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Theory of Acoustically Induced Optical Harmonic Generation

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Our recent theory of nonlinear electrodynamics of elastic anisotropic dielectrics is applied to acoustically induced optical harmonic generation (AIOHG) in which two input optical waves and an input acoustic wave are mixed to produce an output optical wave at a frequency displaced from the optical harmonic by the much lower acoustic frequency. The susceptibility governing AIOHG is derived from a fundamental point of view for acentric dielectrics of arbitrary symmetry. It consists of (a) a direct effect represented by a fifth-rank material tensor whose symmetry, frequency dispersion, and relation to other nonlinearities are derived, and (b) five indirect contributions, three being two-step processes and two being three-step processes. The indirect contributions are expressible in terms of lower-order directly measurable material tensors and various wave vectors of the interacting waves. Because of the latter dependence these contributions possess symmetry *different* from the direct effect and from each other. They can be comparable in magnitude to the direct effect. Rotations present in shear waves are shown to contribute to AIOHG to an extent comparable to that from strains in materials whose second-order optical mixing tensor is large. This shows that the displacement gradient, *not* the strain, is the measure of elastic deformation relevant to AIOHG. The form of the phase-matched output AIOH wave is derived for waves having an arbitrary orientation in an anisotropic medium. The concept of *double phase matching* is introduced, whereby not only the output wave is phase matched, but also the intermediate step in one of two types of two-step indirect contributions. Under this condition the output wave grows as the fourth power of the crystal length if pump depletion is negligible. Double phase matching can give an output-power enhancement, compared to single phase matching, of $\sim 10^9$ under reasonable conditions.

I. INTRODUCTION

Recently the interaction of three optical fields and one acoustic field was observed under phase-matching conditions.[1] The experiment took the form of observing an optical output field which was at a frequency displaced from the second harmonic of an input optical field by the much lower frequency of an input acoustic field. The interaction has been called acoustically induced optical harmonic generation (AIOHG). The experiments were performed in a collinear interaction geometry in insulating GaAs, a cubic piezoelectric crystal.

One way of visualizing the interaction is to consider the distortion of the crystal caused by the acoustic wave as changing slightly the second-harmonic generation coefficient. The change in this coefficient reverses sign every half-acoustic wavelength since the distortion is of opposite sense every half-wavelength. If the acoustic half-wavelength is made equal to the coherence length of normal second-harmonic generation, then, when the radiated harmonic and its driving polarization become out of phase, the sign reversal of the interaction coefficient allows them to remain in phase. Thus, the interaction will be phase matched under this condition, which can be shown to be algebraically identically to wave-vector conservation in the interaction. In other words, the acoustic wave vector has made up for the wave-vector mismatch occurring in the normal second-harmonic generation.

The same interaction can be used in an acoustically controlled parametric optical process. Here an input acoustic field and an input optical field would mix to produce two output optical fields in a phase-matched interaction. Harris, Wallace, and Quate[2] have considered this process from a phenomenological point of view. They assumed the nonlinear susceptibility governing the interaction consisted of two two-step processes—optical mixing and acousto-optic scattering taken in either order. Their calculations based on this formulation indicated the strength of the over-all interaction to be too small to be useful for acoustically controlled optical parametric devices for reasonable strain levels.

Recently we have developed a classical theory of nonlinear electrodynamics of elastic anisotropic dielectrics.[3] 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. A consistent set of coupled electromagnetic field equations and dynamical force equations governing the motion of the various mechanical degrees of freedom of the material medium are obtained. The theory predicts the symmetry of any nonlinear, as well as linear, interaction of electromagnetic waves and various eigenmodes of the solid such as acoustic, ionic, and electronic vibrations; it predicts the various multistep indirect 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 interaction of three optical fields and one acoustic field. The calculation will be carried through in a form applicable to mixing of two input optical and one input acoustic wave in a noncollinear interaction in a medium of arbitrary symmetry and orientation. However, the susceptibility which we will derive from a fundamental point of view will be applicable to whatever form in which the interaction occurs.

