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Theory of the Photoelastic Interaction

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We apply our recent theory of nonlinear electrodynamics of elastic anisotropic dielectrics to the photoelastic interaction and derive for the first time from a fundamental point of view the form of the photoelastic susceptibility for materials of arbitrary symmetry. An indirect photoelastic effect is shown to exist in piezoelectric crystals. Its susceptibility cannot be represented as an ordinary tensor; it possesses different symmetry than the normal or direct photoelastic susceptibility, and it can be comparable in size to the latter. The direct photoelastic susceptibility can be represented by a fourth-rank tensor, but it lacks the traditionally assumed symmetry upon interchange of the last two (elastic) indices. The independent elastic variable relevant to the photoelastic interaction is the displacement gradient, not the strain as believed since 1841. This arises because rotations can play as significant a role as strains do in photoelasticity of strongly birefringent crystals in the presence of shear distortions. The form of the derived photoelastic tensor gives information about the various origins of the effect and can predict the frequency dispersion of the effect. Though the nonlinear polarization derived here is equally applicable to Brillouin scattering, specific application is made in this paper to acousto-optic scattering, and the form of the phase-matched output wave is derived for waves having an arbitrary orientation to an anisotropic medium.

I. INTRODUCTION

The first nonlinear optical effect discovered was the photoelastic effect observed in 1816 by Brewster.[1] The interpretation of the effect in anisotropic media has been based until now on the phenomenological formulation made by Pockels,[2] which stated that the change in the inverse dielectric tensor $(\delta\kappa^{-1})_{ij}$ was proportional to the strain S_{kl} according to

$$(\delta\kappa^{-1})_{ij}=p_{ijkl}S_{kl}\ , \tag{1.1}$$

where p_{ijkl} is the Pockels photoelastic tensor. Because $(\delta\kappa^{-1})_{ij}$ and S_{kl} are both symmetric tensors, p_{ijkl} should be symmetric upon interchange of i and j and upon interchange of k and l. It should also possess the crystal symmetry of an ordinary fourth-rank tensor.[3]

The modern era of photoelasticity studies began with Brillouin's pioneering work[4] on light scattering from thermal fluctuations in solids (Brillouin scattering). He showed that the scattered light would be shifted in frequency from the incident light by an acoustic frequency. The necessity of quasimomentum conservation in the scattering interaction causes the acoustic frequency active in the scattering process to depend upon the angle of scattering. Brillouin scattering was first observed by Gross[5] in liquids where only longitudinal elastic vibrations play a role. Leontowitsch and Mandlestam[6] calculated the expected scattered intensity in terms of the elastic and photoelastic constants of a solid. Raman and Venkateswaran[7] in gypsum and Sibaiya[8] in Rochelle salt observed Brillouin scattering from transverse as well as longitudinal elastic vibrations. Chandrasekharan[9] pointed out that for unpolarized incident light as many as 12 Brillouin frequency components could be scattered in a birefringent medium. Chiao *et al.*[10] first observed stimulated Brillouin scattering.

Debye and Sears[11] and Lucas and Biquard[12] performed the first scattering of light from coherently generated acoustic waves (acousto-optic scattering). The spatial form of the scattered light in these experiments was explained by Raman and Nath.[13] Mueller studied the photoelastic behavior of cubic crystals theoretically, using polarizable point ions and the anisotropy of the Coulomb potential and the Lorentz local field,[14] and suggested methods of determining the photoelastic tensor by acousto-optic scattering.[15] Such measurements were first made by Galt[16] and by Burstein and Smith[17] on cubic crystals and extended to birefringent crystals by Narasimhamurty.[18] Dixon[19] added considerably to the understanding of acousto-optic scattering in anisotropic media.

Recently we have developed a classical theory of nonlinear electrodynamics of elastic anisotropic dielectrics.[20] The theory is formulated from a microscopic point of view before passage to the continuum limit is made. Of crucial importance to obtaining the correct nonlinear terms is a formulation which allows for finite deformations of the elastic medium. Construction of an appropriately invariant stored-energy function is at the heart of the development. The theory predicts the symmetry of any nonlinear, as well as linear, interaction of electromagnetic waves with various eigenmodes of the solid such as acoustic, ionic, and electronic vibrations; it predicts the various multistep contributions to the over-all interaction and their symmetry and so interrelates various nonlinear interactions; it can predict the dispersion of the susceptibility that governs the nonlinear interaction. The theory is valid for wavelengths of the interacting waves which are long compared to unit-cell dimensions.

In this paper we apply the general theory to the simplest interaction between an input acoustic wave with an input optic wave and derive the photoelastic susceptibility relevant to either acousto-optic or Brillouin scattering. This is the first time that the form of the photoelastic susceptibility has been derived for materials of arbitrary symmetry from a classical point of view that is fundamental (in the sense that the susceptibility is obtained deductively from a Lagrangian which describes a solid as an aggregation of moving charged mass points in interaction with an electromagnetic field). We find that the susceptibility consists of two parts: One is a fourth-rank tensor possessing material symmetry which represents what we will call the direct effect; the other is not a simple fourth-rank tensor but rather a fourth-rank tensor function of the acoustic wave-vector direction. The latter part, which represents what we will call the indirect effect, exists only in piezoelectric materials and represents the macroscopic two-step contribution to photoelasticity through the piezoelectric and electro-optic effects. This indirect part has *different, often lower, symmetry* than the direct photoelastic effect. The indirect effect can, in fact, make a significant contribution to the total effect in certain geometries in some nonferroelectric crystals such as α-iodic acid. It can also be very large in ferroelectric crystals such as lithium niobate. The present theory, however, does not apply to ferroelectric crystals.

The history of the indirect photoelastic effect is quite murky. An awareness of its existence can be found in the literature[21] as evidenced by a realization that the values of the photoelastic tensor components may vary depending on the electromagnetic boundary conditions present on the crystal during measurement. Nevertheless, review papers[1,22] on the photoelastic interaction have completely ignored the indirect photoelastic effect. We find, however, after an extensive search that the form of the indirect photoelastic effect was derived by Chapelle and Taurel[23] though we can find no reference to their work by later workers in the fields of Brillouin and acousto-optic scattering. Concurrent with our work, Coquin[24] has considered the indirect photoelastic effect and its importance in ferroelectric lithium niobate, Brody and Cummins[25] have studied the effect by Brillouin scattering near the ferroelectric transition temperature in potassium dihydrogen phosphate, and Wemple and DiDomenico[26] have estimated its size in oxygen octohedra ferroelectrics from a phenomenological treatment. That the indirect photoelastic effect has a symmetry different from the direct effect seems to have been unrecognized by all workers except Chapelle and Taurel.

We also find that the photoelastic susceptibility tensor which represents the direct effect and hence the properly defined photoelastic tensor p_{ijkl} does *not* possess the traditionally assumed symmetry upon interchange of the last two (acoustic) indices.[27] We show that this occurs because the displacement gradient is the natural independent acoustic variable for photoelastic measurements. From the time of Neumann's work[1] (in 1841) unitl now, the strain, or sometimes the stress, has been believed to be the proper variable. The displacement gradient is needed because the rotation of volume elements, which involves the antisymmetric combination of displacement gradients, contributes to light scattering from shear distortions in strongly birefringent media to an extent comparable to the strain, which is the symmetric combination of displacement gradients. The deviations from equality of p_{ijkl} and p_{ijlk} can be large for common crystals such as calcite, sodium nitrate, etc.

Finally, we will show what internal effects contribute to the photoelastic susceptibility, how these are related to other measurable quantities, and what dispersion is expected for the photoelastic tensor.

Our method is as follows: In Sec. II we begin with the continuum Lagrangian of the problem, passage to the continuum limit from the discrete-particle Lagrangian having been discussed previously.[20] The dynamical equations for the degrees of freedom of the material medium and the electromagnetic field equations are obtained from the Lagrangian. These equations are then expressed in terms of a more convenient internal coordinate system. A polynomial form of the stored energy, expressed in rotationally invariant quantities, is adopted. Next, the equations are linearized in the displacement and expressed in the spatial frame. Only terms that involve at most two fields are retained, as is appropriate for the photoelastic effect.

In Sec. III the various fields are expanded in Fourier series in time and only terms whose frequency is the sum of the optical and acoustical frequencies are retained. The wave equation is formed and the nonlinear driving polarization determined, the latter being the heart of the problem. The wave equation is then solved, neglecting pump-wave depletion. The Poynting vector is obtained for a near phase-matched output but for otherwise arbitrarily oriented waves with respect to the anisotropic medium.

In Sec. IV the symmetry of the derived nonlinear polarization is discussed while in Sec. V its origin and dispersion are discussed. The results of these sections have been summarized above.

II. FORMULATION

We wish to develop the theory of the interaction of an electromagnetic wave at an optical frequency with an acoustic wave at a much lower frequency. To handle vibrations at optical frequencies, the theory, of course, must be dynamic in nature. Since the wavelength of light, as well as the wavelength of sound, is large compared to a unit-cell dimension, a long-wavelength theory is adequate. In this paper we will not deal with effects, such as optical activity, which involve wave-vector dispersion and so represent deviations from the truly long-wavelength limit. The general theory,[20] on which this treatment is based, however, is capable of handling such effects. We will simplify the problem here in two other respects, though generalizations including them are straightforward. An ideal dielectric, that is a material which contains no free charge, will be considered, and loss in the various modes of vibration of the solid will be ignored, although the latter is easy to add. Also, only nonmagnetic and nonferroelectric materials will be considered. Our interest is in anisotropic media and the theory will be formulated so as to apply to crystals of any symmetry as well as to isotropic media. Our treatment will include piezoelectric crystals and will demonstrate the anomalous photoelastic properties of such crystals.

We will begin here by writing the total Lagrangian L of the system in the continuum limit[20] as a sum of a particle Lagrangian I_P, an electromagnetic field Lagrangian L_F, and a field-particle interaction Lagrangian L_I:

$$L = L_P + L_F + L_I \,, \tag{2.1}$$

where

$$L_P = \int \left(\frac{1}{2} \sum_{\alpha=1}^{N} \rho^\alpha [\dot{\vec{x}}^\alpha(\vec{X}, t)]^2 - \rho^0 \bar{\Sigma}\,(\vec{x}^\beta(\vec{X}, t), \vec{x}^\beta_{,A}(\vec{X}, t)) \right) d\vec{X} \,, \tag{2.2}$$

$$L_F = \frac{1}{2}\epsilon_0 \int [\vec{E}^2(\vec{z}, t) - c^2 \vec{B}^2(\vec{z}, t)]\, d\vec{z} \,, \tag{2.3}$$

$$L_I = \int \sum_{\alpha=1}^{N} q^\alpha [\dot{\vec{x}}^\alpha(\vec{X}, t) \cdot \vec{A}(\vec{x}^\alpha(\vec{X}, t)) - \Phi(\vec{x}^\alpha(\vec{X}, t))]\, d\vec{X} \tag{2.4}$$

$$= \int [\vec{j}(\vec{z}, t) \cdot \vec{A}(\vec{z}, t) - q(\vec{z}, t)\, \Phi(\vec{z}, t)]\, d\vec{z} \,. \tag{2.5}$$

In the Lagrangian we regard the vector and scalar potentials $\vec{A}$ and Φ as the generalized coordinates describing the electromagnetic field. They are functions of position, which is expressed in a laboratory Cartesian coordinate system $\vec{z}$, and are related to the electric field $\vec{E}$ and the magnetic induction $\vec{B}$ by the familiar relations

$$\vec{E}(\vec{z}, t) = -\vec{\nabla}\Phi(\vec{z}, t) - \frac{\partial \vec{A}(\vec{z}, t)}{\partial t} \,, \tag{2.6}$$

$$\vec{B}(\vec{z}, t) = \vec{\nabla} \times \vec{A}(\vec{z}, t) \,, \tag{2.7}$$

where rationized mks units are employed. In Eq. (2.3) ϵ_0 is the permittivity of free space and c is the free-space velocity of light. We regard the N different continuum particle positions $\vec{x}^\alpha$, which we choose to express as components x_i^α in a common Cartesian frame called the spatial frame, as the coordinates describing the mechanical motion; N is the number of different particles, ions and electrons in a primitive unit cell. Each $\vec{x}^\alpha$ is to be regarded as a function of $\vec{X}$ and t, the time. The quantity $\vec{X}$ is a continuum variable that designates or names a material point in a frame, called the material frame, which we choose to be Cartesian and as such is a time-independent quantity. A dot over a variable represents the partial time derivative holding $\vec{X}$ fixed, often called the material time derivative. The quantities ρ^α and q^α are, respectively, the mass and charge, taken as constants, of the αth particle divided by the primitive cell volume. The term $\rho^0\bar{\Sigma}$ of Eq. (2.2) is the stored-mechanical-energy density of the material medium. The mass density of the medium ρ^0 is conventionally introduced as a factor in the stored-energy density to make $\bar{\Sigma}$ the stored energy per unit mass. The quantity $\vec{x}^\alpha_{,A}$, of which $\bar{\Sigma}$ is a function, denotes $\partial \vec{x}^\alpha / \partial X_A$, where A denotes the component of $\vec{X}(A = 1, 2, 3)$. When a component of $\vec{x}^\alpha$ need be specified, a lower-case Latin letter will be used as a subscript to distinguish it from the upper-case Latin letters used to denote components of $\vec{X}$. We make the important physical assumption in Eq. (2.2) that the dependence of $\bar{\Sigma}$ on the derivatives of $\vec{x}^\alpha$ with respect to X_A higher than the first will be negligible. We assume a homogeneous medium; hence $\bar{\Sigma}$ does not depend explicitly on $\vec{X}$. It is also important that $\bar{\Sigma}$ possess displacement and rotational invariance.[20] The charge and current densities of Eq. (2.5) are defined by

$$q(\vec{z}, t) = \sum_{\alpha=1}^{N} q^\alpha \int \delta[\vec{z} - \vec{x}^\alpha(\vec{X}, t)]\, d\vec{X} \,, \tag{2.8}$$

$$\vec{j}(\vec{z}, t) = \sum_{\alpha=1}^{N} q^\alpha \int \dot{\vec{x}}^\alpha(\vec{X}, t)\, \delta[\vec{z} - \vec{x}^\alpha(\vec{X}, t)]\, d\vec{X} \,, \tag{2.9}$$

where $\delta[\vec{z} - \vec{x}^\alpha]$ is the Dirac δ function.

