ with respect to the double interchange of α, γ with β, δ. This property, which has not been mentioned above, results from the fact that a strain applied to the solid corresponding to a pure rotation costs no energy.[16] This symmetry will not manifest itself in $d^{\alpha\beta\gamma\delta\rho}$ or $f^{\alpha\beta\gamma\delta\rho\sigma}$.

Returning to the main discussion, the question of how many independent components the elastic tensor $c^{\alpha\beta\gamma\delta}$ has is equivalent to the group-theoretic question: How many times does the reducible representation $R_c = [[V^2]^2]$ contain the identity representation of T_d?[32] The general formula for the characters of this representation, which may be found by the successive application of formulas derived by Lyubarskii,[28] is

$$\chi_{R_c}(g) = \tfrac{1}{4}\chi_V(g^4) + \tfrac{3}{8}[\chi_V(g^2)]^2$$
$$+ \tfrac{1}{4}\chi_V(g^2)[\chi_V(g)]^2 + \tfrac{1}{8}[\chi_V(g)]^4 \; . \tag{17}$$

Table I shows the characters of R_c for the group T_d. Using the standard formula for the decomposition of reducible representations,[33] we find that, as expected, R_c contains the identity representation exactly three times, corresponding to the three nondispersive elastic constants in cubic materials.

It is a simple matter to apply this formalism to $d^{\alpha\beta\gamma\delta\rho}$ and $f^{\alpha\beta\gamma\delta\rho\sigma}$. $d^{\alpha\beta\gamma\delta\rho}$ transforms according to the representation $R_d = \{V^2\}[V^3]$ (curly brackets denote the antisymmetric part), and $f^{\alpha\beta\gamma\delta\rho\sigma}$ transforms according to $R_f = [V^2][V^4]$. Again following Lyubarskii,[28] the characters of these representations are

TABLE I. Character table of T_d, the point group of GaAs (following Ref. 33). Also shown are the reducible representations relevant to the symmetry analysis of elastic constants.

T_d (43m)	E	$8C_3$	$3C_2$	$6\sigma_d$	$6S_4$
A_1	1	1	1	1	1
A_2	1	1	1	-1	1
E	2	-1	2	0	0
T_1	3	0	-1	-1	1
$V = T_2$	3	0	-1	1	-1
$R_c = [[V^2]^2]$	21	0	5	5	1
$R_d = \{V^2\}[V^3]$	30	0	2	-2	0
$R_f = [V^2][V^4]$	90	0	6	6	0

$$\chi_{R_d}(g) = [\tfrac{1}{2}\chi_V^2(g) - \tfrac{1}{2}\chi_V(g^2)][\tfrac{1}{3}\chi_V(g^3) + \tfrac{1}{2}\chi_V(g^2)\chi_V(g) + \tfrac{1}{6}\chi_V^3(g)] , \tag{18}$$

$$\chi_{R_f}(g) = [\tfrac{1}{2}\chi_V^2(g) + \tfrac{1}{2}\chi_V(g^2)][\tfrac{1}{4}\chi_V(g^4) + \tfrac{1}{3}\chi_V(g^3)\chi_V(g) + \tfrac{1}{8}\chi_V^2(g^2) + \tfrac{1}{4}\chi_V(g^2)\chi_V^2(g) + \tfrac{1}{24}\chi_V^4(g)] \tag{19}$$

and they are given in Table I. When these representations are reduced, we find that there is one distinct component of the $d^{\alpha\beta\gamma\delta\rho}$ tensor and six distinct components of the $f^{\alpha\beta\gamma\delta\rho\sigma}$ tensor.

In order to complete the construction of the dynamical matrix, we must know explicitly the irreducible basis tensors of c, d, and f. (This is analogous to the need to find the irreducible spherical tensors in a spherically symmetric scattering problem.[34]) There are irreducible tensors corresponding to each of the ordinary elastic constants c^{1111}, c^{1212}, and c^{1122} (Ref. 29); a typical element of the tensor corresponding to c^{1212}, for example, is $\hat{x}_1\hat{x}_2\hat{x}_1\hat{x}_2$.[15] The other elements are obtained by suitable permutations and relabelings of the tensor indices.

There is a single basis tensor for the first dispersive elastic tensor d. We will call the elastic constant associated with the irreducible basis tensor d^{12223}, which is indicative of how the basis tensor is constructed: a representa-tive element of the tensor is $\hat{x}_1\hat{x}_2\hat{x}_2\hat{x}_2\hat{x}_3$, with other elements obtained by the same permutations as before, with the sign changes which are dictated by the antisymmetry of $d^{\alpha\beta\gamma\delta\rho}$ in α,γ. The complete basis tensor contains 36 terms.

For the other dispersive elastic tensor f, we call the six elastic constants associated with the six irreducible basis tensors f^{111111}, f^{122122}, f^{112222}, f^{122133}, f^{112233}, and f^{211222}. Again, the nomenclature is used to indicate a typical term in the basis tensor.

Finally, we can state the main result of this subsection, which is the equations of motion (13) rewritten with the minimum number of free phenomenological constants as determined by symmetry. If we rewrite Eq. (13) as

$$\rho\omega^2\epsilon_i = D_{ij}\epsilon_j , \tag{20}$$

then the dynamical matrix D_{ij} is

$$D_{ii} = c^{1111}k_i^2 + c^{1212}(k_j^2 + k_k^2) + f^{111111}k_i^4 + f^{122122}(k_j^4 + k_k^4) + 6f^{122133}k_j^2k_k^2 + 6f^{211222}(k_i^2k_j^2 + k_i^2k_k^2) , \tag{21a}$$

$$D_{ij} = 2c^{1122}k_ik_j + 3id^{12223}(k_i^2k_k - k_j^2k_k) + 4f^{112222}(k_ik_j^3 + k_i^3k_j) + 12f^{112233}(k_ik_jk_k^2) . \tag{21b}$$

Here the subscripts i, j, and k denote Cartesian directions, and we require $i \neq j$, $j \neq k$, and $k \neq i$. Equation (21) gives the dynamical matrix which we use to model dispersive phonon propagation in GaAs.

D. Comparison with lattice dynamics and phonon imaging

We would like to get a little ahead of ourselves at this point and quote the final results of the analysis we have performed to extract the values of the dispersive elastic constants from previous lattice-dynamical calculations for GaAs. An explanation of how we extract these constants will require a detailed digression on microscopic lattice-dynamical theory, which we will defer until the next section.

Table II gives the values of the third- and fourth-order dispersive constants, as well as the ordinary elastic constants, for three lattice-dynamics models[12,13] of GaAs.

(1) A shell model.[10,11] The ions in the crystal are thought of as surrounded by massless spherical shells, and the ions and shells interact via short-range first- and second-neighbor forces (i.e., they are connected by springs) as well as via a Coulomb interaction. There are 14 free

TABLE II. Nondispersive and dispersive elastic constants for the lattice-dynamical models for GaAs as described in the text.

	Shell model	Dipole model	Rigid-ion model	Valence-shell model	Expt.
c^{1111} (dyn/cm^2) ($\times 10^{12}$)	1.20	1.19	1.19	1.20	1.18
c^{1122} ($\times 10^{12}$)	0.57	0.57	0.56	0.61	0.56
c^{1212} ($\times 10^{12}$)	0.60	0.60	0.59	0.46	0.59
d^{12223} (dyn/cm) ($\times 10^3$)	-1.8	-4.1	-1.8	$+1.4$	
f^{111111} (dyn) ($\times 10^{-4}$)	-0.26	-0.40	-0.41	-0.43	
f^{122122} ($\times 10^{-4}$)	-2.04	-1.05	-0.90	-0.79	
f^{112222} ($\times 10^{-4}$)	-0.16	-0.03	-0.04	-0.21	
f^{122133} ($\times 10^{-4}$)	-0.48	-0.45	-0.33	-0.07	
f^{112233} ($\times 10^{-4}$)	$+0.19$	$+0.08$	$+0.05$	$+0.13$	
f^{211222} ($\times 10^{-4}$)	-0.24	-0.22	-0.25	-0.25	

parameters in this model, which [like the constants in models (2)–(4) below] are determined by fitting to the ordinary elastic constants[35] and to neutron-scattering data.[36]

(2) A deformation-dipole model.[8] In this model a dynamic dipole moment is taken to point along the instantaneous direction of each near-neighbor bond. The forces between these dipoles, along with ion-ion Coulomb interactions and short-range mechanical forces, define the model, which has 15 free parameters.