The susceptibility governing AIOHG consists of six major contributions. We will describe them here in the order they appear in Eq. (5.40). The first term represents the direct interaction of the two input optical waves with the one input acoustic wave. It is a fifth-rank tensor and possesses the symmetry of the material. It does not possess symmetry upon interchange of the two elastic indices. This arises because material rotations occurring in the presence of shear distortions can give contributions comparable to those arising from strains in materials of large second-order optical constants (optical-mixing or harmonic-generation tensor). The antisymmetric contribution to the susceptibility from rotation is calculated for GaAs in Sec. V of this paper and is found to be comparable with the measured susceptibility which includes both symmetric and antisymmetric parts. The elastic asymmetry just described is analogous to that derived by us for the ordinary lowest-order photoelastic interaction[4,5] and recently observed.[6] It forces one to consider the displacement gradient, not the strain, as the basic measure of elastic deformation relevant to photoelastic interactions of all orders.

There are five indirect contributions to the AIOH susceptibility:

(a) One two-step indirect process uses the piezoelectric effect to produce an electric field from the input acoustic wave and then mixes this electric field with two electrical fields of the input optical waves via the third-order optical-mixing tensor. The susceptibility representing this indirect effect is a fifth-rank tensor function of the acoustic wave-vector direction. Because of this functional dependence it will have a rather low symmetry, *different* from that of the direct effect. The lower symmetry here is analogous to that found for the indirect photoelastic effect (piezoelectric effect in combination with the electro-optic effect).[5]

(b) A second two-step indirect effect arises from acousto-optic scattering of the input optical wave from the acoustic wave followed by mixing of this scattered wave with the input optical wave via the second-order optical-mixing effect. The susceptibility governing this indirect effect is a fifth-rank tensor function of the wave vectors of the input optical and acoustic waves and the acousto-optic scattered (intermediate) wave. Because of this dependence this contribution to the total susceptibility also has a different symmetry from the direct effect. Included in the acousto-optic scattering process is the contribution made by rotations, as well as strains, when acoustic shear waves are used in optically anisotropic media.[4–6]

(c) A three-step indirect effect arises from the production of an electric field from the input acoustic wave via the piezoelectric effect, optical mixing of this low-frequency electric field with the input optical wave electric field via the electro-optic effect, and a second optical mixing involving the electric field at the acoustically shifted optical frequency with the electric field of the input optical wave. This indirect effect can be equally well described as acousto-optic scattering via the *indirect* photoelastic effect[5] followed by second-order optical mixing. The susceptibility governing this three-step indirect effect is a fifth-rank tensor function of the acoustic and optical wave vectors and the wave vector of the acoustically shifted optical frequency wave. This indirect effect also has a symmetry differing from the others.

(d) A third two-step indirect effect arises from second-order optical mixing (sum-frequency generation, or harmonic generation if the two input optical frequencies are the same) of the two input optical waves followed by acousto-optic scattering of the sum-frequency optical wave with the input acoustic wave via the photoelastic interaction. The latter interaction, once again, contains contributions from rotations as well as strains. The susceptibility controlling this indirect effect is a fifth-rank tensor function of the input acoustic and optical wave vectors and the optical sum-frequency wave vector. The symmetry of this contribution also differs from the rest.

(e) A second three-step interaction arises from second-order optical mixing of the two input optical waves, the production of a low-frequency electric field from the acoustic wave by the piezoelectric effect, and the mixing of the sum-frequency electric field with the low-frequency electric field via the electro-optic interaction. The susceptibility producing this indirect effect is a fifth-rank tensor function of the input acoustic and optical wave vectors, and the sum-frequency wave vector. This three-step indirect effect can equally well be described as sum-frequency generation followed by acousto-optic scattering via the *indirect* photoelastic effect.[5]