The dynamical equations of the electromagnetic field are obtained from the Lagrange equations for the generalized coordinates Φ and $\vec{A}$ with the Lagrangian densities defined in Eqs. (2.3) and (2.5):

$$\epsilon_0 \vec{\nabla} \cdot \vec{E}(\vec{z}, t) = q(\vec{z}, t) \,, \tag{2.10}$$

$$\frac{1}{\mu_0} \vec{\nabla} \times \vec{B}(\vec{z}, t) - \epsilon_0 \frac{\partial \vec{E}(\vec{z}, t)}{\partial t} = \vec{j}(\vec{z}, t) \,, \tag{2.11}$$

where μ_0 is the permeability of free space. The charge and current densities can be expressed in a multipole expansion about the center of mass $\bar{\vec{x}}$ of the primitive unit cell. Magnetic dipole, electric quadrupole,[20] and higher-order terms will be dropped. The free-charge density will be set to zero and the corresponding current dropped since a dielectric is being considered. Equations (2.10) and (2.11) then become

$$\epsilon_0 \vec{\nabla}\cdot\vec{E}(\vec{z},t) = -\vec{\nabla}\cdot\vec{P}(\vec{z},t)\,, \tag{2.12}$$

$$\frac{1}{\mu_0}\vec{\nabla}\times\vec{B}(\vec{z},t) - \epsilon_0\frac{\partial\vec{E}(\vec{z},t)}{\partial t} = \frac{\partial\vec{P}(\vec{z},t)}{\partial t} + \vec{\nabla}\times[\vec{P}(\vec{z},t)\times\dot{\vec{x}}(\vec{z},t)]\,, \tag{2.13}$$

where the polarization $\vec{P}(\vec{z},t)$ is given by

$$\vec{P}(\vec{z},t) = \sum_{\alpha=1}^{N}\frac{q^\alpha\vec{x}^\alpha(\vec{z},t)}{J(\vec{z},t)} \equiv \frac{\vec{p}(\vec{z},t)}{J(\vec{z},t)}\,, \tag{2.14}$$

and the Jacobian of the transformation from $\vec{X}$ to $\bar{\vec{x}}$ is given by

$$J(\vec{z},t) \equiv \left(\det\frac{\partial x_i(\vec{X},t)}{\partial X_A}\right)_{\vec{z}=\bar{\vec{x}}(\vec{X},t)}\,. \tag{2.15}$$

By comparison of Eqs. (2.12) and (2.13) with the conventional forms of Maxwell's equations, it is seen that the electric displacement $\vec{D}$ and the magnetic field $\vec{H}$ must be given by

$$\vec{D}(\vec{z},t) = \epsilon_0\vec{E}(\vec{z},t) + \vec{P}(\vec{z},t)\,, \tag{2.16}$$

$$\vec{H}(\vec{z},t) = (1/\mu_0)\vec{B}(\vec{z},t) + \dot{\vec{x}}(\vec{z},t)\times\vec{P}(\vec{z},t)\,. \tag{2.17}$$

Note that in the electric dipole approximation an effective magnetization term arises from the motion of the polarization. The other two of Maxwell's equations, besides Eqs. (2.12) and (2.13), are direct consequences of the definitions in Eqs. (2.6) and (2.7), namely,

$$\vec{\nabla}\times\vec{E}(\vec{z},t) + \frac{\partial\vec{B}(\vec{z},t)}{\partial t} = 0\,, \tag{2.18}$$

$$\vec{\nabla}\cdot\vec{B}(\vec{z},t) = 0\,. \tag{2.19}$$

The dynamical equations of particle motion are obtained from the Lagrange equations for the coordinates $\vec{x}^\alpha$ with the Lagrangian densities defined in Eqs. (2.2) and (2.4). We obtain[20]

$$\rho^\alpha\ddot{\vec{x}}^\alpha = \vec{f}^\alpha + \vec{q}^\alpha[\vec{E}(\vec{x}^\alpha) + \dot{\vec{x}}^\alpha\times\vec{B}(\vec{x}^\alpha)]\,, \tag{2.20}$$

where

$$f_i^\alpha = -\rho^0\frac{\partial\bar{\Sigma}}{\partial x_i^\alpha} + \rho^0\frac{\partial}{\partial X_A}\,\frac{\partial\bar{\Sigma}}{\partial x_{i,A}^\alpha} \tag{2.21}$$

and $\vec{X}$ is still the independent variable. Repeated tensor subscripts as in the second term on the right-hand side of Eq. (2.21) indicate summation over those subscripts.

At this point we transform from the set of particle coordinates $\vec{x}^\alpha$ to a set of internal coordinates[20,28] $\vec{y}^\mu$ which retains the diagonality of the kinetic energy, which contains the continuum coordinate vector $\bar{\vec{x}}$ representing the center of mass of the primitive unit cell, and which possesses displacement invariance for all members except $\bar{\vec{x}}$. It will be important later in the construction of a rotationally invariant stored energy to have coordinates which are in the form of N vectors (symmetry coordinates or normal coordinates are not generally in such a form). The inclusion of the center of mass as one of the new coordinates will be important in the expansion of the stored energy where derivatives with respect to $\vec{X}$ of $\bar{\vec{x}}$ and the other $\vec{y}^\mu$ will be handled differently. Transformation to internal coordinates is not a restrictive step; at a later point in the development a second transformation to either symmetry or normal coordinates can be made.

We will represent the transformation between these sets of coordinates by

$$\vec{y}^\mu(\vec{X},t) = \sum_{\alpha=1}^{N} U^{\mu\alpha}\vec{x}^\alpha(\vec{X},t), \quad \mu = 0, 1, 2, \ldots, N-1\,. \tag{2.22}$$

Note that we have shifted the range of numbering of $\vec{y}^\mu$ with respect to $\vec{x}^\alpha$. This is chosen to allow denoting the c.m. coordinate $\bar{\vec{x}}$ by $\vec{y}^0$:

$$\vec{y}^0 = \bar{\vec{x}} = \sum_{\alpha=1}^{N}\frac{\rho^\alpha\vec{x}^\alpha}{\rho^0}\,. \tag{2.23}$$

The inverse transformation of Eq. (2.22) will be represented by

$$\vec{x}^\alpha = \sum_{\mu=0}^{N-1} V^{\alpha\mu}\vec{y}^\mu\,. \tag{2.24}$$

The diagonality of the kinetic energy in the internal coordinate system

$$\sum_{\alpha=1}^{N}\rho^\alpha(\dot{\vec{x}}^\alpha)^2 = \sum_{\mu=0}^{N-1} m^\mu(\dot{\vec{y}}^\mu)^2 \tag{2.25}$$

defines the effective mass density m^μ associated with the μth internal coordinate of the medium. It can be shown[20] from Eqs. (2.22)–(2.25) that

$$\rho^\alpha V^{\alpha\mu} = m^\mu U^{\mu\alpha}\,. \tag{2.26}$$

The dynamical equations of particle motion, Eq. (2.20), can be transformed to the internal coordinate representation

$$m^\mu\ddot{\vec{y}}^\mu = \sum_{\alpha=1}^{N} V^{\alpha\mu}\vec{f}^\alpha + \sum_{\alpha=1}^{N} V^{\alpha\mu}q^\alpha[\vec{E}(\vec{x}^\alpha) + \dot{\vec{x}}^\alpha\times\vec{B}(\vec{x}^\alpha)]\,, \tag{2.27}$$

where $\mu = 0, 1, 2, \ldots, N-1$. Denote

$$f_i^\mu = \sum_{\alpha=1}^{N} V^{\alpha\mu}f_i^\alpha = -\rho^0\frac{\partial\hat{\Sigma}}{\partial y_i^\mu} + \rho^0\frac{\partial}{\partial X_A}\,\frac{\partial\hat{\Sigma}}{\partial y_{i,A}^\mu}\,, \tag{2.28}$$

where now

$$\hat{\Sigma} = \hat{\Sigma}(\vec{x}_{,A}, \vec{y}^\nu), \quad \nu = 1, 2, \ldots, N-1, \tag{2.29}$$

the change to new independent variables being indicated by the caret. Note that because of displacement invariance, $\hat{\Sigma}$ is not a function of the c.m. coordinate $\vec{y}^0=\vec{x}$. We neglect its dependence on $\vec{y}^{\nu}_{,A}(\nu=1, 2, \ldots, N-1)$; hence the second term of Eq. (2.28) is needed only when $\mu=0$. This represents the lowest-order approximation in the long-wavelength limit and is equivalent to omission of wave-vector dispersion.

The $\vec{E}(\vec{x}^{\alpha})$ and $\vec{B}(\vec{x}^{\alpha})$ fields of Eq. (2.27) may be expanded in a Taylor series about the c.m. $\vec{x}$ by regarding the variable

$$\vec{x}^{\alpha}(\vec{X}, t)-\vec{x}(\vec{X}, t)=\vec{u}^{\alpha}(\vec{X}, t) \tag{2.30}$$

as a small quantity. This procedure is justified because it has been shown[20] that the local electric field effects can be included in the stored-energy density $\hat{\Sigma}$. Only when treating phenomena such as optical activity,[20] where the phase change of a light wave between atoms of the same cell becomes important, must this expansion be generalized. Only electric monopole and electric dipole terms will be retained; magnetic dipole and quadrupole terms[20] and higher-order terms will be dropped here. Equation (2.27) can then be put in the form

$$\begin{aligned} m^{\mu}\ddot{\vec{y}}^{\mu} &= \vec{f}^{\mu}+q^{\mu}[\vec{E}(\vec{x})+\dot{\vec{x}}\times\vec{B}(\vec{x})]+(\vec{p}^{\mu}\cdot\vec{\nabla})\vec{E}(\vec{x}) \\ &\quad +\dot{\vec{p}}^{\mu}\times\vec{B}(\vec{x})+\dot{\vec{x}}\times(\vec{p}^{\mu}\cdot\vec{\nabla})\vec{B}(\vec{x}), \\ &\mu=1, 2, \ldots, N-1 \ . \end{aligned} \tag{2.31}$$

The effective charge q^{μ} associated with the μth internal coordinate is given by

$$q^{\mu}=\sum_{\alpha=1}^{N} V^{\alpha\mu}q^{\alpha} , \tag{2.32}$$

and the effective polarization associated with it is given by

$$\vec{p}^{\mu}=\sum_{\alpha=1}^{N} V^{\alpha\mu}q^{\alpha}\vec{u}^{\alpha}=\sum_{\nu=1}^{N-1} q^{\mu\nu}\vec{y}^{\nu} , \tag{2.33}$$

where

$$q^{\mu\nu}=\sum_{\alpha=1}^{N} V^{\alpha\mu}q^{\alpha}V^{\alpha\nu}=q^{\nu\mu} . \tag{2.34}$$

We note also that

$$q^{\mu 0}=q^{0\mu}=q^{\mu} , \tag{2.35}$$

since $V^{\alpha 0}=1$.

The c.m. equation can be represented formally the same as Eq. (2.31) with $\mu=0$. However, in an ideal dielectric,

$$q^0=\sum_{\alpha=1}^{N} q^{\alpha}=0 \tag{2.36}$$

and

$$\vec{p}^0=\sum_{\alpha=1}^{N} q^{\alpha}\vec{u}^{\alpha}=\sum_{\nu=1}^{N-1} q^{\nu}\vec{y}^{\nu}=\vec{p} . \tag{2.37}$$

We also have

$$m^0=\rho^0 \tag{2.38}$$

and

$$f_i^0=\rho^0\frac{\partial}{\partial X_A}\frac{\partial\Sigma}{\partial x_{i,A}} . \tag{2.39}$$

With these simplifications the dynamical equation for the c.m. becomes

$$\begin{aligned} \rho^0\ddot{\vec{x}} &= \vec{f}^0+(\vec{p}\cdot\vec{\nabla})\vec{E}(\vec{x})+\dot{\vec{p}}\times\vec{B}(\vec{x}) \\ &\quad +\dot{\vec{x}}\times(\vec{p}\cdot\vec{\nabla})\vec{B}(\vec{x}) . \end{aligned} \tag{2.40}$$

Construction of a rotationally invariant stored energy density $\hat{\Sigma}$ has been discussed previously.[20,29] The most convenient method of guaranteeing the rotational invariance of $\hat{\Sigma}$ is by expressing it as a function of rotationally invariant variables. If $\hat{\Sigma}$ is to be expressed as a polynomial in these variables, regarded as a truncation of an infinite series expansion, then it is sufficient[20] to choose E_{AB} and Λ^{μ}_{A}, defined by

$$E_{AB}=\tfrac{1}{2}(\vec{x}_{,A}\vec{x}_{,B}-\delta_{AB})=E_{BA} , \tag{2.41}$$

$$\Lambda^{\mu}_{A}=R_{iA}y^{\mu}_{i}, \quad \mu=1, 2, \ldots, N-1 , \tag{2.42a}$$

where

$$R_{iA}=x_{i,B}(C^{-1/2})_{BA} , \tag{2.42b}$$

$$C_{AB}=\delta_{AB}+2E_{AB} \tag{2.42c}$$

as the independent rotationally invariant variables. The deletion of the caret from Σ will indicate its dependence on these new variables. The quantity E_{AB} is the finite strain tensor, R_{iA} is the rotation tensor, C_{AB} is the deformation tensor, and Λ^{μ}_{A} is a set of $N-1$ variables which, since it involves the internal coordinates $\vec{y}^{\mu}(\mu\neq 0)$, can be thought of as polarization-like coordinates though we have found it convenient not to introduce the effective charge in the definition. This is avoided since our treatment must include infrared inactive vibration modes that do not possess any associated effective charge.