(3) A rigid-ion model.[8] This is a specialization of the deformation-dipole model above with the bond polarizabilities set equal to zero. Eleven free parameters remain.

(4) A valence-force-field model.[10,11] A reparametrization of the shell model introduced in (1) above.

Although the dynamical variables of these models consist of microscopic atomic displacements, the various interatomic interactions of these models are no more calculable from first principles than the elastic constants of the present model. This is reflected in the wide variation in the values of the dispersive elastic constants which are obtained from fitting to these models. (The ordinary elastic constants are in good agreement simply because they have been used to constrain the parameters of these models.) Given the variability of the d's and f's obtained, it is not surprising that these models give very different predictions for the ballistic-phonon—propagation patterns for GaAs observed in the dispersive regime[7] around $\omega \approx 1$ THz. Reference 7 showed that the shell model gives a better overall prediction than the other lattice-dynamical calculations for the focusing pattern; therefore, the numbers in the first column of Table II are probably closest to the true elastic constants of GaAs. Refinement of these estimates for the dispersive elastic constants by direct comparison with experiment would be appropriate, but has not been attempted in the present work.

There exists a very elementary dimensional argument for why the values of the dispersive elastic constants fall within the orders of magnitude which they show in Table II; that is, why the values for $d^{\alpha\beta\gamma\delta\rho}$ fall in the range 10^3 dyn/cm, and those of $f^{\alpha\beta\gamma\delta\rho}$ in the range 10^{-4} dyn. $d^{\alpha\beta\sigma\delta\rho}$ has the dimensions of an ordinary elastic constant times a length. The only characteristic length scale in the solid is the interatomic spacing, on the order of 10^{-8} cm. Since $c^{\alpha\beta\gamma\delta}$ is on the order 10^{+12} dyn/cm^2 (which itself

can be made plausible by a dimensional argument relating c to a cohesive energy per unit volume), we expect d's on the order of $(10^{12}$ dyn/cm$^2)(10^{-8}$ cm$) = 10^4$ dyn/cm, which is about right. In exactly the same way we argue that $f^{\alpha\beta\gamma\delta\rho\sigma}$ should typically be c times an interatomic length squared: $f = (10^{12}$ dyn/cm$^2)(10^{-8}$ cm$)^2 \approx 10^{-4}$ dyn.

As mentioned above, GaAs should not exhibit acoustic activity; however, the presence of a nonzero d elastic coefficient may be measurable by sonic probes. In the [110] direction, one of the transverse modes is slightly mixed with the longitudinal wave by the d elastic constant (and vice versa), leading to modes with elliptical polarization. The ratio of the minor to major axes of this ellipse for the (quasi)longitudinal mode grows linearly with frequency, and is given by

$$\frac{3\sqrt{2}d^{12223}}{c^{1111}+c^{1122}}\left[\frac{\rho}{c^{1212}}\right]^{1/2}\omega .$$

Thus, if a purely transverse ultrasonic wave is launched into the crystal in the [110] direction, both a (quasi)transverse and a (quasi)longitudinal wave will be propagated with an amplitude ratio given by this expression. At $\nu = 250$ MHz, this amplitude ratio would be 10^{-3}, which may be detectable by present-day ultrasonic techniques.

To show that the inclusion of higher-order dispersive elastic constants is indeed crucial for describing GaAs lattice dynamics in the THz regime, as well as to illustrate some of the general behavior of the elastic model defined by Eqs. (13) and (20) and (21), we show in Fig. 2 phonon dispersions along several high-symmetry and one general direction in reciprocal space for GaAs as obtained by the shell model, compared with ω vs $\mathbf{k}$ obtained from the dispersive theory equations (20) and (21), and with the purely nondispersive sound theory (i.e., $d^{\alpha\beta\gamma\delta\rho} = f^{\alpha\beta\gamma\delta\rho\sigma} = 0$). As these figures show, the dispersive elastic theory is capable of reproducing ω vs $\mathbf{k}$ for this model (and so presumably for experiment as well) up to frequencies on the order of and larger than 1 THz. By contrast, the nondispersive theory does not follow the dispersions into this frequency regime nearly as well; the inclusion of $d^{\alpha\beta\gamma\delta\rho}$ and $f^{\alpha\beta\gamma\delta\rho\sigma}$ is clearly not superfluous. Similar re-
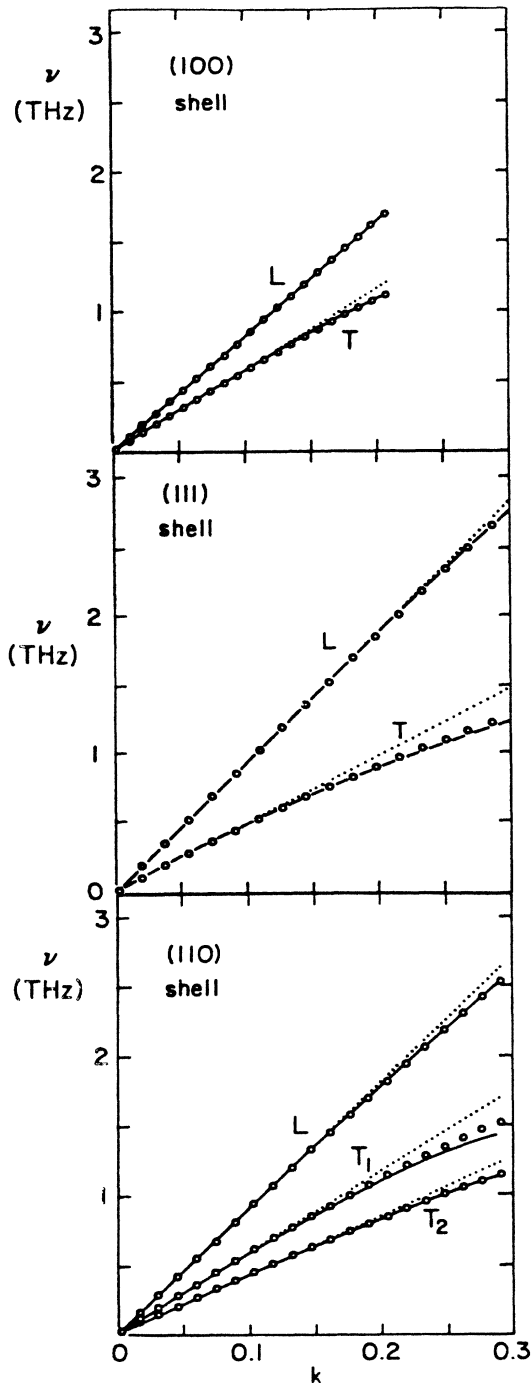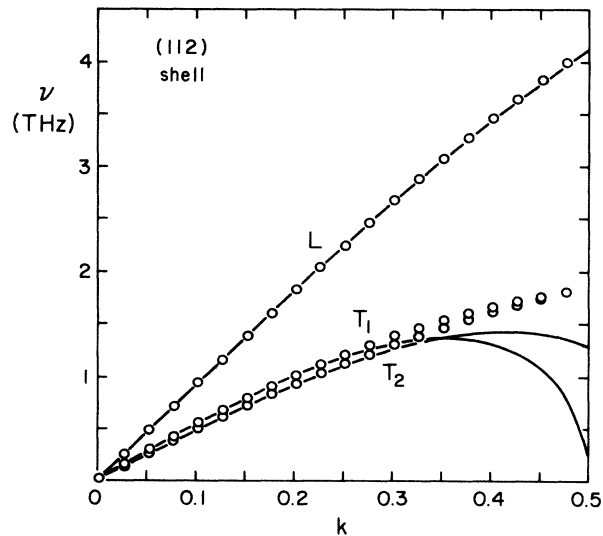
FIG. 3. Comparison of the shell-model lattice-dynamical calculation (open circles) with the present dispersive elastic model (curves) for elastic parameters as given in Table II for dispersions in a more general direction, [112]. Larger wave vectors are shown; this illustrates how the elastic model eventually fails at large energy. The failure for the shell model is more extreme than for the others because of the larger elastic constants which correspond to the shell model (see Table II).

sults have been obtained for the other lattice-dynamical models. The reason that the dispersive model works well is that, as we will show in detail below, the dispersive elastic theory represents a Taylor-series expansion of the real microscopic lattice dynamics up to fourth order in wave vector k.[37] Of course, like any truncated Taylor series, the approximation must eventually fail; evidence of the beginning of this failure may be seen in Fig. 3 in the plot going in the [112] direction, for which we have gone to somewhat larger $|k|$ than for the other directions. Since most of the dispersive corrections to the dynamical matrix equation (17) are negative (i.e., generally $d, f < 0$, as they must be to reproduce the general phenomenon that sound velocities decrease with increasing k), the dispersions eventually turn downward (unphysically). However, this does not happen until k has reached a large fraction of the Brillouin-zone dimensions.