The form of the derived susceptibility suggests the possibility in an appropriate geometry and material of *double phase matching*. By this we mean the phase matching not only of the output AIOH but also the intermediate wave in one of the indirect processes. For instance, the acousto-optic scattering of the input optical wave could be phase matched. This would require

$$\vec{k}_B = \vec{k}_O + \vec{k}_A \, , \tag{1.1}$$

where the wave vectors are distinguished by B, O, A for the acousto-optic (Brillouin) scattered wave, the input optical wave, and the acoustic wave. Phase matching of the second step, optical mixing of waves of frequencies ω_O and ω_B, requires

$$\vec{k}_C = \vec{k}_B + \vec{k}_O \, , \tag{1.2}$$

where the AIOH wave is denoted by C. If both steps are phase matched according to Eqs. (1.1) and (1.2), the over-all process is therefore required to be phase matched also,

$$\vec{k}_C = 2\vec{k}_O + \vec{k}_A \, . \tag{1.3}$$

Attaining both Eqs. (1.1) and (1.2) simultaneously will require a material of special dispersion and birefringence in conjunction with appropriate orientation and input optical and acoustic frequencies. When such is attained the over-all strength of the process can be made comparable to ordinary optical mixing, since with attainable acoustic and optical powers essentially 100% of the input optical wave can be scattered at the Bragg angle from the acoustic wave. In this way a third-order process takes on the strength of a second-order process. The increased strength of AIOHG under the condition of double phase matching is indicated by the dependence of the output intensity on the *fourth* power of the crystal thickness (in a plane-wave geometry ignoring depletion of the pump waves); under the condition of single phase matching of the output the dependence is the conventional second power of the crystal thickness. It is hoped that implementation of the double-phase-matching concept will make AIOHG and the related parametric process strong enough to be useful.

Our approach will be to derive the basic equations governing AIOHG in Sec. II based on our general theory of nonlinear electrodynamics.[3] The development will lean heavily on that carried out for the ordinary photoelastic interaction in dielectrics.[5] In Sec. III we form the wave equation for the AIOHG. In Sec. IV it is solved near the condition of single phase matching for a general orientation of the waves with respect to an arbitrary anisotropic medium. In Sec. V the symmetry of the nonlinear susceptibility governing AIOHG, its relation to lower-order susceptibilities, and its dispersion are examined. In Sec. VI the wave equation is solved near the condition of double phase matching.

II. FORMULATION OF BASIC EQUATIONS

Our objective is to develop the theory of the interaction of two input optical waves with an input acoustic wave in a dielectric to produce an output optical wave at a frequency which is the sum of the frequencies of the input waves. We exclude ferromagnetic and ferroelectric materials from our treatment because extra contributions may arise in these materials. The nonlinear contributions obtained here are, however, present in such materials. Furthermore, we will not consider here effects of wave-vector dispersion, such as optical rotation (activity), or loss in the various vibration modes of the solid.

Because the interaction under consideration is linear in the acoustic variable, we can and must linearize the equations in the acoustic displacement. Because of this, the treatment can follow the photoelastic interaction treatment[5] exactly except that three—as well as two—input-field driving terms must be included. Because of this we present here only those changes in the photoelastic-effect development necessitated by the higher-order interaction being studied.

In order to obtain all possible three-field interaction terms we must expand the stored-energy density $\rho^0\Sigma$, given in Eq. (2.43) of Ref. 5, to higher order according to

$$\begin{aligned}\rho^0\Sigma(\Lambda_A^\alpha, E_{BC}) = {} & \sum_{\alpha,\beta} {}^{(2,0)}H_{AB}^{\alpha\beta}\Lambda_A^\alpha\Lambda_B^\beta + \sum_{\alpha} {}^{(1,1)}H_{ABC}^{\alpha}\Lambda_A^\alpha E_{BC} + {}^{(0,2)}H_{ABCD}E_{AB}E_{CD} + \sum_{\alpha,\beta,\gamma} {}^{(3,0)}H_{ABC}^{\alpha\beta\gamma}\Lambda_A^\alpha\Lambda_B^\beta\Lambda_C^\gamma \\ & + \sum_{\alpha,\beta} {}^{(2,1)}H_{ABCD}^{\alpha\beta}\Lambda_A^\alpha\Lambda_B^\beta E_{CD} + \sum_{\alpha} {}^{(1,2)}H_{ABCDE}^{\alpha}\Lambda_A^\alpha E_{BC}E_{DE} + \sum_{\alpha,\beta,\gamma,\delta} {}^{(4,0)}H_{ABCD}^{\alpha\beta\gamma\delta}\Lambda_A^\alpha\Lambda_B^\beta\Lambda_C^\gamma\Lambda_D^\delta \\ & + \sum_{\alpha,\beta,\gamma} {}^{(3,1)}H_{ABCDE}^{\alpha\beta\gamma}\Lambda_A^\alpha\Lambda_B^\beta\Lambda_C^\gamma E_{DE} + \sum_{\alpha,\beta} {}^{(2,2)}H_{ABCDEF}^{\alpha\beta}\Lambda_A^\alpha\Lambda_B^\beta E_{CD}E_{EF} \, .\end{aligned} \tag{2.1}$$