On the basis of the above discussion we can expand $\rho^0\Sigma$ as

$$\begin{aligned} \rho^0\Sigma(E_{AB}, \Lambda^{\alpha}_{A}) &= \sum_{\alpha,\beta} {}^{(2,0)}H^{\alpha\beta}_{AB}\Lambda^{\alpha}_{A}\Lambda^{\beta}_{B}+\sum_{\alpha} {}^{(1,1)}H^{\alpha}_{ABC}\Lambda^{\alpha}_{A}E_{BC}+{}^{(0,2)}H_{ABCD}E_{AB}E_{CD} \\ &\quad +\sum_{\alpha,\beta,\gamma} {}^{(3,0)}H^{\alpha\beta\gamma}_{ABC}\Lambda^{\alpha}_{A}\Lambda^{\beta}_{B}\Lambda^{\gamma}_{C}+\sum_{\alpha,\beta} {}^{(2,1)}H^{\alpha\beta}_{ABCD}\Lambda^{\alpha}_{A}\Lambda^{\beta}_{B}E_{CD}+\sum_{\alpha} {}^{(1,2)}H^{\alpha}_{ABCDE}\Lambda^{\alpha}_{A}E_{BC}E_{DE} . \end{aligned} \tag{2.43}$$

In Eq. (2.43) the "post"-superscripts α, β, γ individually span the range 1, 2, $\ldots$, $N-1$. The "pre"-superscripts provide a handy designation for the various $\overleftrightarrow{H}$'s. The subscripts are tensor indices and summation is implied for repeated subscripts. Such a convention cannot be applied to the "post"-superscripts. The series expansion has been carried far enough to yield all two field-driving terms for the force equations, as needed for acousto-optic mixing, except those which would contain two c.m.- (acoustic-) type fields, whose contributions would be negligible. The absence of terms in Eq. (2.43) involving ${}^{(1,0)}H_A^\alpha$ and ${}^{(0,1)}H_{AB}$ reflect assumptions that the *natural state* of the medium (the state where external fields and stresses are absent) is unpolarized and unstrained, respectively. Thus ferroelastics and ferroelectrics are excluded from the discussion. The stored-energy function is assumed to be a single-valued function of the values of the independent variables. This excludes materials exhibiting hysteresis from the discussion. It also means that the stored energy is not dependent on the rate of approach to its value. Hence the $\overleftrightarrow{H}$ coefficients are tensor constants which are independent of frequency. They characterize the material medium and so possess the group symmetry of the medium. If S represents a transformation of the group, then[20]

$$H^{\alpha\beta\cdots}_{AB\,CD\cdots} = H^{S(\alpha)S(\beta)\cdots}_{A'B'C'D'\cdots} S_{A'A}S_{B'B}S_{C'C}S_{D'D}\cdots \quad . \tag{2.44}$$

The derivative of the stored-energy density needed for calculation of $\vec{f}^\mu$ ($\mu = 1, 2, \ldots, N-1$) is

$$\frac{\partial\Sigma}{\partial y_i^\mu} = R_{iA}\frac{\partial\Sigma}{\partial\Lambda_A^\mu} \quad . \tag{2.45}$$

The derivative needed for $\vec{f}^0$ is

$$\frac{\partial\Sigma}{\partial x_{i,A}} = x_{i,B}\frac{\partial\Sigma}{\partial E_{AB}} + \sum_{\nu=1}^{N-1} y_j^\nu \frac{\partial\Sigma}{\partial\Lambda_B^\nu}\,\frac{\partial R_{jB}}{\partial x_{i,A}} \quad , \tag{2.46a}$$

$$\begin{aligned}\frac{\partial R_{jB}}{\partial x_{i,A}} = {}& \delta_{ij}(\delta_{AB} - E_{AB} - \cdots) \\ & + \tfrac{1}{2}x_{j,D}\,[-x_{i,D}\delta_{AB} - x_{i,B}\delta_{AD} \\ & + \tfrac{3}{2}(x_{i,D}E_{AB} + x_{i,C}\delta_{AD}E_{CB} \\ & + x_{i,C}\delta_{AB}E_{DC} + x_{i,B}E_{DA}) - \cdots] \,.\end{aligned} \tag{2.46b}$$

In order to bring the equations into a more tractable form we define a deformation variable, the displacement $\vec{u}$, by

$$\vec{u}(\vec{X}, t) = \vec{x}(\vec{X}, t) - \vec{X} \,, \tag{2.47}$$

where the vectors are now referred to a common Cartesian coordinate system. Since the normal photoelastic effect is linear in the elastic variable, we can regard the displacement $\vec{u}$ as small and retain only terms in the expansion linear in $\vec{u}$. For an undistorted material the displacement $\vec{u}$ will be zero; from Eq. (2.47) we see that this means that the designation of the material points, denoted by $\vec{X}$, is just their undistorted positions in the spatial frame. Equation (2.47) leads directly to the following relations,

$$x_{i,A} = \delta_{iA} + u_{i,A} \,, \tag{2.48}$$

$$\frac{\partial y_a^\alpha}{\partial X_B} = \frac{\partial y_a^\alpha}{\partial x_B} + u_{i,B}\, y_{a,i}^\alpha \,, \tag{2.49}$$

needed to simplify the force equations. The relations

$$R_{iA} \cong \delta_{iA} + \tfrac{1}{2}(u_{i,A} - u_{A,i}) \,, \tag{2.50}$$

$$\Lambda_A^\alpha \cong y_A^\alpha + \tfrac{1}{2}\, y_i^\alpha (u_{i,A} - u_{A,i}) \,, \tag{2.51}$$

$$\begin{aligned}\Lambda_{A,B}^\alpha \cong {}& \frac{\partial y_A^\alpha}{\partial x_B} + u_{i,B}\, y_{A,i}^\alpha + \tfrac{1}{2} y_{i,B}^\alpha (u_{i,A} - u_{A,i}) \\ & + \tfrac{1}{2}\, y_i^\alpha (u_{i,AB} - u_{A,iB}) \,,\end{aligned} \tag{2.52}$$

$$E_{AB} \cong \tfrac{1}{2}(u_{A,B} + u_{B,A}) \,, \tag{2.53}$$

correct through terms linear in $\vec{u}$, are also needed. It should be noted that because of the use of a common Cartesian coordinate system in Eq. (2.47) there no longer is a distinction between upper- and lower-case tensor subscripts. We will denote $\partial y_a^\alpha/\partial x_B$ in Eq. (2.49) and (2.52) henceforth by $y_{a,b}^\alpha$.

At this point the independent variable of the force equations (2.31) and (2.40) is still $\vec{X}$. It is more convenient to solve the equations in the spatial rather than material frame. The equations thus need to be transformed to make $\vec{x}$ the independent variable. A caret on the $\vec{u}$ and $\vec{y}^\mu$ functions should now be used to denote their new functional dependence:

$$\hat{\vec{u}}(\vec{x}, t) = \vec{u}(\vec{X}, t) \,, \tag{2.54}$$

$$\hat{\vec{y}}^\alpha(\vec{x}, t) = \vec{y}^\alpha(\vec{X}, t) \quad . \tag{2.55}$$

The time derivatives holding $\vec{X}$ fixed must be transformed to time derivatives holding $\vec{x}$ fixed. To first order in $\hat{\vec{u}}$

$$\dot{\vec{x}} \cong \left(\frac{\partial\hat{\vec{u}}}{\partial t}\right)_{\vec{x}} \,, \tag{2.56}$$

$$\ddot{\vec{x}} \cong \left(\frac{\partial^2\hat{\vec{u}}}{\partial t^2}\right)_{\vec{x}} \,, \tag{2.57}$$

$$\dot{\vec{y}}^\alpha \cong \left(\frac{\partial\hat{\vec{y}}^\alpha}{\partial t}\right)_{\vec{x}} + \hat{\vec{y}}^\alpha_{,i}\left(\frac{\partial\hat{u}_i}{\partial t}\right)_{\vec{x}} \,, \tag{2.58}$$

$$\ddot{\vec{y}}^\alpha \cong \left(\frac{\partial^2\hat{\vec{y}}^\alpha}{\partial t^2}\right)_{\vec{x}} + 2\left(\frac{\partial\hat{u}_i}{\partial t}\right)_{\vec{x}}\left(\frac{\partial\hat{\vec{y}}^\alpha_{,i}}{\partial t}\right)_{\vec{x}} + \hat{\vec{y}}^\alpha_{,i}\left(\frac{\partial^2\hat{u}_i}{\partial t^2}\right)_{\vec{x}} \quad . \tag{2.59}$$

Though from this point on $\hat{\vec{u}}$ and $\hat{\vec{y}}^\alpha$ will be used, we will for simplicity of notation omit the carets. Also, at this point we can identify $\vec{x}$ with $\vec{z}$, the position in the laboratory coordinate system. With the aid of Eqs. (2.45)–(2.59), Eq. (2.31) becomes

$$m^{\mu}\frac{\partial^2 y_i^{\mu}}{\partial t^2}=-2m^{\mu}\frac{\partial u_j}{\partial t}\frac{\partial y_{i,j}^{\mu}}{\partial t}-m^{\mu}\frac{\partial^2 u_j}{\partial t^2}y_{i,j}^{\mu}+q^{\mu}E_i+q^{\mu}\epsilon_{ijk}\frac{\partial u_j}{\partial t}B_k+\sum_{\nu}q^{\mu\nu}y_j^{\nu}E_{i,j}+\sum_{\nu}q^{\mu\nu}\epsilon_{ijk}\frac{\partial y_j^{\nu}}{\partial t}B_k-2\sum_{\beta}{}^{(2,0)}H_{ib}^{\mu\beta}y_b^{\beta}$$

$$-{}^{(1,1)}H_{ibc}^{\mu}u_{b,c}-\sum_{\beta}{}^{(2,0)}H_{ab}^{\mu\beta}(y_b^{\beta}u_{i,a}-y_b^{\beta}\mu_{a,i}+y_j^{\beta}u_{j,b}\delta_{ia}-y_j^{\beta}u_{b,j}\delta_{ia})-3\sum_{\beta,\gamma}{}^{(3,0)}H_{ibc}^{\mu\beta\gamma}y_b^{\beta}y_c^{\gamma}-2\sum_{\beta}{}^{(2,1)}H_{ibcd}^{\mu\beta}y_b^{\beta}u_{c,d}\,,$$

$$\mu=1,2,\ldots,N-1 \qquad (2.60)$$

where ϵ_{ijk} is the usual permutation symbol and Eq. (2.40) becomes

$$m^0\frac{\partial^2 u_i}{\partial t^2}=\sum_{\nu}q^{\nu}y_j^{\nu}E_{i,j}+\sum_{\nu}q^{\nu}\epsilon_{ijk}\frac{\partial y_j^{\nu}}{\partial t}B_k+\sum_{\alpha}{}^{(1,1)}H_{adi}^{\alpha}y_{a,d}^{\alpha}+2\,{}^{(0,2)}H_{abdi}u_{a,bd}+\sum_{\alpha,\beta}{}^{(2,0)}H_{ad}^{\alpha\beta}(y_{a,g}^{\alpha}y_j^{\beta}+y_a^{\alpha}y_{j,g}^{\beta})(\delta_{ij}\delta_{gd}-\delta_{jg}\delta_{id})$$

$$+\sum_{\alpha}{}^{(1,1)}H_{ade}^{\alpha}(y_{a,d}^{\alpha}u_{i,e}+y_a^{\alpha}u_{i,de})+\frac{1}{2}\sum_{\alpha}{}^{(1,1)}H_{adi}^{\alpha}(2y_{a,j}^{\alpha}u_{j,d}+y_j^{\alpha}u_{j,ad}+y_{j,d}^{\alpha}u_{j,a}-y_j^{\alpha}u_{a,jd}-y_{j,d}^{\alpha}u_{a,j})$$

$$+\frac{1}{2}\sum_{\alpha}{}^{(1,1)}H_{dbc}^{\alpha}(y_{j,g}^{\alpha}u_{b,c}+y_j^{\alpha}u_{b,cg})(\delta_{ij}\delta_{gd}-\delta_{jg}\delta_{id})+2\sum_{\alpha,\beta}{}^{(2,1)}H_{abdi}^{\alpha\beta}y_{a,d}^{\alpha}y_b^{\beta}+2\sum_{\alpha}{}^{(1,2)}H_{abcdi}^{\alpha}(y_{a,d}^{\alpha}u_{b,c}+y_a^{\alpha}u_{b,cd})\,. \qquad (2.61)$$

In these two equations only forces involving at most two fields have been retained. All summations over Greek-letter superscripts range over $1, 2, \ldots, N-1$ The first two force terms on the right-hand side of Eq. (2.60) are of kinematic origin, while the next three arise from the interaction energy, and the remainder arise from the stored energy. The first two force terms on the right-hand side of Eq. (2.61) arise from the interaction energy and the remainder arise from the stored energy.

The right-hand sides of the field equations (2.12) and (2.13) must also be expanded in terms of $\vec{u}$. To do this we have, to first order in $\vec{u}$,

$$1/J(\vec{z},t)=1-\vec{\nabla}\cdot\vec{u}(\vec{z},t)\,. \qquad (2.62)$$

Therefore,

$$\epsilon_0\vec{\nabla}\cdot\vec{E}(\vec{z},t)+\vec{\nabla}\cdot\vec{p}(\vec{z},t)=\vec{\nabla}\cdot[\vec{p}(\vec{z},t)\vec{\nabla}\cdot\vec{u}(\vec{z},t)]\,, \qquad (2.63)$$

$$\frac{1}{\mu_0}\vec{\nabla}\times\vec{B}(\vec{z},t)-\epsilon_0\frac{\partial\vec{E}(\vec{z},t)}{\partial t}-\frac{\partial\vec{p}(\vec{z},t)}{\partial t}=-\frac{\partial}{\partial t}[\vec{p}(\vec{z},t)\vec{\nabla}\cdot\vec{u}(\vec{z},t)]+\vec{\nabla}\times\left(\vec{p}(\vec{z},t)\times\frac{\partial\vec{u}(\vec{z},t)}{\partial t}\right)\,. \qquad (2.64)$$

The N-vector matter equations represented by Eqs. (2.60) and (2.61) along with the field equations (2.18), (2.19), (2.63), and (2.64) form the basis for a study of the photoelastic effect.

III. WAVE EQUATION

The photoelastic effect has been studied in at least three types of experiments: The oldest method is the measurement of induced birefringence in a medium subjected to a static uniform load. The other two methods, described above, are Brillouin scattering and acousto-optic scattering. We choose to treat the problem in the form of acousto-optic scattering, though the nonlinear polarization that we derive is equally applicable to the other experimental approaches.