III. THE DISPERSIVE ELASTIC MODEL

A. Relationship to microscopic lattice dynamics

We now return to the discussion which we deferred earlier of how the connection is made between our dispersive elastic model and a traditional microscopic lattice-dynamics description of vibrations in solids.

My primary source material for this subject has been the book of Lax,[14] which contains informative discussions of many of the subjects treated here. Aside from the practical result of this subsection—providing a means of extracting the dispersive elastic constants from a microscopic model—it is of some fundamental significance to show that the phenomenological model of Eq. (4) can be

FIG. 2. Comparison of the shell-model lattice-dynamical calculation (open circles) with the present dispersive elastic model (solid curves) and with ordinary nondispersive sound theory (dotted curves) for the phonon dispersions of GaAs along the [100], [111], and [110] directions. The elastic parameters are found in Table II. k is in units of $2\pi/a$, where $a = 5.65$ Å is the cubic lattice constant for GaAs. Phonon frequencies are in THz. Agreement between the dispersive continuum and lattice models is almost perfect through the experimentally important regime up to 1 THz. (This is true for other phonon-propagation directions and lattice models.) The nondispersive model fails noticeably for the transverse branches near $\nu = 1$ THz.

developed as a systematic approximation to the actual microscopic dynamics of the solid.

The microscopic action of the solid in the harmonic approximation is (Ref. 14, pp. 326 and 327)

$$A = \sum_{\alpha,i,\beta,j} \int dt \left[\tfrac{1}{2} M_\alpha (\dot{w}^{\alpha i})^2 \delta_{\alpha\beta} \delta_{ij} - \tfrac{1}{2} \sum_{\mu,\nu} w_\mu^{\alpha i} K_{\mu\nu}^{\alpha i \beta j} w_\nu^{\beta j} \right] .$$

(22)

Here, the fundamental dynamical variable $w_\mu^{\alpha i}$, rather than being a continuum displacement field, is the displacement from equilibrium of atom α in unit cell i in direction μ. M_α is the mass of the αth atom, and $K_{\mu\nu}^{\alpha i \beta j}$ is the "matrix of force constants" connecting atom α (unit cell i) with atom β (unit cell j). It is certainly not clear *a priori* that these microscopic displacement vectors can be averaged suitably to give the continuum displacement field of Eq. (4). The identification can be made, however, by comparing the equations of motion.

To obtain the equations of motion analogous to Eq. (13) from Eq. (24), we find the variational minimum of the action, then exploit the translational symmetry of the solid to write the eigenvector solutions as plane waves:

$$w_\mu^{\alpha i} = \sqrt{M_\alpha} b_\mu^\alpha(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{X}^{\alpha i}} e^{i\omega t} .$$

(23)

$\mathbf{X}^{\alpha i}$ is the equilibrium position of atom α in unit cell i. [This is nearly the same as Eq. (11.2.6) in Ref. 14, except with a different mass factor.] The resulting equations of motion are

$$\rho \omega^2(\mathbf{k}) b_\mu^\alpha(\mathbf{k}) = \sum_{\beta,\nu} R_{\mu\nu}^{\alpha\beta}(\mathbf{k}) b_\nu^\beta(\mathbf{k}) ,$$

(24)

with

$$R_{\mu\nu}^{\alpha\beta}(\mathbf{k}) \equiv \frac{\rho}{\sqrt{M_\alpha M_\beta}} \sum_i K_{\mu\nu}^{\alpha i \beta j} e^{i\mathbf{k}\cdot(\mathbf{X}^{\beta j} - \mathbf{X}^{\alpha i})} .$$

(25)

[cf. Eq. (11.2.8b), Ref. 14.] Superficially, Eqs. (24) and (13) are identical; however, while Eq. (13) contains three equations of motion, Eq. (24) contains $3N$, N being the number of atoms per unit cell. If $N = 1$, then indeed the elastic constants of Eq. (13) can be read off trivially from a power-series expansion of Eq. (25). However, for $N > 1$, (and, in particular, $N = 2$, the case of present concern) the identification is considerably more subtle.

We begin this identification by applying a unitary transformation $\underline{U}$ to the dynamical matrix $\underline{R}$ which block-diagonalizes it for $\mathbf{k} = 0$:

$$\underline{R}^U(\mathbf{k}=0) = \underline{U}^T \underline{R}(\mathbf{k}=0)\underline{U} = \begin{bmatrix} \underline{D}^a & \underline{0}_{3\times3} \\ \underline{0}_{3\times3} & \underline{D}^o \end{bmatrix} .$$

(26)

The matrix $\underline{U}$ consists of the $\mathbf{k} = 0$ eigenvectors, which for a solid with a zinc-blende structure can be written down exactly by symmetry. $\underline{D}^a$ and $\underline{D}^o$ are the acoustic and optic contributions to the dynamical matrix. The 3×3 subblock $\underline{D}^a$ in Eq. (26) is identified with the elastic dynamical matrix of Eq. (9) at $\mathbf{k} = 0$. Of course, this identification is rather trivial $[\underline{D}^a(\mathbf{k}=0) = \underline{0}_{3\times3}]$, but it provides a footing upon which to consider $\mathbf{k} \neq 0$. For, in the basis of Eq. (26), the coupling of the acoustic and op-

tic blocks remains small (of order k) at small $\mathbf{k}$:

$$\underline{R}^U(\mathbf{k}) = \underline{U}^T \underline{R}(\mathbf{k})\underline{U} = \begin{bmatrix} \underline{D}^a + Q(k^2)\mathbb{1}_{3\times3} & Q(\mathbf{k})\mathbb{1}_{3\times3} \\ Q(\mathbf{k})\mathbb{1}_{3\times3} & \underline{D}^o + Q(\mathbf{k})\mathbb{1}_{3\times3} \end{bmatrix} .$$

(27)

Equation (27) assumes that the dynamical matrix is analytic in $\mathbf{k}$ in the vicinity of $\mathbf{k} = 0$. This is actually not the case for polar crystals, in which[1] the dynamical matrix $\underline{D}(\mathbf{k})$ contains terms of the form $k_\alpha k_\beta / k^2$ which are related to the development of a long-range electric field accompanying strains of the solid, and which are responsible for the LO-TO splitting at $\mathbf{k} = 0$. However, the effect of this nonanalyticity is small for the acoustic dispersion of GaAs, and it will be ignored in the development which follows. (In the Appendix we touch upon the modifications of the derivation of the elastic theory which are required, and show that the resulting corrections lead only to small modifications of the predictions for ballistic-phonon propagation.) Thus it should be possible to find a small additional orthogonal transformation [which we will write as $\exp(i\,\delta\underline{H})$, where $\delta\underline{H}$ is a small Hermitian matrix], which again decouples the acoustic and optic subspaces,

$$\exp(-i\,\delta\underline{H})\underline{R}^U(\mathbf{k})\exp(i\,\delta\underline{H}) = \begin{bmatrix} \underline{D}^a(\mathbf{k}) & \underline{0}_{3\times3} \\ \underline{0}_{3\times3} & \underline{D}^o(\mathbf{k}) \end{bmatrix} ,$$

(28)

and permits us to identify the phenomenological elastic dynamical matrix $\underline{D}^a(\mathbf{k})$ at all $\mathbf{k}$. Since $\delta\underline{H}$ is small, we will be able to write a systematic perturbation expansion for it in $\mathbf{k}$. (See, however, the Appendix.)

Before proceeding with this perturbation analysis, we must discuss the constraints which symmetry imposes on the transformation matrix $\delta\underline{H}$. These restrictions are quite essential, because without them there are an infinite number of distinct orthogonal transformations which satisfy Eq. (28).