Consider an input optical wave at an angular frequency ω_O and an input acoustic wave at an angular frequency ω_A. These can, in general, mix to give all orders of sum and difference frequencies ($\omega_O\pm\omega_A$, $\omega_O\pm 2\omega_A$, $2\omega_O\pm\omega_A, \ldots$) by interacting one or more times with various orders of nonlinearities. The term photoelastic effect, as used here, will refer to the lowest-order mixing of the two input waves to yield an output frequency of either $\omega_O+\omega_A$ or $\omega_O-\omega_A$. In order to select out a particular frequency component we expand each of the fields $\vec{u}$, $\vec{y}^{\mu}$, $\vec{E}$, and $\vec{B}$ in a Fourier series of the form

$$\vec{Z}(\vec{z},t)=\frac{1}{2}\sum_{m,n=-\infty}^{\infty}\vec{Z}(\vec{z},t;m,n)\,, \qquad (3.1)$$

where

$$\vec{Z}(\vec{z},t;m,n)=\vec{Z}(\vec{z};m,n)\,e^{-i(m\omega_O+n\omega_A)t}\,, \qquad (3.2)$$

$$\vec{Z}(\vec{z};-m,-n)=\vec{Z}^*(\vec{z};m,n)\,. \qquad (3.3)$$

The solutions for acousto-optic scattering are $m=1$, $n=\pm 1$. It is fortunate that an iterative technique can be used for solution of this problem, that is, the $(m,n)=(1,\pm 1)$ solutions can be obtained from a *linear* problem in terms of the (1, 0) and (0, 1) solutions which have been obtained[20] separately and independently of the $(1,\pm 1)$ solutions. The iterative technique can, in fact, be applied to a great variety of nonlinear optical problems of current interest. It is limited by two assumptions: (a) Depletion of the input of pump waves is negligible, that is, lower-order (in m, n indices) fields enter higher-order equations only as waves of constant amplitude; (b) variation of the linear properties of the medium by the nonlinear interaction is negligible for the solution of the linear problem, or more generally, products of l fields can be neglected in the solution of a problem of order $k=|m|+|n|$ for all $l>k$. The first assumption will be violated if a combination of the interaction strength and interaction length is too large while the second will be violated in interactions such as the self-focusing of light.

The $\vec{u}$, $\vec{y}^{\mu}$, $\vec{E}$, and $\vec{B}$ fields in the form of Eqs. (3.1)–(3.3) can now be substituted into Eqs. (2.18), (2.19), (2.60), (2.61), (2.63), and (2.64). For notational convenience we consider the $(m, n) = (1, 1)$ problem only; the $(1, -1)$ problem is handled analogously. We obtain

$$\vec{\nabla} \cdot \vec{B}(\vec{z}; 1, 1) = 0 , \tag{3.4}$$

$$\epsilon_0 \vec{\nabla} \cdot \vec{E}(\vec{z}; 1, 1) + \sum_{\alpha} q^{\alpha} \vec{\nabla} \cdot \vec{y}^{\alpha}(\vec{z}; 1, 1) = Q(\vec{z}; 1, 1) , \tag{3.5}$$

$$(1/\mu_0) \vec{\nabla} \times \vec{B}(\vec{z}; 1, 1) + i\epsilon_0 \omega_B \vec{E}(\vec{z}; 1, 1) + i\omega_B \sum_{\alpha} q^{\alpha} \vec{y}^{\alpha}(\vec{z}; 1, 1) = \vec{I}(\vec{z}; 1, 1) , \tag{3.6}$$

$$\vec{\nabla} \times \vec{E}(\vec{z}; 1, 1) - i\omega_B \vec{B}(\vec{z}; 1, 1) = 0 , \tag{3.7}$$

$$-\omega_B^2 m^{\mu} y_i^{\mu}(\vec{z}; 1, 1) - q^{\mu} E_i(\vec{z}; 1, 1) + 2\sum_{\beta} {}^{(2,0)}H_{ib}^{\mu\beta} y_b^{\beta}(\vec{z}; 1, 1) + {}^{(1,1)}H_{ibc}^{\mu} u_{b,c}(\vec{z}; 1, 1) = F_i^{\mu}(\vec{z}; 1, 1) , \tag{3.8}$$

$$-\omega_B^2 m^0 u_i(\vec{z}; 1, 1) - \sum_{\alpha} {}^{(1,1)}H_{adi}^{\alpha} y_{a,d}^{\alpha}(\vec{z}; 1, 1) - 2\, {}^{(0,2)}H_{abdi} u_{a,bd}(\vec{z}; 1, 1) = G_i(\vec{z}:1, 1) , \tag{3.9}$$

where

$$\begin{aligned} F_i^{\mu}(\vec{z}; 1, 1) = \frac{1}{2}\Big(& 2\omega_O \omega_A m^{\mu} u_j(0, 1) y_{i,j}^{\mu}(1, 0) + \omega_A^2 m^{\mu} u_j(0, 1)\, y_{i,j}^{\mu}(1, 0) \\ & - i\omega_A q^{\mu} \epsilon_{ijk} u_j(0, 1) B_k(1, 0) + \sum_{\alpha} q^{\mu\alpha} y_j^{\alpha}(1, 0) E_{i,j}(0, 1) - i\omega_O \sum_{\alpha} q^{\mu\alpha} \epsilon_{ijk} y_j^{\alpha}(1, 0) B_k(0, 1) \\ & - \sum_{\beta} {}^{(2,0)}H_{ab}^{\mu\beta} [y_b^{\beta}(1, 0) u_{i,a}(0, 1) - y_b^{\beta}(1, 0) u_{a,i}(0, 1) - y_j^{\beta}(1, 0) u_{b,j}(0, 1) \delta_{ia} + y_j^{\beta}(1, 0) u_{j,b}(0, 1) \delta_{ia}] \\ & - 3 \sum_{\beta,\gamma} {}^{(3,0)}H_{ibc}^{\mu\beta\gamma} y_b^{\beta}(1, 0) y_c^{\gamma}(0, 1) - 2\sum_{\beta} {}^{(2,1)}H_{ibcd}^{\mu\beta} y_b^{\beta}(1, 0) u_{c,d}(0, 1) \\ & + \text{interchange of } (1, 0) \text{ and } (0, 1)\Big) e^{i(\vec{k}_O + \vec{k}_A) \cdot \vec{z}} = F_i^{\mu}(1, 1) e^{i(\vec{k}_O + \vec{k}_A) \cdot \vec{z}}, \end{aligned} \tag{3.10}$$

$$G_i(\vec{z}; 1, 1) = G_i(1, 1) e^{i(\vec{k}_O + \vec{k}_A) \cdot \vec{z}} , \tag{3.11}$$

$$\begin{aligned} I_i(\vec{z}; 1, 1) = \tfrac{1}{2} \{ & i[\omega_O + \omega_A] \sum_{\alpha} q^{\alpha} y_i^{\alpha}(1, 0) u_{j,j}(0, 1) + i\omega_A [\delta_{il}\delta_{jm} - \delta_{im}\delta_{jl}] [\sum_{\alpha} q^{\alpha} y_m^{\alpha}(1, 0) u_{l,j}(0, 1) \\ & + \sum_{\alpha} q^{\alpha} y_{m,j}^{\alpha}(1, 0) u_l(0, 1)] + \text{interchange of } (1, 0) \text{ and } (0, 1)\} e^{i(\vec{k}_A + \vec{k}_O) \cdot \vec{z}} \\ = {} & I_i(1, 1) e^{i(\vec{k}_A + \vec{k}_O) \cdot \vec{z}} , \end{aligned} \tag{3.12}$$

$$\begin{aligned} Q(\vec{z}; 1, 1) = {} & \tfrac{1}{2} [\sum_{\alpha} q^{\alpha} y_{i,i}^{\alpha}(1, 0) u_{j,j}(0, 1) + \sum_{\alpha} q^{\alpha} y_i^{\alpha}(1, 0) u_{j,ji}(0, 1) + \text{interchange of } (1, 0) \text{ and } (0, 1)] e^{i(\vec{k}_A + \vec{k}_O) \cdot \vec{z}} \\ = {} & Q(1, 1) e^{i(\vec{k}_A + \vec{k}_O) \cdot \vec{z}} , \end{aligned} \tag{3.13}$$

$$\omega_B = \omega_O + \omega_A . \tag{3.14}$$

In Eqs. (3.10)–(3.13) the derivative notation now means multiplication by the product of the imaginary unit and the appropriate wave vector. In Eqs. (3.4)–(3.9) the linear terms have been grouped on the left-hand side and the nonlinear driving terms, involving products of two fields in this case, placed on the right-hand side. It should be noted that in the definitions of the nonlinear driving terms in Eqs. (3.10)–(3.13), interchange of $(1, 0) \longleftrightarrow (0, 1)$ requires the concurrent interchange $\omega_O \longleftrightarrow \omega_A$ wherever the frequencies appear. The explicit expression for $\vec{G}(1, 1)$ was omitted since it will not be needed. The dependence on $\vec{z}$ given in Eqs. (3.10)–(3.13) has arisen from the assumption that the input optic and acoustic fields are plane waves of constant amplitude characterized by wave vectors $\vec{k}_O$ and $\vec{k}_A$, respectively. We are thus neglecting the depletion of the input optic and acoustic waves in the photoelastic interaction.

It should be noted at this point that the two scalar Maxwell equations (3.4) and (3.5) are redundant. Equation (3.4) is just the divergence of Eq. (3.7) and Eq. (3.5) combined with Eq. (3.6), with the time dependence retained, gives a relation that can be called the conservation of nonlinear charge:

$$\frac{\partial Q(\vec{z}, t; 1, 1)}{\partial t} + \vec{\nabla} \cdot \vec{I}(\vec{z}, t; 1, 1) = 0 . \tag{3.15}$$

The solution of the system of Eqs. (3.6)–(3.9) can be simplified by observing that because the output wave is an electromagnetic wave, the linear term in Eq. (3.8) involving ${}^{(1,1)}\overleftrightarrow{H}$ is of order $(v_A/v_O)^2 \sim 10^{-10}$ times the magnitude of the dominant terms in the equation and so may be neglected (v_A and v_O denote velocities of acoustic and optic waves in the medium). Note that the neglect of the ${}^{(1,1)}\overleftrightarrow{H}$ term in Eq. (3.8) uncouples the equation for $\vec{u}$ from the remainder of the equations and shows that the $\vec{G}(1,1)$ function plays an insignificant role in driving the output optical field. Define $\Upsilon_{ab}^{\alpha\beta}(\omega)$ by

$$\epsilon_0 \sum_\beta \Upsilon_{ab}^{\alpha\beta}(\omega)[2\,{}^{(2,0)}H_{bc}^{\beta\gamma} - \omega^2 m^\beta \delta^{\beta\gamma}\delta_{bc}] = \delta^{\alpha\gamma}\delta_{ac} . \tag{3.16}$$

Hence Eq. (3.8) gives

$$y_j^\nu(\vec{z};1,1) = \epsilon_0 \sum_\mu \Upsilon_{ji}^{\nu\mu}(\omega_B)[q^\mu E_i(\vec{z};1,1) + F_i^\mu(\vec{z};1,1)] . \tag{3.17}$$

Since $\Upsilon_{ij}^{\nu\mu}$ is the inverse of a quantity which is symmetric upon simultaneous interchange of $(i \longleftrightarrow j)$ and $(\nu \longleftrightarrow \mu)$, we have

$$\Upsilon_{ij}^{\nu\mu}(\omega) = \Upsilon_{ji}^{\mu\nu}(\omega) . \tag{3.18}$$

From Eqs. (3.6) and (3.17) we see that the total linear susceptibility is given by

$$\chi_{ij}(\omega) = \sum_{\nu,\mu} q^\nu \Upsilon_{ij}^{\nu\mu}(\omega) q^\mu . \tag{3.19}$$

Equations (3.6), (3.7), and (3.17) can be combined to yield the wave equation with a driving term

$$(c/\omega_B)^2[E_{j,ij}(\vec{z};1,1) - E_{i,jj}(\vec{z};1,1)] - \kappa_{ij}(\omega_B)E_j(\vec{z};1,1) = \mathcal{P}_i(\vec{z};1,1)/\epsilon_0 , \tag{3.20}$$

where $\kappa_{ij}(\omega)$ is the dielectric tensor given by

$$\kappa_{ij}(\omega) = \delta_{ij}(\omega) + \chi_{ij}(\omega) \tag{3.21}$$

and $\mathcal{P}_i(\vec{z};1,1)$ is the nonlinear driving polarization given by

$$\mathcal{P}_i(\vec{z};1,1) = \epsilon_0 \sum_{\nu,\mu} q^\nu \Upsilon_{ij}^{\nu\mu}(\omega_B) F_j^\mu(\vec{z};1,1) + \frac{iI_i(\vec{z};1,1)}{\omega_B} = \mathcal{P}_i(1,1)\, e^{i(\vec{k}_O + \vec{k}_A)\cdot\vec{z}} . \tag{3.22}$$

The general solution of the inhomogeneous equation (3.20) is the sum of the general solution of the homogeneous equation, called the free wave, and a particular solution of the inhomogeneous equation, called the forced wave. Each of these terms can contain parts corresponding to the ordinary and extraordinary waves. Usually only one of these can be phase matched at a time. Phase matching here is synonymous with satisfying the Bragg condition. We thus wish to consider only one of these waves in a given geometry. It must be one of the eigenvectors of the free wave and only the projection of the forced wave on this eigenvector will contribute to the phase-matched or Bragg-scattered output.

The plane-wave eigenvectors of a free-wave electric field $\vec{\mathcal{E}}^{(\theta)}$ propagating in the direction $\vec{s}$ with a refractive index n must satisfy

$$[\delta_{ij} - s_i s_j]\,\mathcal{E}_j^{(\theta)} = (1/n^2)\kappa_{ij}(\omega)\mathcal{E}_j^{(\theta)} , \tag{3.23}$$

where

$$\vec{s} = \vec{k}/|\vec{k}| , \tag{3.24}$$

$$n = c|\vec{k}|/\omega , \tag{3.25}$$

$\vec{k}$ is the wave vector of a free wave and θ refers to the three possible eigenvectors corresponding to the ordinary wave, the extraordinary wave, and the nonpropagating solution (infinite refractive index). Since the electric field eigenvectors are not an orthogonal set, it is convenient to introduce also the electric displacement eigenvectors $\vec{\mathfrak{D}}^{(\theta)}$. They satisfy

$$[\delta_{ij} - s_i s_j][\kappa^{-1}(\omega)]_{jk}\mathfrak{D}_k^{(\theta)} = (1/n^2)\mathfrak{D}_i^{(\theta)} \tag{3.26}$$

and are related to $\vec{\mathcal{E}}^{(\theta)}$ by

$$\mathfrak{D}_i^{(\theta)} = \kappa_{ij}(\omega)\mathcal{E}_j^{(\theta)} . \tag{3.27}$$

Though the electric displacement eigenvectors also do not form an orthogonal set, $\vec{\mathcal{E}}^{(\theta)}$ and $\vec{\mathfrak{D}}^{(\theta)}$ taken together form a biorthogonal set[30] whose normalization can be chosen so that

$$\vec{\mathcal{E}}^{(\theta)} \cdot \vec{\mathfrak{D}}^{(\varphi)} = \delta^{\theta\varphi} . \tag{3.28}$$

Denoting the free wave that is phase matchable by ξ we have

$$\vec{E}(\vec{z};1,1)_{\text{free}} = C^{(\xi)}\,\vec{\mathcal{E}}^{(\xi)}(\vec{s}_B,\omega_B)\,e^{i\vec{k}_B\cdot\vec{z}} , \tag{3.29}$$

where $C^{(\xi)}$ is an arbitrary scaler constant, $\vec{k}_B$ is the wave vector of the free wave, and $\vec{s}_B$ is defined by

$$\vec{s}_B = \vec{k}_B/|\vec{k}_B| . \tag{3.30}$$

The forced wave can be expressed[20] in terms of the eigenvectors of the free-wave electric field that propagates in the direction of the forced wave as

$$\vec{E}(\vec{z};1,1)_{\text{forced}} = \sum_{\varphi=1}^{3} \frac{\vec{\mathcal{E}}^{(\varphi)}(\vec{s}_F,\omega_B)\,\vec{\mathcal{E}}^{(\varphi)}(\vec{s}_F,\omega_B)\cdot\vec{\mathcal{P}}(1,1)}{\epsilon_0(|\vec{k}_O+\vec{k}_A|^2/|\vec{k}_B|^2 - 1)} \times e^{i(\vec{k}_O+\vec{k}_A)\cdot\vec{z}} , \tag{3.31}$$

where

$$\vec{s}_F = (\vec{k}_O + \vec{k}_A)/|\vec{k}_O + \vec{k}_A| . \tag{3.32}$$

Of the three terms in the forced-wave expansion only the $\varphi = \xi$ term need be retained.

If the diagonal elements of the dielectric tensor in its principal coordinate system are denoted by κ_1, κ_2, κ_3, then in this system

$$\mathcal{E}_i^{(\xi)} = s_i/(n_\xi^2 - \kappa_i)N_\xi , \tag{3.33}$$

$$\mathfrak{D}_i^{(\xi)} = \kappa_i s_i/(n_\xi^2 - \kappa_i)N_\xi , \tag{3.34}$$

$$N_\xi = \left(\sum_{i=1}^{3} \frac{s_i^2\kappa_i}{(n_\xi^2 - \kappa_i)^2}\right)^{1/2} . \tag{3.35}$$

In Eqs. (3.33)–(3.35) the summation convention over repeated indices is not followed. Equations (3.33)–(3.35) are useful for biaxial crystal classes and propagation in general directions. For propagation in principal planes, Eqs. (3.23) and (3.26) should be re-solved. The eigenvectors for uniaxial crystal classes can be found by a careful limiting procedure from Eqs. (3.33)–(3.35) or by a direct procedure given previously.[20]

Since we are interested in the radiation in directions close to that at which phase matching and hence where a fairly intense output occurs, we can neglect the "reflected" electric field at the frequency ω_B in the boundary condition at the input surface of the material medium. We thus require that the projection of $\vec{E}(\vec{z};1,1)$ on $\vec{\mathcal{E}}^{(\xi)}(\vec{s}_F,\omega_B)$, which is the scalar product of $\vec{E}(\vec{z};1,1)$ with $\vec{\mathfrak{D}}^{(\mathcal{E})}(\vec{s}_F,\omega_B)$, be zero at the plane input surface. Hence

$$\vec{E}(\vec{z}_P;1,1)\cdot\vec{\mathfrak{D}}^{(\xi)}(\vec{s}_F,\omega_B)$$

$$=0=C^{(\xi)}\vec{\mathfrak{D}}^{(\xi)}(\vec{s}_F,\omega_B)\cdot\vec{\mathcal{E}}^{(\xi)}(\vec{s}_B,\omega_B)\,e^{i\vec{k}_B\cdot\vec{z}_P}+\frac{\vec{\mathcal{E}}^{(\xi)}(\vec{s}_F,\omega_B)\cdot\vec{\mathcal{P}}(1,1)e^{i(\vec{k}_O+\vec{k}_A)\cdot\vec{z}_P}}{\epsilon_0(|\vec{k}_O+\vec{k}_A|^2/|\vec{k}_B|^2-1)}\,,\qquad(3.36)$$

where $\vec{z}_P$ are the coordinates of the plane input surface, the origin of them being taken to lie in the plane. Since Eq. (3.36) is good only near phase matching, it is a good approximation to take

$$\vec{\mathfrak{D}}^{(\xi)}(\vec{s}_F,\omega_B)\cdot\vec{\mathcal{E}}^{(\xi)}(\vec{s}_B,\omega_B)=1$$

even though the propagation directions are slightly different. Two conditions result from Eq. (3.36):

$$(\Delta\vec{k}_B)_t=(\vec{k}_B-\vec{k}_O-\vec{k}_A)_t=0\,,\qquad(3.37)$$

where t stands for the components tangential to the input plane, and

$$C^{(\xi)}=-\frac{\vec{\mathcal{E}}^{(\xi)}(\vec{s}_F,\omega_B)\cdot\vec{\mathcal{P}}(1,1)}{\epsilon_0(|\vec{k}_O+\vec{k}_A|^2/|\vec{k}_B|^2-1)}\,.\qquad(3.38)$$

Letting n denote the component of a vector along the inward normal to the input surface, we have near phase matching

$$\vec{E}(\vec{z};1,1)=(-2i/\epsilon_0)\vec{\mathcal{E}}^{(\xi)}\vec{\mathcal{E}}^{(\xi)}\cdot\vec{\mathcal{P}}(1,1)\times(|\vec{k}_O+\vec{k}_A|^2/|\vec{k}_B|^2-1)^{-1}\times\sin(\tfrac{1}{2}\Delta\vec{k}_{Bn}\cdot\vec{z})\,e^{i(\vec{k}_B+\vec{k}_O+\vec{k}_A)\cdot\vec{z}/2},\qquad(3.39)$$

$$\vec{H}(\vec{z};1,1)=(-2ic^2/\omega_B)\vec{k}_B\times\vec{\mathcal{E}}^{(\xi)}\vec{\mathcal{E}}^{(\xi)}\cdot\vec{\mathcal{P}}(1,1)\times(|\vec{k}_O+\vec{k}_A|^2/|\vec{k}_B|^2-1)^{-1}\times\sin(\tfrac{1}{2}\Delta\vec{k}_{Bn}\cdot\vec{z})\,e^{i(\vec{k}_B+\vec{k}_O+\vec{k}_A)\cdot\vec{z}/2},\qquad(3.40)$$

where terms of order $(\vec{k}_B\cdot\vec{z})^{-1}$ have been neglected. The Poynting vector is thus

$$\vec{S}(\vec{z};1,1)=\frac{2c^2k_B^4}{\epsilon_0\omega_B}\,[(\vec{\mathcal{E}}^{(\xi)})^2\vec{k}_B-(\vec{\mathcal{E}}^{(\xi)}\cdot\vec{k}_B)\vec{\mathcal{E}}^{(\xi)}]\times\left|\frac{\vec{\mathcal{E}}^{(\xi)}\cdot\vec{\mathcal{P}}(1,1)}{(\vec{k}_B+\vec{k}_O+\vec{k}_A)\cdot\Delta\vec{k}_{Bn}}\right|^2\sin^2(\tfrac{1}{2}\Delta\vec{k}_{Bn}\cdot\vec{z})\,.\qquad(3.41)$$

This expression applies within the material medium at distances near enough to the input surface so that pump depletion is negligible and at directions close to the phase-matching direction. Except for the latter condition Eq. (3.41) applies to input waves having arbitrary orientations with respect to the anisotropic medium. Phase matching occurs in this plane-wave geometry when

$$(\Delta\vec{k}_B)_n=(\vec{k}_B-\vec{k}_O-\vec{k}_A)_n=0\,,\qquad(3.42)$$

since the boundary condition has required the tangential components to vanish already. The Poynting vector remains finite and in fact reaches its maximum under this condition.

IV. SYMMETRY OF NONLINEAR POLARIZATION

It is apparent from the wave equation (3.20) or the Poynting vector given in Eq. (3.41) that the interaction strength is governed by the nonlinear driving polarization $\vec{\mathcal{P}}(1,1)$ given by Eq. (3.22) with (3.10) and (3.12). By estimating magnitudes of the various terms in $\vec{F}^{\mu}(1,1)$ and $\vec{I}(1,1)$ we find that for the frequencies used in photoelastic studies and for crystals presently studied only force terms in $\vec{F}^{\mu}(1,1)$ arising from the stored energy and the first term given in $\vec{I}(1,1)$ are significant (see the Appendix). Therefore

$$\begin{aligned}\mathcal{P}_i(1,1)=&-(\tfrac{1}{2}\epsilon_0)\sum_{\rho,\mu,\beta}q^{\rho}\Upsilon^{\rho\mu}_{im}(\omega_\beta)\big({}^{(2,0)}H^{\mu\beta}_{lb}\delta_{mk}\\&-{}^{(2,0)}H^{\mu\beta}_{kb}\delta_{ml}\,{}^{(2,0)}H^{\mu\beta}_{mk}\delta_{bl}+{}^{(2,0)}H^{\mu\beta}_{ml}\delta_{bk}\\&+2\,{}^{(2,1)}H^{\mu\beta}_{mbkl}\big)y^{\beta}_b(1,0)\mu_{k,l}(0,1)\\&-3\epsilon_0\sum_{\rho,\mu,\beta,\gamma}q^{\rho}\Upsilon^{\rho\mu}_{im}(\omega_B)\,{}^{(3,0)}H^{\mu\beta\gamma}_{mbc}y^{\beta}_b(1,0)y^{\gamma}_c(0,1)\\&-\tfrac{1}{2}\delta_{ib}\delta_{kl}\sum_{\beta}q^{\beta}y^{\beta}_b(1,0)u_{k,l}(0,1).\end{aligned}\qquad(4.1)$$

Solutions of the linear problems $(m,n)=(1,0)$ and $(0,1)$ in this notation are readily obtainable from Eqs. (3.4)–(3.9), with the nonlinear driving functions set equal to zero, and have been discussed previously.[20] The results relevant to the present development are

$$y^{\beta}_b(1,0)=\epsilon_0\textstyle\sum_{\nu}\Upsilon^{\beta\gamma}_{bj}(\omega_0)q^{\nu}E_j(1,0),\qquad(4.2)$$

$$y^{\gamma}_c(0,1)=-\epsilon_0\sum_{\theta,\varphi}\left(\delta_{cs}\delta^{\gamma\theta}-\sum_{\eta}\frac{\Upsilon^{\gamma\eta}_{cr}(\omega_A)q^{\eta}q^{\theta}a_ra_s}{a_p\kappa_{pq}(\omega_A)a_q}\right)\times\Upsilon^{\theta\varphi}_{se}(\omega_A)\,{}^{(1,1)}H^{\varphi}_{ekl}\,u_{k,l}(0,1),\qquad(4.3)$$

where

$$\vec{a}=\vec{k}_A/|\vec{k}_A|\,,\qquad(4.4)$$

and terms in Eq. (4.3) of order $(v_A/v_0)^2\approx10^{-10}$ times the others have been dropped. The term involving $\vec{a}$ arises from the piezoelectrically created longitudinal electric field which is in the direction of $\vec{a}$. As there does not seem to be any way of preventing this field by the use of boundary conditions in the usual manner the acousto-optic scattering is done, we have included it here. If photoelasticity is studied by the use of static loads on crystals, boundary conditions can be imposed which will eliminate the piezoelectrically created electric field and hence this term would then be deleted. The expression we will obtain for $\vec{\mathcal{P}}(1,1)$ by the use of Eq. (4.3) will be specific to acoustic waves because of the presence of $\vec{a}$. Because of its presence the static limit cannot be obtained directly; it can be obtained only through the solution of an appropriate boundary-value problem. Equation (4.1) can now be expressed as

$$\mathcal{P}_i(1,1)=\epsilon_0\left(\chi_{i\,j\,kl}^{\omega_B\omega_O\omega_A}-\frac{2d_{ijr}^{\omega_B\omega_O\omega_A}a_r a_s e_{skl}^{\omega_A}}{\epsilon_0 a_p\kappa_{pq}(\omega_A)a_q}\right)\times E_j(1,0)\,u_{k,l}(0,1), \qquad (4.5)$$

where

$$\begin{aligned}\chi_{i\,j\,kl}^{\omega_B\omega_O\omega_A}=&-\tfrac{1}{2}\chi_{ij}(\omega_O)\,\delta_{kl}\\&-(\tfrac{1}{2}\epsilon_0)\sum_{\rho,\mu,\beta,\nu}\Upsilon_{im}^{\rho\mu}(\omega_B)\Upsilon_{bj}^{\beta\nu}(\omega_O)\,q^\nu q^\rho\\&\times\left({}^{(2,0)}H_{lb}^{\mu\beta}\,\delta_{mk}+{}^{(2,0)}H_{ml}^{\mu\beta}\,\delta_{bk}\right.\\&\left.-{}^{(2,0)}H_{kb}^{\mu\beta}\,\delta_{ml}-{}^{(2,0)}H_{mk}^{\mu\beta}\,\delta_{bl}+2\,{}^{(2,1)}H_{mbkl}^{\mu\beta}\right)\\&+3\epsilon_0^2\sum_{\rho,\nu,\beta,\mu,\gamma,\varphi}\Upsilon_{im}^{\rho\mu}(\omega_B)\Upsilon_{bj}^{\beta\gamma}(\omega_O)\\&\times\Upsilon_{ce}^{\gamma\varphi}(\omega_A)q^\nu q^\rho\,{}^{(3,0)}H_{mbc}^{\mu\beta\gamma}\,{}^{(1,1)}H_{ekl}^{\varphi},\end{aligned} \qquad (4.6)$$

$$\begin{aligned}d_{i\,j\,r}^{\omega_B\omega_O\omega_A}=&-\tfrac{3}{2}\epsilon_0^2\sum_{\rho,\mu,\beta,\nu,\eta,\gamma}\Upsilon_{im}^{\rho\mu}(\omega_B)\Upsilon_{bj}^{\beta\nu}(\omega_O)\\&\times\Upsilon_{cr}^{\gamma\eta}(\omega_A)q^\nu q^\eta q^\rho\,{}^{(3,0)}H_{mbc}^{\mu\beta\gamma},\end{aligned} \qquad (4.7)$$

$$e_{skl}^{\omega_A}=-\epsilon_0\sum_{\theta,\varphi}\Upsilon_{se}^{\theta\varphi}(\omega_A)q^\theta\,{}^{(1,1)}H_{ekl}^{\varphi}\,. \qquad (4.8)$$