First, we consider how the microscopic dynamical matrix $\underline{R}^U$ and the phenomenological elastic dynamical matrix $\underline{D}$ transform under rotations. Suppose the coordinate system is rotated by $\tilde{u}_\alpha = S_{\alpha\beta} u_\beta$ (we use $\underline{S}$ for the rotation matrix to avoid confusion with the dynamical matrix $\underline{R}$). We wish to know how the dynamical matrix of Eq. (13) transforms. Just taking the leading-order term (the others behave identically),

$$\tilde{D}_{\alpha\gamma}^a(\mathbf{k}) = \tilde{c}^{\alpha\beta\gamma\delta} k_\beta k_\delta$$

$$= S_{\alpha i} c^{ijkl} S_{\gamma k}(S_{\beta j} k_\beta)(S_{\delta l} k_l)$$

$$= S_{\alpha i} D_{ij}^a(S_{\beta j} k_\beta) S_{\alpha k} ,$$

(29)

or, more schematically,

$$\tilde{D}^a(\mathbf{k}) = \underline{S}\underline{D}^a(\underline{S}^T\mathbf{k})\underline{S}^T .$$

(30)

The microscopic dynamical matrix satisfies a very similar equation under rotation of the coordinate system. The requirement that the microscopic action [Eq. (22)] be vari-

ant under rotation imposes the following requirement on the matrix of force constants:

$$\widetilde{K}^{\,ai\beta j}_{\mu\nu}=\sum_{l,m}S_{\mu l}K^{ai\beta j}_{lm}S_{\nu m}\;.\qquad(31)$$

Equation (31) permits us to write, for the dynamical matrix [see Eq. (25)],

$$\widetilde{R}^{\,\alpha\beta}_{\mu\nu}=\frac{\rho}{\sqrt{M_\alpha M_\beta}}\sum_{j,l,m}S_{\mu l}K^{ai\beta j}_{lm}e^{iS_{\mu l}k_\mu(X^{\beta j}_l-X^{ai}_l)}S_{\nu m}\;.$$

$$=\sum_{l,m}S_{\mu l}\frac{\rho}{\sqrt{M_\alpha M_\beta}}\sum_{j}K^{ai\beta j}_{lm}e^{iS_{\mu l}k_\mu(X^{\beta j}_l-X^{ai}_l)}S_{\nu m}\;.\qquad(32)$$

In matrix notation, we find that the rotation matrix acts separately on each 3×3 block of the dynamical matrix:

$$\begin{bmatrix}\widetilde{\underline{R}}^{\,11}(\mathbf{k}) & \widetilde{\underline{R}}^{\,12}(\mathbf{k})\\ \widetilde{\underline{R}}^{\,21}(\mathbf{k}) & \widetilde{\underline{R}}^{\,22}(\mathbf{k})\end{bmatrix}=\begin{bmatrix}\underline{S} & \underline{0}_{3\times3}\\ \underline{0}_{3\times3} & \underline{S}\end{bmatrix}\begin{bmatrix}\underline{R}^{11}(\underline{R}^{T}\mathbf{k}) & \underline{R}^{12}(\underline{R}^{T}\mathbf{k})\\ \underline{R}^{21}(\underline{R}^{T}\mathbf{k}) & \underline{R}^{22}(\underline{R}^{T}\mathbf{k})\end{bmatrix}\begin{bmatrix}\underline{S}^{T} & \underline{0}_{3\times3}\\ \underline{0}_{3\times3} & \underline{S}^{T}\end{bmatrix}\;.\qquad(33)$$

Since the matrix

$$\begin{bmatrix}\underline{S} & \underline{0}_{3\times3}\\ \underline{0}_{3\times3} & \underline{S}\end{bmatrix}$$

commutes with the $\underline{U}$ defined by Eq. (26), Eq. (33) applies to the rotated dynamical matrix $\underline{R}^{\,U}$ as well.

We now use the symmetry conditions, Eqs. (30) and (33), to constrain the $\mathbf{k}$-dependent small rotation $\exp(i\,\delta\underline{H})$. Inserting Eq. (28) into Eq. (33) gives

$$\exp[i\,\delta\widetilde{\underline{H}}(\mathbf{k})]\begin{bmatrix}\widetilde{\underline{D}}^{\,a}(\mathbf{k}) & \underline{0}_{3\times3}\\ \underline{0}_{3\times3} & \widetilde{\underline{D}}^{\,o}(\mathbf{k})\end{bmatrix}\exp[-i\,\delta\widetilde{\underline{H}}(\mathbf{k})]=\begin{bmatrix}\underline{S} & \underline{0}_{3\times3}\\ \underline{0}_{3\times3} & \underline{S}\end{bmatrix}\exp[i\,\delta\underline{H}(\underline{S}^{T}\mathbf{k})]\begin{bmatrix}\underline{D}^{a}(\underline{S}^{T}\mathbf{k}) & \underline{0}_{3\times3}\\ \underline{0}_{3\times3} & \underline{D}^{o}(\underline{S}^{T}\mathbf{k})\end{bmatrix}$$

$$\times\exp[-i\,\delta H(\underline{S}^{T}\mathbf{k})]\begin{bmatrix}\underline{S}^{T} & \underline{0}_{3\times3}\\ \underline{0}_{3\times3} & \underline{S}^{T}\end{bmatrix}\;.\qquad(34)$$

Comparing this with Eq. (30) gives a condition on the upper 3×3 block of $\exp(i\,\delta\underline{H})$:

$$\underline{S}=\exp[-i\,\delta\underline{H}(\mathbf{k})]^{aa}\underline{S}\exp[i\,\delta\underline{H}(\underline{R}^{T}\mathbf{k})]^{aa}\;.\qquad(35)$$

Suppose we require that the optic submatrix $\underline{D}^{o}$ satisfy the same transformation relation under rotation as $\underline{D}^{a}$ [Eq. (30)], i.e.,

$$\widetilde{\underline{D}}^{\,o}(\mathbf{k})=\underline{S}\,\widetilde{\underline{D}}^{\,o}(\underline{S}^{T}\mathbf{k})\underline{S}^{T}\;.\qquad(36)$$

While this is not guaranteed by any general symmetry principle, it certainly has no effect on the condition we are interested in, namely the rotational invariance of the acoustic subblock. If Eq. (36) is required, then Eq. (35) is extended to read

$$\begin{bmatrix}\underline{S} & \underline{0}_{3\times3}\\ \underline{0}_{3\times3} & \underline{S}\end{bmatrix}=\exp[-i\,\delta\widetilde{\underline{H}}(\mathbf{k})]\begin{bmatrix}\underline{S} & \underline{0}_{3\times3}\\ \underline{0}_{3\times3} & \underline{S}\end{bmatrix}$$

$$\times\exp[i\,\delta\underline{H}(\underline{S}^{T}\mathbf{k})]\;,\qquad(37)$$

or

$$\exp[i\,\delta\widetilde{\underline{H}}(\mathbf{k})]=\begin{bmatrix}\underline{S} & \underline{0}_{3\times3}\\ \underline{0}_{3\times3} & \underline{S}\end{bmatrix}$$

$$\times\exp[i\,\delta\underline{H}(\underline{S}^{T}\mathbf{k})]\begin{bmatrix}\underline{S}^{T} & \underline{0}_{3\times3}\\ \underline{0}_{3\times3} & \underline{S}^{T}\end{bmatrix}\;.\qquad(38)$$

That is, the unitary transformation $\exp[i\,\delta\underline{H}(\mathbf{k})]$ relating the microscopic dynamical matrix to the elastic continuum dynamical matrix satisfies the same rotational invariance condition as the dynamical matrix itself. [See Eq. (30).] In addition, by performing a power-series expansion of $\exp(i\,\delta\underline{H})$ in Eq. (38) and equating powers of $\delta\underline{H}$, we can show

$$\delta\widetilde{\underline{H}}(\mathbf{k})=\begin{bmatrix}\underline{S} & \underline{0}_{3\times3}\\ \underline{0}_{3\times3} & \underline{S}\end{bmatrix}\delta\underline{H}(\underline{S}^{T}\mathbf{k})\begin{bmatrix}\underline{S}^{T} & \underline{0}_{3\times3}\\ \underline{0}_{3\times3} & \underline{S}^{T}\end{bmatrix}\;.\qquad(39)$$

Since δH is Hermitian and satisfies an equation identical to Eq. (33), it must have exactly the same form as a dynamical matrix. Its 3×3 diagonal blocks, $\delta\underline{H}^{aa}$ and δH^{oo}, are of exactly the same form as $\underline{D}^{a}$ or $\underline{D}^{o}$, and therefore may be written using the general wave-vector expansion of Eq. (13):

$$\delta H^{aa,oo}(\mathbf{k})=c'^{\alpha\beta\gamma\delta}k_\beta k_\delta+id'^{\alpha\beta\gamma\delta\rho}k_\beta k_\delta k_\rho$$

$$+f'^{\alpha\beta\gamma\delta\sigma}k_\beta k_\delta k_\rho k_\sigma+\cdots\;.\qquad(40)$$

The real constants $c'^{\alpha\beta\gamma\delta}$, $d'^{\alpha\beta\gamma\delta\rho}$, and $f'^{\alpha\beta\gamma\delta\rho\sigma}$ are not numerically related to the elastic constants, but they have all the same symmetry properties.