The $\chi_{ijkl}^{\omega_B\omega_O\omega_A}$ is the true fourth-rank photoelasticity tensor representing what we will call the direct photoelastic effect. The second term in Eq. (4.5), which we will call the indirect photoelastic effect, exists only in piezoelectric crystals. The quantity $e_{jkl}^{\omega_A}$ is the conventional piezoelectric tensor relating the strain S_{kl} to the polarization $\vec{P}$ by

$$P_j(0,1)=\sum\nolimits_\gamma q^\gamma y_j^\gamma(0,1)=\epsilon_0\chi_{jk}(\omega_A)E_k(0,1)+e_{jkl}^{\omega_A}S_{kl}(0,1), \qquad (4.9)$$

$$S_{kl}=\tfrac{1}{2}(u_{k,l}+u_{l,k})\,. \qquad (4.10)$$

The quantity $d_{i\,j\,k}^{\omega_B\omega_O\omega_A}$ is the direct electro-optic tensor expressed in the form of the optical mixing tensor. It relates two input electric field amplitudes to a nonlinear polarization by

$$\mathcal{P}_i^{\omega_B}=D\epsilon_0 d_{i\,j\,k}^{\omega_B\omega_O\omega_A}E_j^{\omega_O}E_k^{\omega_A}, \qquad (4.11)$$

where

$$\begin{aligned}D&=1\quad\text{if }\omega_O=\omega_A\\&=2\quad\text{if }\omega_O\neq\omega_A.\end{aligned} \qquad (4.12)$$

It is related to the more conventional Pockels electro-optic tensor r_{ijk} by

$$d_{i\,j\,k}^{\omega_B\omega_O\omega_A}=-\tfrac{1}{4}\kappa_{il}(\omega_B)r_{lmk}\kappa_{mj}(\omega_O), \qquad (4.13)$$

where the Pockels electro-optic tensor relates the change in the inverse dielectric tensor to the electric field causing the change by

$$(\delta\kappa^{-1})_{ij}=r_{ijk}E_k\,. \qquad (4.14)$$

The factor of 4 in Eq. (4.13) arises from $D=2$ and from $d_{i\,j\,k}^{\omega_B\omega_O\omega_A}$ relating to one of two $(\omega_O\pm\omega_A)$ Fourier components while r_{lmk} is tradionally quoted as a static value $(\omega_A=0)$.

Because of the presence of the acoustic wave-vector direction $\vec{a}$ in the term in Eq. (4.5) representing the indirect photoelastic effect, it does not transform as a simple fourth-rank tensor and hence cannot be represented as such. It is instead a fourth-rank tensor function of the acoustic wave-vector direction. Because of this the indirect photoelastic effect has *different* symmetry than the direct effect. Furthermore, the value of its effective susceptibility in Eq. (4.5) can vary even for a particular set of the four tensor indices depending on the direction of the acoustic wave vector. The indirect effect does possess symmetry upon interchange of k and l and also upon interchange of the i and j indices provided ω_B and ω_O are also interchanged simultaneously. Since these frequencies are so close together, for all practical purposes there is symmetry upon interchange simply of i and j. Table I lists the possible nonzero components of the indirect photoelastic effect that can arise from an arbitrary direction of the acoustic wave vector for the various crystal classes.

The indirect photoelastic effect can make a significant contribution to the total effect for certain geometries in some nonferroelectric crystals such as α-HIO_3. For instance, for an acoustic wave propagating in the [011] direction in α-HIO_3, $|p_{1212}|_{\text{indirect}}=0.009$ or about 10% of the reported value[31] of p_{1212}. For an acoustic wave propagating in the [110] direction, $|p_{3232}|_{\text{indirect}}=0.022$, a still larger value. Since p_{3232} has not yet been measured a percentage comparison cannot be made. For an acoustic wave propagating in the [111] direction, $|p_{3212}|_{\text{indirect}}=0.013$, a photoelastic coefficient whose direct contribution vanishes because of symmetry requirements. These values are based on measured values for the piezoelectric coefficients,[32] constant-stress dielectric constants[33] altered through the piezoelectric coefficient to give the constant-strain dielectric constants appropriate to high-frequency acoustic waves, electro-optic coefficients,[34] and optical dielectric constants.[35] Though this theory as presently formulated does not apply to ferroelectrics, it is clear that an in-

TABLE I. Possible components of the *indirect* photoelastic effect in contracted notation for various crystal classes. X indicates components which can be nonzero for a general acoustic wave-vector direction. Isotropic substances, cubic classes $m3$, 432, and $m3m$, hexagonal classes $6/m$ and $6/mmm$, trigonal classes $\bar{3}m$ and $\bar{3}$, tetragonal classes $4/mmm$ and $4/m$, orthorhombic class mmm, monoclinic class $2/m$, and triclinic class $\bar{1}$ are nonpiezoelectric and so possess no indirect photoelastic effect.

Cubic 23 or $\bar{4}3m$

$$\begin{bmatrix} 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&X&X&X\\ 0&0&0&X&X&X\\ 0&0&0&X&X&X \end{bmatrix}$$

Hexagonal $\bar{6}m2$ or $\bar{6}$

$$\begin{bmatrix} X&X&0&0&0&X\\ X&X&0&0&0&X\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ X&X&0&0&0&X \end{bmatrix}$$

Hexagonal $6mm$ or 6

$$\begin{bmatrix} X&X&X&X&X&0\\ X&X&X&X&X&0\\ X&X&X&X&X&0\\ X&X&X&X&X&0\\ X&X&X&X&X&0\\ 0&0&0&0&0&0 \end{bmatrix}$$

Hexagonal 622

$$\begin{bmatrix} 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&X&X&0\\ 0&0&0&X&X&0\\ 0&0&0&0&0&0 \end{bmatrix}$$

Trigonal $3m$ or 3

$$\begin{bmatrix} X&X&X&X&X&X\\ X&X&X&X&X&X\\ X&X&X&X&X&X\\ X&X&X&X&X&X\\ X&X&X&X&X&X\\ X&X&X&X&X&X \end{bmatrix}$$

Trigonal 32

$$\begin{bmatrix} X&X&0&X&X&X\\ X&X&0&X&X&X\\ 0&0&0&0&0&0\\ X&X&0&X&X&X\\ X&X&0&X&X&X\\ X&X&0&X&X&X \end{bmatrix}$$

Tetragonal $\bar{4}2\,m$

$$\begin{bmatrix} 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&X&X&X\\ 0&0&0&X&X&X\\ 0&0&0&X&X&X \end{bmatrix}$$

Tetragonal 422

$$\begin{bmatrix} 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&X&X&0\\ 0&0&0&X&X&0\\ 0&0&0&0&0&0 \end{bmatrix}$$

Tetragonal $4mm$ or 4

$$\begin{bmatrix} X&X&X&X&X&0\\ X&X&X&X&X&0\\ X&X&X&X&X&0\\ X&X&X&X&X&0\\ X&X&X&X&X&0\\ 0&0&0&0&0&0 \end{bmatrix}$$

Tetragonal $\bar{4}$

$$\begin{bmatrix} X&X&0&X&X&X\\ X&X&0&X&X&X\\ 0&0&0&0&0&0\\ X&X&0&X&X&X\\ X&X&0&X&X&X\\ X&X&0&X&X&X \end{bmatrix}$$

Orthorhombic $mm2$

$$\begin{bmatrix} X&X&X&X&X&0\\ X&X&X&X&X&0\\ X&X&X&X&X&0\\ X&X&X&X&X&0\\ X&X&X&X&X&0\\ 0&0&0&0&0&0 \end{bmatrix}$$

Orthorhombic 222

$$\begin{bmatrix} 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&X&X&X\\ 0&0&0&X&X&X\\ 0&0&0&X&X&X \end{bmatrix}$$

Monoclinic m or 2

$$\begin{bmatrix} X&X&X&X&X&X\\ X&X&X&X&X&X\\ X&X&X&X&X&X\\ X&X&X&X&X&X\\ X&X&X&X&X&X\\ X&X&X&X&X&X \end{bmatrix}$$

Triclinic 1

$$\begin{bmatrix} X&X&X&X&X&X\\ X&X&X&X&X&X\\ X&X&X&X&X&X\\ X&X&X&X&X&X\\ X&X&X&X&X&X\\ X&X&X&X&X&X \end{bmatrix}$$

direct photoelastic effect of the form given in Eq. (4.5) will exist in ferroelectrics also. In fact, the indirect effect can be comparable in magnitude to the direct effect in ferroelectric $\mathrm{LiNbO_3}$, particularly for the p_{3333} coefficient.

After suitable subsidiary measurements have been made the indirect photoelastic effect can be removed from the total susceptibility, represented by the large parentheses in Eq. (4.5), which is measured in an acousto-optic scattering experiment in a piezoelectric crystal. The remaining susceptibility represents the direct photoelastic effect and is a fourth-rank tensor which possesses the symmetry of the crystal group. It also has symmetry upon interchange of i and j to the extent that the difference of the frequencies ω_B and ω_O is negligible. Examination of Eq. (4.6) shows, however, that the direct-effect susceptibility is *not* symmetric upon interchange of k and l, unlike the Pockels phenomenological formulation of Eq. (1.1). Thus the strain, which is the symmetric combination of displacement gradients, cannot be used here as the independent variable to describe elastic deformation. Rather, the displacement gradient itself is seen to be the natural measure of elastic deformation relevant to the photoelastic effect. This represents a departure from the view, held since the work of Neumann[1] (in 1841) that the strain is such a measure.

It is useful to divide the susceptibility of Eq. (4.6) into parts symmetric in k and l, indicated by parentheses around the subscripts, and antisymmetric in k and l, indicated by brackets around the subscripts:

$$\begin{aligned} \chi_{i}{}^{\omega_B}{}_{j}{}^{\omega_O}{}_{(kl)}{}^{\omega_A} = &-\tfrac{1}{2}\chi_{ij}(\omega_O)\delta_{kl} \\ &-\epsilon_0 \sum_{\rho,\mu,\beta,\nu} \Upsilon^{\rho\mu}_{im}(\omega_B)\Upsilon^{\beta\nu}_{bj}(\omega_O)q^{\nu}q^{\rho}\,{}^{(2,1)}H^{\mu\beta}_{mbkl} \\ &+3\epsilon_0^2 \sum_{\rho,\mu,\beta,\gamma,\varphi,\nu} \Upsilon^{\rho\mu}_{im}(\omega_B) \\ &\times\Upsilon^{\beta\nu}_{bj}(\omega_O)\Upsilon^{\gamma\varphi}_{ce}(\omega_A)q^{\nu}q^{\rho}\,{}^{(3,0)}H^{\mu\beta\gamma}_{mbc}\,{}^{(1,1)}H^{\varphi}_{ekl}\,, \end{aligned} \tag{4.15}$$

$$\begin{aligned} \chi_{i}{}^{\omega_B}{}_{j}{}^{\omega_O}{}_{[kl]}{}^{\omega_A} = &-\tfrac{1}{2}\epsilon_0 \sum_{\rho,\mu,\beta,\nu} \Upsilon^{\rho\mu}_{im}(\omega_B)\Upsilon^{\beta\nu}_{bj}(\omega_O)q^{\nu}q^{\rho} \\ &\times\left({}^{(2,0)}H^{\mu\beta}_{lb}\delta_{mk} - {}^{(2,0)}H^{\mu\beta}_{kb}\delta_{ml} + {}^{(2,0)}H^{\mu\beta}_{ml}\delta_{bk} \right. \\ &\left. - {}^{(2,0)}H^{\mu\beta}_{mk}\delta_{bl}\right) \quad . \end{aligned} \tag{4.16}$$

Note that the first term on the right-hand side of Eq. (4.15) involves directly measurable tensors of a lower order and as such could be regarded as an indirect effect. We choose, however, to include it in the direct effect. The susceptibility of Eq. (4.15) can be related to the Pockels photoelastic tensor by

$$\chi_{i}{}^{\omega_B}{}_{j}{}^{\omega_O}{}_{(kl)}{}^{\omega_A} = -\tfrac{1}{2}\kappa_{im}(\omega_B)p_{mn(kl)}\kappa_{nj}(\omega_O) \quad , \tag{4.17}$$

where $p_{mn(kl)}$ is synonymous with the photoelastic tensor of Eq. (1.1) and the factor of $\frac{1}{2}$ arises since $p_{mn(kl)}$, like r_{lmk} above, is quoted as a static value. Similarly, we can define a tensor $p_{mn[kl]}$, which has no analog in the Pockels formulation, by

$$\chi_{i}{}^{\omega_B}{}_{j}{}^{\omega_O}{}_{[kl]}{}^{\omega_A} = -\tfrac{1}{2}\kappa_{im}(\omega_B)p_{mn[kl]}\kappa_{nj}(\omega_O) \quad . \tag{4.18}$$

The antisymmetric tensor of Eq. (4.16) can be more readily understood if ${}^{(2,0)}\overleftrightarrow{\mathrm{H}}$ is eliminated from Eq. (4.16) with the use of Eq. (3.16). Therefore

$$\begin{aligned} \chi_{i}{}^{\omega_B}{}_{j}{}^{\omega_O}{}_{[kl]}{}^{\omega_A} = &-\frac{1}{4}\sum_{\rho,\nu,\mu} q^{\rho}q^{\nu} \\ &\times\{\Upsilon^{\rho\mu}_{ik}(\omega_B)[\epsilon_0\omega_O^2 m^{\mu}\Upsilon^{\nu\mu}_{jl}(\omega_O)+\delta^{\nu\mu}\delta_{jl}] \\ &-\Upsilon^{\rho\mu}_{il}(\omega_B)[\epsilon_0\omega_O^2 m^{\mu}\Upsilon^{\nu\mu}_{jk}(\omega_O)+\delta^{\nu\mu}\delta_{jk}] \end{aligned}$$

$$+\Upsilon^{\mu\nu}_{kj}(\omega_O)\left[\epsilon_0\omega_B^2 m^\mu \Upsilon^{\rho\mu}_{il}(\omega_B)+\delta^{\rho\mu}\delta_{il}\right]$$

$$-\Upsilon^{\mu\nu}_{lj}(\omega_O)\left[\epsilon_0\omega_B^2 m^\mu \Upsilon^{\rho\mu}_{ik}(\omega_B)+\delta^{\rho\mu}\delta_{ik}\right]\Big\} \qquad (4.19)$$

It is seen that the four terms involving either ω_O^2 or ω_B^2 are almost exactly canceling. Since the remainder from this near cancellation is of order ω_A/ω_O times the dominant terms it can be neglected. Hence

$$\chi^{\omega_B\omega_O\omega_A}_{i\,j[kl]} \cong \tfrac{1}{4}\left[\kappa_{il}(\omega_B)\delta_{jk}-\kappa_{ik}(\omega_B)\delta_{jl}+\kappa_{lj}(\omega_O)\delta_{ik}-\kappa_{kj}(\omega_O)\delta_{il}\right] \quad . \qquad (4.20)$$

Note that, to this approximation, this antisymmetric susceptibility can be calculated from the dielectric tensor alone.[27] Equations (4.18) and (4.20) can be combined to yield an antisymmetric part of the photoelastic tensor

$$p^{\omega_B\omega_O\omega_A}_{i\ j\ \ kl}=\tfrac{1}{2}\Big\{[\kappa^{-1}(\omega_O)]_{lj}\delta_{ik}-[\kappa^{-1}(\omega_O)]_{kj}\delta_{il}+[\kappa^{-1}(\omega_B)]_{il}\delta_{kj}-[\kappa^{-1}(\omega_B)]_{ik}\delta_{lj}\Big\} \; . \qquad (4.21)$$

Contributions to the nonlinear polarization of Eq. (4.5) from the antisymmetric susceptibility given by Eq. (4.20) will arise only from the antisymmetric combination of displacement gradients, which forms the mean rotation tensor

$$\tilde{R}_{ij}=\tfrac{1}{2}(u_{i,j}-u_{j,i}) \quad . \qquad (4.22)$$

We thus see from Eqs. (4.20) and (4.22) that the antisymmetric susceptibility arises from the rotation of the linear optical anisotropy, that is, the dielectric tensor, within volume elements in the acoustic wave. Since rotation occurs only in acoustic waves possessing a shear component, as seen from Eq. (4.22), only antisymmetric susceptibility components that combine with shear-wave components can occur.