It would appear that Eq. (40) puts a rather strong con-

straint on the form of $\delta\underline{H}$. By applying one more symmetry, time reversal, we show that $\delta\underline{H}^{aa}$ and $\delta\underline{H}^{oo}$ are even more strongly constrained: they must, in fact, be *zero*.

The reality of the potential energy (see Sec. II A) requires that the dynamical matrix go to its complex conju-

gate when $\mathbf{k} \to -\mathbf{k}$:

$$\underline{R}^*(\mathbf{k}) = \underline{R}(-\mathbf{k}) , \tag{41}$$

and also, $[\underline{D}^{a,o}(\mathbf{k})]^* = \underline{D}^{a,o}(-\mathbf{k})$. We apply these conditions to Eq. (28), which defines $\exp(i\,\delta\underline{H})$:

$$\exp[i\,\delta\underline{H}^*(\mathbf{k})]\underline{R}^*(\mathbf{k})\exp[-i\,\delta\underline{H}^*(\mathbf{k})] = \exp[i\,\delta\underline{H}^*(\mathbf{k})]\underline{R}(-\mathbf{k})\exp[-i\,\delta\underline{H}^*(\mathbf{k})]$$

$$= \begin{bmatrix} [\underline{D}^a(\mathbf{k})]^* & \underline{0}_{3\times 3} \\ \underline{0}_{3\times 3} & [D^o(\mathbf{k})]^* \end{bmatrix} = \begin{bmatrix} \underline{D}^a(-\mathbf{k}) & \underline{0}_{3\times 3} \\ \underline{0}_{3\times 3} & \underline{D}^o(-\mathbf{k}) \end{bmatrix}$$

$$= \exp[-i\,\delta H(-\mathbf{k})]\underline{R}(-\mathbf{k})\exp[i\,\delta\underline{H}(-\mathbf{k})] . \tag{42}$$

This equation is true if

$$\delta\underline{H}^*(\mathbf{k}) = -\delta\underline{H}(-\mathbf{k}) . \tag{43}$$

This requires that the coefficients of the power series in Eq. (40) be pure imaginary; however, we have already shown that they must be real. Therefore, they are zero, and $\delta\underline{H}^{aa} = \delta\underline{H}^{oo} = \underline{0}_{3\times 3}$.

These constraints permit a unique perturbation-theoretic solution for the parts of $\delta\underline{H}$ which remain, $\delta\underline{H}^{ao}$ and $\delta\underline{H}^{oa} = (\delta\underline{H}^{ao})^T$. This perturbation solution is developed as problem (11.10.7) of Ref. 14; we will review the steps and present the solution here. First, the microscopic dynamical matrix $\underline{R}^U(\mathbf{k})$ of Eq. (27) is divided into three parts: $\underline{R}^U(\mathbf{k}) = \underline{H}_0 + \underline{Y}(\mathbf{k}) + \underline{X}(\mathbf{k})$, where

$$\underline{H}_0 \equiv \begin{bmatrix} E^a\mathbf{1}_3 & \underline{0}_{3\times 3} \\ \underline{0}_{3\times 3} & E^o\mathbf{1}_3 \end{bmatrix} \tag{44}$$

$[E^a = 0$, see below Eq. (26)],

$$\underline{Y}(\mathbf{k}) \equiv \begin{bmatrix} \underline{R}^U_{aa}(\mathbf{k}) - E^a\mathbf{1}_3 & \underline{0}_{3\times 3} \\ \underline{0}_{3\times 3} & \underline{R}^U_{oo}(\mathbf{k}) - E^o\mathbf{1}_3 \end{bmatrix} , \tag{45}$$

and

$$\underline{X}(\mathbf{k}) \equiv \begin{bmatrix} \underline{0}_{3\times 3} & \underline{R}^U_{ao}(\mathbf{k}) \\ \underline{R}^U_{oa}(\mathbf{k}) & \underline{0}_{3\times 3} \end{bmatrix} . \tag{46}$$

$\underline{X}(\mathbf{k})$ will serve as the small parameter of the theory, being of order $\mathbf{k}$. We wish to find the transformation $\exp(i\,\delta\underline{H})$ which satisfies Eq. (28). Since $\delta\underline{H}$ is small, we perform a power-series expansion of the exponential and collect the terms as commutators. Using $\delta\underline{H}^{aa} = \delta\underline{H}^{oo} = \underline{0}_{3\times 3}$, we find

$$\begin{bmatrix} \underline{D}^a(\mathbf{k}) & \underline{0}_{3\times 3} \\ \underline{0}_{3\times 3} & \underline{D}^o(\mathbf{k}) \end{bmatrix} = \underline{H}_0 + \underline{Y} - \frac{1}{2!}[[(\underline{H}_0 + \underline{Y}), \delta\underline{H}], \delta\underline{H}]$$

$$+ \frac{1}{4!}[[[[(\underline{H}_0 + \underline{Y}), \delta\underline{H}], \delta\underline{H}], \delta\underline{H}], \delta\underline{H}] + \cdots - i[\underline{X}, \delta\underline{H}] + \frac{i}{3!}[[[\underline{X}, \delta\underline{H}], \delta\underline{H}], \delta\underline{H}] + \cdots , \tag{47}$$

and

$$\begin{bmatrix} \underline{0}_{3\times 3} & \underline{D}^{ao}(\mathbf{k}) \\ \underline{D}^{oa}(\mathbf{k}) & \underline{0}_{3\times 3} \end{bmatrix} = \underline{X} - \frac{1}{2!}[[\underline{X}, \delta\underline{H}], \delta\underline{H}] + \cdots - i[(\underline{H}_0 + \underline{Y}), \delta\underline{H}] + \frac{i}{3!}[[[(\underline{H}_0 + \underline{Y}), \delta\underline{H}], \delta\underline{H}], \delta H] + \cdots . \tag{48}$$

However, we want $\underline{D}^{ao} = \underline{D}^{oa} = \underline{0}_{3\times 3}$; we use this to solve for $\delta\underline{H}$. Equation (48) can be solved iteratively: the first iterate of this equation is

$$i[\underline{H}_0, \delta\underline{H}] = \underline{X} , \tag{49}$$

which can be solved:

$$\delta\underline{H} = -i \begin{bmatrix} \underline{0}_{3\times 3} & \dfrac{\underline{R}^U_{ao}(\mathbf{k})}{E^a - E^o} \\ \dfrac{\underline{R}^U_{oa}(\mathbf{k})}{E^o - E^a} & \underline{0}_{3\times 3} \end{bmatrix} \equiv -i\underline{X}^H . \tag{50}$$

We have used a notation of Lax:[14] if $[\underline{H}_0, \underline{A}] = \underline{Z}$, then

$\underline{A} = \underline{Z}^H$. Subsequent iterations lead to a series solution for $\delta\underline{H}$:

$$\delta\underline{H} = -i\underline{X}^H + i[\underline{Y},\underline{X}^H]^H - \cdots . \tag{51}$$

Finally, Eq. (51) may be inserted into Eq. (47) to obtain the desired result:

$$\begin{bmatrix} \underline{D}^a(\mathbf{k}) & \underline{0}_{3\times3} \\ \underline{0}_{3\times3} & \underline{D}^o(\mathbf{k}) \end{bmatrix} = \sum_{j=0}^{\infty} \underline{K}^{(j)} , \tag{52}$$

$$\underline{K}^{(0)} = \underline{H}_0, \quad \underline{K}^{(1)} = \underline{Y} ,$$

$$\underline{K}^{(2)} = \tfrac{1}{2}[\underline{X}^H, \underline{X}] ,$$

$$\underline{K}^{(3)} = \tfrac{1}{2}[\underline{X}, [\underline{Y}, \underline{X}^H]^H] , \tag{53}$$

$$\underline{K}^{(4)} = \tfrac{1}{2}[\underline{X}, \{[[\underline{Y},\underline{X}^H]^H, \underline{Y}]^H + \tfrac{2}{3}[\underline{K}^{(2)},\underline{X}^H]^H\}]$$
$$- \tfrac{1}{12}[[\underline{K}^{(2)},\underline{X}^H], \underline{X}^H] .$$

Equations (52) and (53) provide the desired connection between the microscopic lattice-dynamical theory and the phenomenological elastic theory. From the derivation, it is clear that the connection is unique, establishing the uniqueness of the elastic model. (We have not explored the case of more than two atoms per unit cell in detail. In this case more conditions need to be imposed on δH because there is freedom in the choice of basis vectors for the $\mathbf{k}=0$ optic modes in the matrix U [see Eq. (26)]. It appears certain, however, that a unique elastic theory can be obtained in this case as well.) The terms $K^{(j)}$ in the expansion equation (52) correspond to successively higher powers of $\mathbf{k}$, although the identification is not one to one. For the acoustic dynamical matrix, $K^{(0)}$ contributes nothing, $K^{(1)}$ and $K^{(2)}$ contribute $\mathbf{k}^2$ corrections as well as all higher powers of $\mathbf{k}$ ($K^{(2)}$ describes the so-called "internal shift"[14] in nonprimitive crystals), $K^{(3)}$ contributes $\mathbf{k}^3$ and all higher powers, and $K^{(4)}$ contributes $\mathbf{k}^4$ and all higher powers. Since we are interested only up to $\mathbf{k}^4$, we can stop here. In principle, however, Eq. (52) defines a power-series expansion to all orders in $\mathbf{k}$ of the elastic dynamical matrix. We speculate that in crystals with O_h symmetry (e.g., Si, Ge), since the acoustic branches have cusps at the X point in the Brillouin zone[14] (because of the degeneracy of the LA and LO modes at the X point for these crystals), this power series has a finite radius of convergence. In solids with T_d symmetry, the LA and LO modes are nondegenerate at the X point; this means that the acoustic dispersions are smooth everywhere, suggesting that the power series defined by Eq. (52) has an infinite radius of convergence.

B. Computational procedure

Finally, we can explain how the elastic constants in Table II have been extracted from the various lattice-dynamical models. Using any of these models,[8-11] we can compute the dynamical matrix $\underline{R}^U(\mathbf{k})$ [Eq. (27)] for any value of $\mathbf{k}$. We then use Eqs. (52) and (53) to evaluate the elastic dynamical matrix; these expressions are correct up to order k^4, and contain high-order terms as well. To extract the elastic constants, we simply fit to the expressions for certain elastic dynamical matrix elements along high-symmetry directions in k. The ones we use are [using Eq. (21)]

$$D^a_{xx}(k,0,0) = c^{1111}k^2 + f^{111111}k^4 ,$$

$$D^a_{yy}(k,0,0) = c^{1212}k^2 + f^{122122}k^4 ,$$

$$D^a_{xx}(k,k,0) = (c^{1111}+c^{1212})k^2$$
$$+ (f^{111111}+f^{122122})k^4 + 6f^{211222}k^4 ,$$

$$D^a_{xy}(k,k,0) = 2c^{1122}k^2 + 8f^{112222}k^4 ,$$

$$D^a_{xz}(k,k,0) = i3d^{12223}k^3 ,$$

$$D^a_{zz}(k,k,0) = 2c^{1212}k^2 + 2f^{122122}k^4 + 6f^{122133}k^4 , \tag{54}$$

$$D^a_{xy}(k,k,k) = 2c^{1122}k^2 + 8f^{112222}k^4 + 12f^{112233}k^4 ,$$

$$D^a_{xx}(k,k,k) = c^{1111}k^2 + 2c^{1212}k^2$$
$$+ (f^{111111} + 2f^{122122} + 6f^{122133}$$
$$+ 12f^{211222})k^4 .$$

Fitting these is sufficient to overdetermine the elastic constants; the internal consistency with which the equations is satisfied is about 10%, indicating roughly 10% accuracy for the elastic constants of Table II.

IV. DISCUSSION

In this section we will begin by considering various approximation schemes which may be applied to our elastic model that could lead to a simpler description or one which is more useful as a heuristic tool.

First, as is common in elastic theories, we consider the isotropic approximation. Here we assume that the phonon eigenfrequencies and eigenvectors are the same, no matter what the direction of propagation. Algebraically, this means that the dynamical matrices for $\mathbf{k}$ and for a rotated direction $\underline{S}\mathbf{k}$ are related by

$$\underline{D}(\underline{S}\mathbf{k}) = \underline{S}^T \underline{D}(\mathbf{k})\underline{S} . \tag{55}$$

This is superficially similar to Eq. (30), but it is, in fact, much more restrictive, and it requires relationships among the various elastic constants. These are

$$2c^{1122} = c^{1111} - c^{1212} , \tag{56a}$$

$$d^{12223} = 0 , \tag{56b}$$

$$f^{122133} = \tfrac{1}{3}f^{122122} , \tag{56c}$$

$$f^{211222} = \tfrac{1}{6}f^{111111} + \tfrac{1}{6}f^{122122} \tag{56d}$$

$$f^{112222} = \tfrac{1}{4}f^{111111} - \tfrac{1}{4}f^{122122} , \tag{56e}$$

$$f^{112233} = \tfrac{1}{12}f^{111111} - \tfrac{1}{12}f^{122122} . \tag{56f}$$

Thus in this approximation, the theory requires no third-order elastic constants and only two independent fourth-order constants. (And, as is well known,[16] the number of ordinary elastic constants is reduced from three to two.) The reason that the fifth-rank tensor vanishes is that the underlying symmetry group, T_d, has a mirror plane which, coupled to the assumed complete rotational symmetry, introduces an effective inversion sym-

metry which requires all odd-rank tensors to vanish. The two fourth-order constants correspond to very simple physical quantities: f^{111111} controls the dispersion of the longitudinal wave and f^{122122} controls the dispersion of the transverse waves.

A comparison of Eqs. (56) with Table II shows that, while the assumption of isotropy is not particularly good for the lowest-order elastic constants c, nevertheless it correctly predicts the general ordering of the magnitudes of the six fourth-order dispersive elastic constants. Thus it should be true that f^{111111} still primarily controls longitudinal dispersion and f^{122122} primarily transverse dispersion. This is suggestive, since we see that the greatest distinguishing feature of the shell model (which fits experiment[7]) compared with the others (which do not) is a much larger value of f^{122122}. This indicates that the most troublesome and crucial feature of constructing good lattice-dynamical descriptions of dispersion at long wavelength is to get a sufficiently large dispersion transverse-acoustic branch. This is to be expected, since it is known that the TA branch in zinc-blende and diamond crystals has very unusual dispersion at large wave vector,[38] and is frequently difficult to fit using lattice dynamics. We seem to see a reflection of this difficulty at long wavelength in the uncertainty of f^{122122}.

This suggests several possible approximate schemes for obtaining a better fit to ballistic-phonon—propagation experiments[7] using the present model. One could make the isotropic approximation for the f's (but *not*, say, for the c's and d's), fix the value of f^{111111} to be the average of those obtained from the lattice dynamics of Table II, and permit f^{122122} to be an adjustable parameter. Another possible approach would not make an isotropic approximation for the other f's, but would simply freeze them at some average value taken from Table II, again letting f^{122122} be an adjustable fitting parameter. Either approach reflects the prejudice of the present analysis that f^{122122} plays the greatest role in determining the observed dispersions. More numerical work will be required to substantiate this hypothesis.

Future work developing out of the present model could move in several directions. Hopefully, phonon imaging or similar techniques will permit the estimation of higher-order elastic constants for a whole series of homologous compounds (e.g., the III-V's).[35,39] Dispersive trends should be more evident by use of the present theory than by comparison of the disparate lattice-dynamical models for these systems. Again, we speculate that f^{122122} is the crucial parameter determining trends through a series.