The number of antisymmetric components which can exist in various crystals can be determined from either Eq. (4.20) or (4.21). The components of the dielectric tensor in these equations must be expressed in the rectangular crystallographic coordinate system. For crystals of orthorhombic or higher symmetry the principal coordinate system of the dielectric tensor coincides with the crystallographic coordinate system which makes the determination very simple. Isotropic substances and cubic crystals have no antisymmetric components since they are optically isotropic (optical activity is neglected in this paper). All uniaxial crystals (hexagonal, tetragonal, and trigonal systems) have four nonzero antisymmetric components. Biaxial crystals of orthorhombic symmetry have six. Crystals of monoclinic symmetry have one principal axis fixed in the x_2 crystallographic direction; the orientation of the other two principal axes depends on wavelength. Fourteen nonzero antisymmetric photoelastic components result for crystals of this symmetry. The principal axes can have a general orientation with respect to the crystallographic axes in triclinic crystals. Thirty-six antisymmetric components result in this case. These results are summarized in Table II.

TABLE II. Nonzero components of the antisymmetric photoelastic tensor, Eq. (4.21). The quantity $p_{mn}=\frac{1}{2}\epsilon_{nkl}\times p_{m[kl]}$, where m is the normal contraction of the first pair of indices, is plotted. Nonzero components, indicated by a dot, are joined by a line when equal. All components are zero for isotropic substances and cubic crystals.

Hexagonal, tetragonal, trigonal systems	Orthorhombic system	Monoclinic system	Triclinic system
0 0 0	0 0 0	0 • 0	• • •
0 0 0	0 0 0	0 0 0	• • •
0 0 0	0 0 0	0 • 0	• • •
• 0 0	• 0 0	• 0 •	• • •
0 • 0	0 • 0	0 • 0	• • •
0 0 0	0 0 •	• 0 •	• • •

Though the rotational effect may be very small in weakly birefringent crystals, it can be comparable to the ordinary strain contribution in strongly birefringent crystals. For instance, for calcite in the visible,

$$(p_{2332}-p_{2323})/\tfrac{1}{2}(p_{2332}+p_{2323})=98\% \qquad (4.23)$$

if the measured value[36] of $p_{23(23)}$ is used for the denominator.

V. ORIGIN AND DISPERSION OF PHOTOELASTIC SUSCEPTIBILITY

Since we have discussed the origin of the antisymmetric part of the direct photoelastic effect as well as the indirect photoelastic effect in Sec. IV, our remarks in this section will concern only the symmetric part of the direct photoelastic effect. The susceptibility representing this portion is given by Eq. (4.15). Examination of that equation indicates that there are three major contributions [minor contributions were dropped in passing from Eq. (3.22) to Eq. (4.15)] to the symmetric susceptibility. The first term on the right-hand side of Eq. (4.15) represents simply the change of the number of oscillators per unit volume due to the compressional and dilatational effects present in longitudinal acoustic waves. Note that the frequency dependence of this term arises from a single susceptibility factor and that it has a higher symmetry than the remaining terms.

The second group of terms on the right-hand side of Eq. (4.15), distinguished by a frequency dependence determined by two susceptibility-like factors, contains ${}^{(2,1)}\overleftrightarrow{\mathrm{H}}$. The tensor constant ${}^{(2,1)}\overleftrightarrow{\mathrm{H}}$ represents the internal direct, or one-step, mixing of the acoustic and optic fields. It has the lowest symmetry of any term in Eq. (4.15). Its components with $\mu = \beta$ can be interpreted as the contribution from the μth oscillator arising from the deformation-altered resonant frequency. The components of ${}^{(2,1)}\overleftrightarrow{\mathrm{H}}$ with $\mu \neq \beta$ represent the contribution from the deformation-altered oscillator strengths arising from the interaction of the μth and βth oscillators caused by the deformation.

The last term in Eq. (4.14) represents an internal or microscopic two-step contribution to the symmetric part of the direct photoelastic susceptibility since it involves both ${}^{(1,1)}\overleftrightarrow{\mathrm{H}}$ and ${}^{(3,0)}\overleftrightarrow{\mathrm{H}}$. It is physically distinguishable from the other conributions by its dispersion which is given by a product of three susceptibility like factors and by its symmetry which differs from that of the ${}^{(2,1)}\overleftrightarrow{\mathrm{H}}$ term. It arises from the mixing of the polarization induced by the input optical field via ${}^{(3,0)}\overleftrightarrow{\mathrm{H}}$ with the internal displacement produced via ${}^{(1,1)}\overleftrightarrow{\mathrm{H}}$ by the input acoustic field. This internal two-step contribution does not occur in all materials; symmetry conditions can cause both ${}^{(3,0)}\overleftrightarrow{\mathrm{H}}$ and ${}^{(1,1)}\overleftrightarrow{\mathrm{H}}$ to be zero for some crystals. Such is the case, for instance, for crystals possessing the rocksalt structure. Conversely, ${}^{(3,0)}\overleftrightarrow{\mathrm{H}}$ and ${}^{(1,1)}\overleftrightarrow{\mathrm{H}}$ need not be zero in a nonpiezoelectric crystal even though by Eqs. (4.7) and (4.8) ${}^{(3,0)}\overleftrightarrow{\mathrm{H}}$ and ${}^{(1,1)}\overleftrightarrow{\mathrm{H}}$ give rise, respectively, to the electro-optic and piezoelectric effects. This is true because symmetry operations affect the "post"-superscripts of the $\overleftrightarrow{\mathrm{H}}$'s via Eq. (2.44) as well as the subscripts. As an example, ${}^{(3,0)}\overleftrightarrow{\mathrm{H}}$ and ${}^{(1,1)}\overleftrightarrow{\mathrm{H}}$ are both nonzero for crystals of the diamond structure.

An important result of the present theory is its prediction of the form of the frequency dispersion of the photoelastic susceptibility. Four things concerning this are important: (i) The various ${}^{(m,n)}\overleftrightarrow{\mathrm{H}}$ by their nature are frequency-independent quantities; (ii) all frequency dependence arises from the dynamical character of the equations used and appears only in $\Upsilon_{ij}^{\alpha\beta}(\omega)$ factors; (iii) contributions to the direct photoelastic susceptibility having different symmetry with respect to lower indices are shown to possess different dispersion; and (iv) the number of ${}^{(m,n)}\overleftrightarrow{\mathrm{H}}$ components present in Eq. (4.15) that must be fit to experimental values depends upon the crystal symmetry and the number of internal coordinates (number of oscillators) which are important to the experimental results.

A proof of the correctness of Eq. (4.15) could be furnished by detailed agreement of the predicted dispersion with experiment. There is not, however, a great deal of data available at present on dispersion of the photoelastic tensor elements, especially in spectral regions and materials where the dispersion is great. Of the few available[37–44] the study by Gavini and Cardona[44] is the most extensive in scope and successful in interpretation and hence we wish to consider their work in the framework of the present theory. Wemple and DiDomenico[26] have recently summarized photoelastic dispersion data and drawn attention to the importance of excitons in causing strong dispersion in the photoelastic tensor elements.

To demonstrate our statement that the present theory can account for the dispersion of the photoelastic tensor components we will show that Eq. (4.15) encompasses the successful dispersion analysis of Gavini and Cardona.[44] They accounted for the strong dispersion encountered near the exciton absorption edge in various alkali halides. They found it necessary to consider the three excitons that correspond to the three valence-band maxima at the center of the Brillouin zone, two of which are degenerate in the absence of stress and one which is separated by the spin-orbit splitting. They also considered an interband (valence-to-conduction-band) oscillator which contributed constant amounts to the photoelastic tensor components, its dispersion in the measured region being negligible. Thus in Eq. (4.15) we take the Greek-letter superscripts to represent normal-mode coordinates and to have values one and two for the exciton oscillators degenerate in the undeformed state, three for the exciton oscillator separated by the spin-orbit splitting, and four for the interband oscillator.

For the rocksalt structure ($m3m$) we have

$$
{}^{(3,0)}H^{\mu\beta\gamma}_{mbc} = 0, \quad {}^{(1,1)}H^{\varphi}_{ekl} = 0. \tag{5.1}
$$

Also, for normal-mode coordinates

$$
{}^{(2,0)}H^{\beta\gamma}_{bc} = {}^{(2,0)}H\,\delta^{\beta\gamma}\delta_{bc}\ . \tag{5.2}
$$

The latter allows Eq. (3.16) to be simplified to

$$
\Upsilon^{\alpha\gamma}_{ac}(\omega) = \frac{C^{\gamma}\delta^{\alpha\gamma}\delta_{ac}}{1-(\omega/\omega^{\gamma})^{2}}\,, \tag{5.3}
$$

where

$$
C^{\gamma} = [2\epsilon_0\,{}^{(2,0)}H^{\gamma\gamma}]^{-1}\ , \tag{5.4}
$$

$$
\omega^{\gamma} = [2\,{}^{(2,0)}H^{\gamma\gamma}/m^{\gamma}]^{1/2}\quad . \tag{5.5}
$$

With these simplifications Eq. (4.15) becomes

$$
\chi_{i\ j\ kl}^{\omega_B\omega_O\omega_A} = -\tfrac{1}{2}(\delta_{ij}\delta_{kl})\sum_{\gamma}\frac{(q^{\gamma})^{2}C^{\gamma}}{1-(\omega/\omega^{\gamma})^{2}} - \epsilon_0\sum_{\mu,\beta}\frac{q^{\mu}q^{\beta}C^{\mu}C^{\beta}\,{}^{(2,1)}H^{\mu\beta}_{ijkl}}{[1-(\omega/\omega^{\beta})^{2}][1-(\omega/\omega^{\mu})^{2}]}\,. \tag{5.6}
$$

Even in normal-mode coordinates ${}^{(2,1)}\overleftrightarrow{\mathrm{H}}$ need not be diagonal in μ, β space; it is, however, symmetric upon interchange of μ and β simultaneously with interchange of i and j. The parameters $(q^\gamma)^2 C^\gamma$ and ω^γ, corresponding, respectively, to the γth oscillator strength and resonant frequency, are not free parameters in fitting to photoelasticity data. They can and should be determined by the dispersion of the optical dielectric constant. From Eqs. (3.19), (3.21), and (5.3) we have, for this purpose, in this crystal structure

$$\kappa_{ij}(\omega)=\delta_{ij}\left\{1+\sum_\gamma (q^\gamma)^2 C^\gamma \Big/ \left[1-\left(\frac{\omega}{\omega^\gamma}\right)^2\right]\right\}. \tag{5.7}$$

There is left in Eq. (5.6) a large number of undetermined ${}^{(2,1)}\overleftrightarrow{\mathrm{H}}$ components even for the rocksalt structure. There are three independent components

$$i,j,k,l=1,1,1,1;1,1,2,2;1,2,1,2$$

with respect to the lower tensor indices and there are ten independent combinations of values of ${}^{(2,1)}\overleftrightarrow{\mathrm{H}}$ for μ, β (reduced from 16 because of symmetry upon interchange of μ, β). This means we still have $3\times 10=30$ independent parameters. From a knowledge of the energy bands from which the excitons are formed, Gavini and Cardona derive relations which allow expressing ${}^{(2,1)}\overleftrightarrow{\mathrm{H}}$ values (for a given set of lower indices) denoted by

$$\mu,\ \beta=1,4;2,4;3,4;1,1;2,2;3,3$$

to a single parameter they call α. The values of ${}^{(2,1)}\overleftrightarrow{\mathrm{H}}$ for μ, $\beta=1,2;1,3;2,3$, are taken as negligibly small and the value of ${}^{(2,1)}\overleftrightarrow{\mathrm{H}}$ for μ, $\beta=4,4$, is related to their parameter C. Further it is assumed by them that the effective interband oscillator frequency is very large compared to the exciton oscillator frequencies. In this manner Gavini and Cardona, and we as well, reduce the number of unknowns to two for a given set of tensor indices. Thus the present theory is also able to account for the fine dispersion fit found by those authors and to do so on the same physical basis.

VI. CONCLUDING REMARKS

The present work points up the dangers of phenomenological theories. The Pockels phenomenological formulation of photoelasticity expressed in Eq. (1.1) contains five physical statements: (a) The effect depends only on the strain as an independent variable; (b) p_{ijkl} is symmetric upon interchange of k and l; (c) p_{ijkl} is an ordinary tensor; (d) p_{ijkl} possesses the symmetry of the crystal; and (e) p_{ijkl} is symmetric upon interchange of i and j. The rotation effect invalidates (a) and (b) and the indirect photoelastic effect invalidates (c) and (d). Thus statements (a)–(e) apply only to the *symmetric* part of the *direct* photoelastic susceptibility given by Eq. (4.15). Nonetheless, it has been believed over the years that the Pockels formulation had been properly tested experimentally.[45] Clearly more experimental work is needed in strongly piezoelectric crystals and in strongly birefringent crystals to verify the modifications of the Pockels formulation that have been presented here. It is also worth noting that on the basis of this work the recent phenomenological formulation of nonlinear photoelasticity[46] is clearly in error for birefringent and piezoelectric crystals.