Another line of future investigation could include the search for other physical phenomena which are influenced by dispersion. An obvious one is the low-temperature specific heat. In the classical derivation of the Debye specific heat,[15] the low-temperature T^3 law arises directly from the quadratic increase in the phonon density of states at low frequency, which, in turn, comes directly from the linear ω-vs-k dispersion law. The present extensions to elastic theory permit us to study the leading corrections to the Debye specific heat.[40] Considering the isotropic approximation, the correction to the dispersion law is

$$\rho\omega^2 = ck^2 + fk^4 , \tag{57}$$

or, to lowest order,

$$\omega = v_s k + wk^3 , \tag{58}$$

where

$$v_s = \left[\frac{c}{\rho} \right]^{1/2} , \quad w = \frac{1}{2} \frac{f}{\sqrt{\rho c}} . \tag{59}$$

This implies a correction to the Debye density of states in frequency:

$$D(\omega) = \frac{4\pi}{(2\pi)^3} \left[\frac{\omega^2}{v_s^3} - \frac{5w}{v_s^6} \omega^4 + \cdots \right] . \tag{60}$$

Since f of Eq. (57) is generally negative, w is generally negative and the correction to the density of states is positive; the density of states is typically enhanced compared with a nondispersive model. This should lead to an enhancement of the specific heat, and indeed it does. Performing the standard calculations,[15] we obtain

$$C_V = \frac{2\pi^2}{5} \frac{k_b^4}{\hbar^3 v_s^3} T^3 - \frac{20\pi^4}{63} \frac{wk_b^6}{\hbar^5 v_s^6} T^5 . \tag{61}$$

So, we expect a measurable T^5 enhancement of the specific heat which is entirely a result of lowest-order phonon dispersion. This quantity would thus serve as a check of the elastic constants obtained from the phonon-imaging experiments. In the more realistic anisotropic case, corrections to the specific heat would remain T^5, and v_s and w would simply be replaced by some suitable average over propagation directions.

A rather old experiment of the specific heat of GaAs (Ref. 41) has actually attempted to extract the coefficients of the successive powers of T in the specific heat:

$$C_V = a_1 T^1 + a_3 T^3 + a_5 T^5 + \cdots . \tag{62}$$

This experiment finds $a_1 = 24.8$ ergs/K^2 mol = 1.8 ergs/K^2 cm^3, $a_3 = 467$ ergs/K^4 mol = 34.4 ergs/K^4 cm^3, and $a_5 = 1.43$ ergs/K^6 mol = 0.11 erg/K^6 cm^3. Using Eqs. (59)—(61), we can use these numbers to generate predictions for the isotropic average elastic constants c and f; we obtain $c = 0.6 \times 10^{12}$ dyn/cm^2 and $f = 4 \times 10^{-3}$ dyn. The value for c represents a quite plausible isotropic average of the elastic constants of Table II; on the other hand, the value for f does not correspond so well with the values we have derived from lattice dynamics, being about an order of magnitude too high. Our feeling is that a remeasurement of this specific heat would be worthwhile. The value reported in Ref. 41 for a_5 is quite possibly unreliable, for two reasons: (1) The data were taken over a very limited temperature range, 1—4.2 K; (2) the GaAs was not intrinsic, giving the background linear specific heat mentioned above. Present-day measurement and preparation techniques can improve on both of these points.

To summarize, we have developed the theory of the lowest-order extension of the elastic continuum model of crystals to include dispersive effects. We may think of

this extension either as permitting the elastic constants to acquire a wave-vector dependence, or as including higher-order derivatives of the displacement field in the elastic energy of the solid. We have applied standard group-theoretic techniques to analyze the symmetry properties of the new dispersive elastic constants. This new elastic model is rigorously derivable from microscopic lattice-dynamical descriptions of the solid via a unitary transformation whose derivation we have reviewed. As a consequence, it is not surprising that the present elastic model provides a description of dispersive ballistic-phonon propagation which is of at least equal quality to that of the lattice-dynamical models. Further adjustment of the elastic parameters should result in a numerical fit to experiment which is superior to any previous models. More importantly, the dispersive elastic model provides a more natural language for comparing trends among similar compounds, for understanding which are the most important parameters describing dispersion (as we have done for the TA branches), and for uncovering relationships between dispersive phonon propagation and other physical phenomena like the specific heat.

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APPENDIX: PIEZOELECTRIC CORRECTIONS TO ELASTIC DISPERSION

It is mentioned in Sec. III that the long-wavelength treatment of acoustic waves considered in the present paper is strictly incorrect when the solid is ionic. Ions interact with the electromagnetic field, and as a consequence the acoustic waves are not purely elastic, but they contain an electromagnetic component as well. In this appendix we outline how this effect is included in a more comprehensive phenomenological theory, and we mention how this comes out of a more careful analysis of microscopic lattice-dynamical theory. The conclusion of this section will be that the effect of piezoelectricity on the acoustic modes is negligibly small for GaAs ($<0.5\%$) in both the nondispersive and dispersive regimes. We are therefore justified in ignoring it in analyzing ballistic-phonon propagation.

The proper way of including electromagnetic effects in our phenomenological elastic model is by incorporating the electromagnetic action with our elastic one, Eq. (5): Ignoring dispersive terms for the moment, we have[18,23]

$$A = \int d\mathbf{x}\, dt \left[\tfrac{1}{2}\rho\dot{u}_\alpha\dot{u}_\alpha - \tfrac{1}{2}C^{\alpha\beta\gamma\delta}\partial_\beta u_\alpha \partial_\delta u_\gamma \right.$$

$$\left. + \tfrac{1}{2}\epsilon_{ij}E_i E_j - \frac{1}{2\mu_0}B_i B_i + e^{\alpha\beta\gamma}\partial_\beta u_\alpha E_\gamma \right].$$

(A1)

The first two terms are the purely mechanical energies discussed extensively in the text. The next two terms will produce Maxwell's equations for a medium with dielectric tensor ϵ_{ij} and magnetic permeability μ_0. As usual, $\mathbf{E}$ and $\mathbf{B}$ come from a more basic underlying dynamical variable, the vector potential $\mathbf{A}$:[42]

$$\mathbf{E} = -\frac{1}{c}\dot{\mathbf{A}}, \quad \mathbf{B} = \nabla\times\mathbf{A}.$$

(A2)

The final term in Eq. (A1) gives the linear coupling between mechanical strains and the electric field; e is the piezoelectric tensor.[43] (We will ignore piezomagnetism.[44])

We now obtain two equations of motion, one from minimizing the action with respect to u_α:

$$\rho\ddot{u}_\alpha = c^{\alpha\beta\gamma\delta}\partial_\beta\partial_\delta u_\gamma - e^{\alpha\beta\gamma}\partial_\beta E_\gamma.$$

(A3)

The other is obtained by minimizing with respect to the vector potential A_α:

$$-\frac{1}{\mu_0}(\nabla\times\mathbf{B})_\alpha + \frac{1}{c}\epsilon_{\alpha\beta}\dot{E}_\beta + \frac{1}{c}e^{\alpha\beta\gamma}\partial_\gamma\dot{u}_\beta = 0.$$

(A4)

The existence of a vector potential guarantees that another of Maxwell's equations,

$$\nabla\times\mathbf{E} + \frac{1}{c}\dot{\mathbf{B}} = 0,$$

(A5)

remains unchanged. Combining (A4) and (A5),

$$(\nabla\times\nabla\times\mathbf{E})_\alpha + \frac{\mu_0}{c^2}\epsilon_{\alpha\beta}\ddot{E}_\beta + \frac{\mu_0}{c^2}e^{\alpha\beta\gamma}\partial_\gamma\ddot{u}_\beta = 0.$$

(A6)

We now seek solutions to the coupled wave equations (A3) and (A6). Since we are primarily interested in the soundlike modes with very low frequency, we make a quasistatic approximation for E:[44]

$$\nabla\times\mathbf{E} = 0.$$

(A7)

This permits us to write $\mathbf{E}$ as the gradient of some potential $\mathbf{E} = \nabla\phi$. In terms of ϕ, Eqs. (A3) and (A6) are written

$$\rho\ddot{u}_\alpha = c^{\alpha\beta\gamma\delta}\partial_\beta\partial_\delta u_\gamma - e^{\alpha\beta\gamma}\partial_\beta\partial_\gamma\phi,$$

(A8)

$$\epsilon_{\alpha\beta}\partial_\beta\ddot{\phi} + e^{\alpha\beta\gamma}\partial_\gamma\ddot{u}_\beta = 0.$$

(A9)

Fourier-transforming in space and time gives

$$-\rho\omega^2 u_\alpha = -c^{\alpha\beta\gamma\delta}k_\beta k_\delta u_\gamma + e^{\alpha\beta\gamma}k_\beta k_\gamma\phi,$$

(A10)

$$-i\epsilon_{\alpha\beta}k_\beta\omega^2\phi - ie_{\alpha\beta\gamma}k_\gamma\omega^2 u_\beta = 0.$$

(A11)

We can now eliminate ϕ to obtain a single equation of motion for u_α:

$$\rho\omega^2 u_\alpha = c^{\alpha\beta\gamma\delta}k_\beta k_\delta u_\gamma + \frac{(e^{\alpha\beta\rho}k_\beta k_\rho)(e^{\gamma\gamma\delta}k_\nu k_\delta)}{k_\alpha \epsilon_{\alpha\beta}k_\beta}u_\gamma .$$

$$(A12)$$

We can view this as an ordinary equation for sound waves, but with so-called "piezoelectrically stiffened" elastic constants $\tilde{c}^{\alpha\beta\gamma\delta}$:

$$\tilde{c}^{\alpha\beta\gamma\delta} = c^{\alpha\beta\gamma\delta} + \frac{e^{\alpha\beta\mu}k_\mu k_\nu e^{\gamma\gamma\delta}}{k_i \epsilon_{ij}k_j} .$$

$$(A13)$$

This is a misnomer for several reasons. First, the sound velocities may equally well be softened as stiffened by this new term. Second, it is improper to call $\tilde{c}^{\alpha\beta\gamma\delta}$ a set of elastic constants, since they are k dependent (with a non-analytic dependence around $\mathbf{k}=0$), and hence do not transform properly under rotation. Still, it is very useful to use Eq. (A13) to estimate the magnitude of the piezoelectric stiffening. For crystals with T_d symmetry, only piezoelectric tensor components of the type e^{123} are

nonvanishing.[16] $e^{123}=0.154$ for GaAs (in the conventional rationalized MKSA units[44]). Because of this symmetry, among the cubic elastic constants, the only one to be stiffened is c^{1122}. The magnitude of this stiffening is

$$\Delta c^{1122} \approx \frac{(e^{123})^2}{\epsilon} = 2.2 \times 10^9 \text{ erg/cm}^2 ,$$

$$(A14)$$

taking $\epsilon = 12.5$ for GaAs. Comparing this with c^{1122} of Table II, we find

$$\frac{\Delta c^{1122}}{c^{1122}} = 0.4\% ,$$

$$(A15)$$

which is utterly negligible.

So much was suspected previously.[45,46] The important question for the present work is: Does the piezoelectric effect on the dispersive elastic constants, $\Delta d/d$ and $\Delta f/f$, have similarly small magnitude? The answer is yes, as we now show. When dispersion is included, the action of Eq. (A1) is augmented by the following terms:

$$A_{\text{dispersive}} = \int d\mathbf{x}\,dt(d^{\alpha\beta\gamma\delta\rho}\partial_\beta u_\alpha \partial_{\delta\rho}u_\gamma + f^{\alpha\beta\gamma\delta\rho\sigma}\partial_\beta u_\alpha \partial_{\delta\rho\sigma}u_\gamma + \gamma_{\alpha\beta\gamma}E_\alpha\partial_\gamma E_\beta + \psi_{\alpha\beta\gamma\delta}E_\alpha\partial_{\gamma\delta}E_\beta$$

$$+ g_{\alpha\beta\gamma\delta}\partial_\beta u_\alpha\partial_\delta E_\gamma + h_{\alpha\beta\gamma\delta\rho}\partial_\beta u_\gamma \partial_{\delta\rho}E_\gamma) .$$

$$(A16)$$

γ and ψ are dispersive corrections to the constitutive relation between the crystal polarization $\mathbf{P}$ and the electric field $\mathbf{E}$. γ is known as the optical activity tensor,[16] since it describes the rotation of the plane of polarization of linearly polarized light. ($\gamma=0$ for crystals with T_d symmetry,[16] but we will carry it through the analysis anyway.) g and h are dispersive connections to the piezoelectric tensor.

By working through the identical analysis as above with $A_{\text{dispersive}}$ included, we find piezoelectrically stiffened dispersive elastic constants $\tilde{d}$ and $\tilde{f}$. The result for $\tilde{d}$ is

$$\tilde{d}^{\alpha\beta\gamma\delta\rho} = d^{\alpha\beta\gamma\delta\rho} + \frac{g^{\alpha\beta\gamma\mu}k_\mu k_\nu e^{\nu\delta\rho}}{k_i\epsilon_{ij}k_j}$$

$$+ \frac{e^{\alpha\beta\mu}k_\mu k_\nu g^{\gamma\gamma\delta\rho}}{k_i\epsilon_{ij}k_j} + \frac{e^{\alpha\beta\mu}k_\mu(\gamma_{i\gamma k}k_i k_k)k_\nu e^{\nu\delta\rho}}{(k_i\epsilon_{ij}k_j)^2}$$

$$(A17)$$

This result has appeared as Eq. (32) of Ref. 18. They point out the significance of Eq. (A17) for acoustic activity of solids, which is described by $\tilde{d}$: Equation (A7) shows that acoustic activity is never a purely mechanical effect (described by $d^{\alpha\beta\gamma\delta\rho}$), but is always influenced by coupling to the electromagnetic field. This point was missed in some previous work.[21,47]

A similar expression can be derived for $\tilde{f}$; since the result is rather lengthy, we present it without subscripts, which can be filled in by the interested reader:

$$\tilde{f} = f + \frac{hkke}{k\epsilon k} + \frac{ekkh}{k\epsilon k} + \frac{gkkg}{k\epsilon k} + \frac{gk(\gamma kk)ke}{(k\epsilon k)^2}$$

$$+ \frac{ek(\gamma kk)kg}{(k\epsilon k)^2} + \frac{ek(\gamma kk)^2 ke}{(k\epsilon k)^3} + \frac{ek(\psi kk)ke}{(k\epsilon k)^2} .$$

$$(A18)$$

To estimate the relative importance of Δd and Δf, we must have some estimate for γ, ψ, g and h. Since very little is known about these elastic constants, we resort to the dimensional analysis of Sec. II B, which proved successful for understanding d and f. Again, we assert that the only important length scale is $a_0 \approx 10^{-8}$ cm, the interatomic spacing. Then we estimate

$$\gamma \approx a_0\epsilon \approx 10^{-7} \text{ cm} ,$$

$$(A19a)$$

$$\psi \approx a_0^2\epsilon \approx 10^{-15} \text{ cm}^2 ,$$

$$(A19b)$$

$$g \approx a_0 e \approx 10^{-10} \text{ MKSA unit} ,$$

$$(A19c)$$

$$h \approx a_0^2 e \approx 10^{-20} \text{ MKSA unit} .$$

$$(A19d)$$

The only one of these about which we have some experimental knowledge is γ, the optical activity coefficient. In quartz, $\gamma=2\times10^{-9}$ cm.[48] So, if Eq. (A19) is true, then comparison with Eqs. (A17) and (A18) shows

$$\frac{\Delta f}{f} \approx \frac{\Delta d}{d} \approx \frac{\Delta c}{c} \approx 0.4\% .$$

$$(A20)$$

Given the result from quartz, this is, if anything, an overestimate of the importance of Δd and Δf.

Finally, we mention how the above results are derived from a microscopic lattice-dynamical model; Ref. 1 contains by far the most lucid discussion of this derivation. Our calculation of Sec. III assumes that the microscopic dynamical matrix $R(k)$ is analytic around $\mathbf{k}=0$, and thus can be developed in a power series. This is not the case when the components of the system interact via long-range Coulomb interactions. As Ref. 1 shows, in addition to analytic parts, $R(\mathbf{k})$ contains pieces like

$$R(\mathbf{k})=R_{\text{ana}}(\mathbf{k})+\frac{k_\alpha k_\beta}{k^2} \ . \tag{A21}$$

This nonanalytic piece can be interpreted as the effect of the macroscopic polarization of the solid, thus leading to the sort of phenomenological terms in Eq. (A1). The detailed study of other nonanalytic terms in the microscopic theory, which presumably leads to terms of the sort contained Eq. (A16), we have not undertaken.

*Present and permanent address: IBM Thomas J. Watson Research Center, P.O. Box 218, Yorktown Heights, NY 10598.

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