We showed in Sec. IV that the measure of elastic deformation relevant to the photoelastic effect is the displacement gradient, not the strain or the stress as believed previously. It is worth remarking at this point that this statement applied only to the largest contributions to the photoelastic effect, the ones sizable enough to be measurable within present-day accuracy. Other much smaller contributions to the photoelastic susceptibility, such as that arising from the first and second terms on the right-hand side of Eq. (3.10), depend on the first and second time derivatives of the displacement as well as gradients of the optical electric field. It should be remembered that materials and circumstances may be found in the future where these contributions will be measurable. For such materials additional measures of elastic deformation, the time derivatives of the displacement will be needed for photoelastic studies.

The equations developed in Sec. IV form the basis of a study of dispersion in the photoelastic tensor components in regions where absorption is negligible. If photoelastic dispersion were studied in the restrahlen region, for instance, loss in the lattice oscillators would have to be included in the theory. We leave this for a future study.

Since wave-vector dispersion has been omitted from the treatment of photoelasticity in this paper, care must be exercised in applying the results obtained here to optically active media. If such is done, effects caused by deformation-induced changes in optical activity as well as normal optical activity must be avoided in making comparisons with this theory. Stress can induce optical activity in crystals which normally do not possess it.[47]

Because an indirect photoelastic effect can be very large in ferroelectrics, it will be important to generalize this treatment to include ferroelectric crystals. We leave this for a later paper.

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APPENDIX

We wish to make an order-of-magnitude estimate of the various terms in $\vec{\mathrm{F}}^\mu(1,1)$ of Eq. (3.10) and in $\vec{\mathrm{I}}(1,1)$ of Eq. (3.11) in order to determine which should be kept in the nonlinear polarization $\vec{\mathcal{P}}(1,1)$ of Eq. (3.22). From Eqs. (3.8) and (3.9) with the nonlinear terms set to zero it is possible to make the following estimates:

$$ {}^{(2,0)}\overleftrightarrow{\mathrm{H}} \approx m^R \omega_R^2 , \tag{A1} $$

$$ {}^{(0,2)}\overleftrightarrow{\mathrm{H}} \approx m^0 v_A^2 , \tag{A2} $$

$$ {}^{(1,1)}\overleftrightarrow{\mathrm{H}} \approx v_A \omega_R (m^0 m^R)^{1/2} , \tag{A3} $$

where R denotes the value characteristic of some resonance, ionic or electronic, of the crystal (different superscripts, which have been omitted here, will correspond to different resonant values). We also estimate

$$ {}^{(2,1)}\overleftrightarrow{\mathrm{H}} \approx {}^{(2,0)}\overleftrightarrow{\mathrm{H}} , \tag{A4} $$

$$ {}^{(3,0)}\overleftrightarrow{\mathrm{H}} \approx {}^{(2,0)}\overleftrightarrow{\mathrm{H}}/a \quad , \tag{A5} $$

where a is a typical primitive unit-cell dimension. From the linear problems (0, 1) and (1, 0) the following estimates are possible:

$$ \vec{\mathrm{y}}(0,1) \approx [\omega_A (m^0)^{1/2}/\omega_R (m^R)^{1/2}]\, \vec{\mathrm{u}}(0,1) \quad , \tag{A6} $$

$$ \vec{\mathrm{u}}(1,0) \approx [v_A \omega_R (m^R)^{1/2}/v_O \omega_O (m^0)^{1/2}]\, \vec{\mathrm{y}}(1,0), \tag{A7} $$

$$ \vec{\mathrm{E}}(1,0) \approx [\omega_R^2 m^R/q^R]\, \vec{\mathrm{y}}(1,0) \quad , \tag{A8} $$

$$ \vec{\mathrm{E}}(0,1) \approx [\omega_A q^R (m^0)^{1/2}/\omega_R \epsilon_0 (m^R)^{1/2}]\, \vec{\mathrm{u}}(0,1) \quad , \tag{A9} $$

$$ \vec{\mathrm{B}}(1,0) \approx [\omega_R^2 m^R/q^R v_O]\, \vec{\mathrm{y}}(1,0) \quad , \tag{A10} $$

$$ \vec{\mathrm{B}}(0,1) \approx [v_A \omega_A q^R (m^0)^{1/2}/c^2 \omega_R \epsilon_0 (m^R)^{1/2}]\, \vec{\mathrm{u}}(0,1) \quad . \tag{A11} $$

The charge density q^R can be eliminated by using its relation to the change in the dielectric tensor $\Delta\kappa$ below and above a resonance according to

$$ \Delta\kappa \approx (q^R)^2/\epsilon_0 m^R \omega_R^2 \quad . \tag{A12} $$

Denote the terms on the right-hand side of Eq. (3.10) in order by (1) to (11) with the arguments shown and (1)′ to (11)′ with the arguments interchanged. Their ratios to the last and largest term are approximately

$$ (1)/(11) \approx \omega_O^2 v_A/\omega_R^2 v_O \quad , \tag{A13} $$

$$ (2)/(11) \approx \omega_O \omega_A v_A/\omega_R^2 v_O \quad , \tag{A14} $$

$$ (3)/(11) \approx v_A/v_O \quad , \tag{A15} $$

$$ (4)/(11) \approx \omega_A (m^0)^{1/2} \Delta\kappa/\omega_R (m^R)^{1/2} \quad , \tag{A16} $$

$$ (5)/(11) \approx \omega_O v_A^2 \Delta\kappa (m^0)^{1/2}/\omega_R c^2 (m^R)^{1/2} , \tag{A17} $$

$$ (6),\ (7),\ (8),\ (9)/(11) \approx 1 \quad , \tag{A18} $$

$$ (10)/(11) \approx v_A (m^0)^{1/2}/a\omega_R (m^R)^{1/2} \quad , \tag{A19} $$

$$ (1)'/(11) \approx \omega_A^2 v_A/\omega_R^2 v_O \quad , \tag{A20} $$

$$ (2)'/(11) \approx \omega_A \omega_O v_A/\omega_R^2 v_O \quad , \tag{A21} $$

$$ (3)'/(11) \approx v_A^3 \Delta\kappa/c^2 v_O \quad , \tag{A22} $$

$$ (4)'/(11) \approx \omega_O v_A (m^0)^{1/2}/\omega_R v_O (m^R)^{1/2} \quad , \tag{A23} $$

$$ (5)'/(11) \approx \omega_A v_A (m^0)^{1/2}/\omega_R v_O (m^R)^{1/2} \quad , \tag{A24} $$

$$ (6)',\ (7)',\ (8)',\ (9)'/(11) \approx v_A^2/v_O^2 \quad , \tag{A25} $$

$$ (10)'/(11) \approx v_A (m^0)^{1/2}/a\omega_R (m^R)^{1/2} \quad , \tag{A26} $$

$$ (11)'/(11) \approx v_A^2/v_O^2 \quad . \tag{A27} $$

From these expressions it can be seen that only terms (6)–(11) and (10)′ are significant in size in $\vec{\mathrm{F}}^{\mu}(1,1)$.

If the three types of terms in $\vec{\mathrm{I}}(1,1)$ in Eq. (3.12) are numbered in order (1)–(3) with the arguments shown and (1)′–(3)′ with the arguments interchanged, then their ratios to the first and largest term are

$$ (2)/(1) \sim \omega_A/\omega_B , \tag{A28} $$

$$ (3)/(1) \sim v_A/v_O , \tag{A29} $$

$$ (1)'/(1) \sim v_A^2/v_O^2 , \tag{A30} $$

$$ (2)'/(1) \sim v_A^2/v_O^2 , \tag{A31} $$

$$ (3)'/(1) \sim v_A \omega_A/v_O \omega_B \quad . \tag{A32} $$

Thus only the first term in $\vec{\mathrm{I}}(1,1)$ will be significant in size for an ordinary photoelastic experiment. From Eq. (3.22) it can be seen that both the dominant term in $\vec{\mathrm{I}}(1,1)$ and the dominant terms in $\vec{\mathrm{F}}^{\mu}(1,1)$ contribute similarly $[\sim \omega_A \omega_B q^R y(1,0) u(0,1)/v_A]$ to the nonlinear polarization.

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[1] For a history of photoelasticity see E. G. Coker and L. N. G. Filon, *A Treaties on Photo-Elasticity* (Cambridge U. P., London, 1931), Chap. 3.

[2] F. Pockels, Ann. Physik 37, 144 (1889); 37, 269 (1889); 37, 372 (1889); 39, 440 (1890).

[3] Errors made by Pockels concerning the symmetry of p_{ijkl} for several crystal classes were corrected by S. Bhagavantam, Proc. Indian. Acad. Sci. A16, 359 (1942); Acta Cryst. 5, 591 (1952).

[4] L. Brillouin, Ann. Phys. (Paris) 17, 88 (1922).

[5] E. Gross, Nature 126, 201 (1930); 126, 400 (1930); 126, 603 (1930).

[6] M. Leontowitsch and S. Mandlestam, Jr., Physik Z. Sowjetunion 1, 317 (1931).

[7] C. V. Raman and C. S. Venkateswaran, Nature 142, 250 (1938).

[8] L. Sibaiya, Proc. Indian Acad. Sci. A8, 393 (1938).

[9] V. Chandrasekharan, Proc. Indian. Acad. Sci. A33, 183 (1951).

[10] R. Y. Chiao, C. H. Townes, and B. P. Stoicheff, Phys. Rev. Letters 12, 592 (1964).

[11] P. Debye and F. W. Sears, Proc. Natl. Acad. Sci. U. S. 18, 409 (1932).

[12] R. Lucas and P. Biquard, J. Phys. Radium 3, 464 (1932).

[13] C. V. Raman and N. S. S. Nath, Proc. Indian Acad. Sci. A2, 406 (1935); A2, 413 (1935); A3, 75 (1936).

[14] H. Mueller, Phys. Rev. 47, 947 (1935).

[15] H. Mueller, Z. Krist. A99, 122 (1938).

[16] J. K. Galt, Phys. Rev. 73, 1460 (1948).

[17] E. Burstein and P. L. Smith, Phys. Rev. 74, 229 (1948).

[18] T. S. Narasimhamurty, Acta Cryst. 14, 1176 (1961).

[19] R. W. Dixon, IEEE J. Quantum Electron. QE-3, 85 (1967).

[20] M. Lax and D. F. Nelson (unpublished).

[21] R. Bechmann, *Landolt-Börnstein Critical Tables*, new series edited by K. H. Hellevege (Springer-Verlag, Berlin, 1966), Vol. III/1, p. 125.

[22] G. N. Ramachandran and S. Ramaseshan, in *Handbuch der Physik*, edited by S. Flügge (Springer, Berlin, 1961), Vol. XXV/1, p. 1; K. Vedam and S. Ramaseshan, in *Progress in Crystal Physics*, edited by R. S. Krishnan (Interscience, New York, 1958), Vol. 1, Chap. IV, p. 102; C. F. Quate, C. D. W. Wilkinson, and D. K. Winslow, Proc. IEEE 53, 1604 (1965); W. G. Cady, *Piezoelectricity* (Dover, New York, 1964), Vol. 2, Chap. XXX.

[23] J. Chapelle and L. Taurel, Compt. Rend. 240, 743 (1955).

[24] G. A. Coquin, IEEE Trans. Sonics Ultrasonics SU17, 66 (1970).

[25] E. M. Brody and H. Z. Cummins, Phys. Rev. Letters 23, 1039 (1969).

[26] S. H. Wemple and M. DiDomenico, Jr., Phys. Rev. B 1, 193 (1970).

[27] D. F. Nelson and M. Lax, Phys. Rev. Letters 24, 379 (1970). *Footnote added in proof*: For experimental confirmation see D. F. Nelson and P. D. Lazay, Phys. Rev. Letters 17, 1187 (1970).

[28] E. B. Wilson, Jr., J. C. Decius, and P. C. Cross, *Molecular Vibrations* (McGraw-Hill, New York, 1955), Chap. 4.

[29] R. A. Toupin, J. Rational Mech. Anal. 5, 849 (1956).

[30] P. M. Morse and H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill, New York, 1953), p. 884.

[31] D. A. Pinnow and R. W. Dixon, Appl. Phys. Letters 13, 156 (1968).

[32] S. Haussühl, Acta Cryst. A24, 697 (1968).

[33] W. P. Mason, *Piezoelectric Crystals and Their Application to Ultrasonics* (Van Nostrand, Princeton, N. J., 1950), p. 223.

[34] E. H. Turner (private communication). His preliminary measurements indicate $r_{52} \approx 8\times 10^{-12}$ m/V, $r_{41} \approx 8\times 10^{-12}$ m/V, $r_{63} \approx 4\times 10^{-12}$ m/V, at $\lambda = 6328$ Å.

[35] S. K. Kurtz, T. T. Perry, and J. G. Bergman, Jr., Appl. Phys. Letters 12, 186 (1968).

[36] F. Pockels, Ann. Physik 11, 726 (1903).

[37] A. A. Giardini, J. Opt. Soc. Am. 47, 726 (1957).

[38] R. Srinivasan, Naturwiss, 48, 96 (1961).

[39] V. I. Nikitenko and G. P. Martynenko, Fiz. Tverd. Tela 7, 622 (1965) [Soviet Phys. Solid State 7, 494 (1965)].

[40] B. Tell, J. M. Worlock, and L. J. Martin, Appl. Phys. Letters 6, 123 (1965).

[41] A. Feldman, Phys. Rev. 150, 748 (1966).

[42] K. K. Dubenskii, A. A. Kaplyanskii, and N. G. Lozovskaya, Fiz. Tverd. Tela 8, 2068 (1966) [Soviet Phys. Solid State 8, 1644 (1966)].

[43] A Feldman and D. Horowitz, Jr., J. Appl. Phys. 39, 5597 (1968).

[44] A. Gavini and M. Cardona, Phys. Rev. 177, 1351 (1969).

[45] See Ref. 1, p. 294.

[46] K. Vedam and R. Srinivasan, Acta Cryst. 22, 630 (1967).

[47] G. S. Ranganath and S. Ramaseshan, J. Opt. Soc. Am. 59, 1229 (1969).