Here Σ is the stored energy per unit mass, ρ^0 the mass density, E_{AB} the tensor measure of finite strain $(A, B = 1, 2, 3)$ defined by

$$E_{AB} = \tfrac{1}{2}(x_{j,A}x_{j,B} - \delta_{AB}) = E_{BA} \, , \tag{2.2}$$

Λ_A^μ a set of $N-1\,(\mu = 1, 2, \ldots, N-1)$ polarizationlike vectors ($A=1,2,3$) defined by

$$\Lambda_A^\mu = R_{iA} y_i^\mu \ , \tag{2.3}$$

where R_{iA} is the finite rotation tensor given by

$$R_{iA} = x_{i,B}(C^{-1/2})_{BA} \ , \tag{2.4}$$

$$C_{AB} = \delta_{AB} + 2E_{AB} = x_{j,A}x_{j,B} \ , \tag{2.5}$$

and the material descriptors ${}^{(m,n)}H^{\alpha\beta\cdots}_{ABCD\cdots}$ are *frequency-independent* tensors characteristic of the material medium. The summations over Greek-letter postsuperscripts run from 1 through $N-1$, where N is the number of vector degrees of freedom (ionic and electronic) per primitive unit cell which are important in characterizing the material medium for the interaction considered. In all equations we employ the summation convention over repeated Latin-letter subscripts. The presuperscripts are just handy designations to indicate the number of Λ_A^α and E_{BC} that are associated with it.

The N vectors $\vec{y}^\mu$ ($\mu = 0,1,2,\ldots,N-1$) include the c.m. position vector ($\mu=0$) and $N-1$ internal coordinate vectors whose components are expressed in a Cartesian frame, called the spatial frame, and denoted by lower-case Latin letters as subscripts. The coordinates $\vec{y}^\mu$ are functions of the time t and $\vec{X}$ which is a continuum variable that designates a material point in a reference frame, called the material frame, which we choose also to be Cartesian. As such, $\vec{X}$ is a time-independent quantity. Components in the material frame are denoted by capital Latin-letter subscripts. The coordinate vectors $\vec{y}^\mu$ are related to the N particle position vectors $\vec{x}^\alpha (\alpha = 1,2,\ldots,N)$ by a transformation matrix $U^{\mu\alpha}$,

$$\vec{y}^\mu(\vec{X},t) = \sum_{\alpha=1}^{N} U^{\mu\alpha}\vec{x}^\alpha(\vec{X},t), \quad \mu = 0,1,2,\ldots,N-1 \ . \tag{2.6}$$

The c.m. coordinate $\vec{y}^0$ is given by

$$\vec{y}^0 = \vec{x} = \sum_{\alpha=1}^{N} \rho^\alpha \vec{x}^\alpha/\rho^0 \ , \tag{2.7}$$

where ρ^α is the mass of the αth particle, taken as a constant, divided by the primitive-cell volume. The coordinates $\vec{y}^\mu$ are also chosen to be displacement invariant and to preserve the diagonality of the kinetic energy,

$$\sum_{\alpha=1}^{N} \rho^\alpha(\dot{\vec{x}}^\alpha)^2 = \sum_{\mu=0}^{N-1} m^\mu (\mathring{\vec{y}}^\mu)^2 \ . \tag{2.8}$$

This equation defines the mass density m^μ associated with the μth internal coordinate of the medium.

The $3N$ force equations representing the material medium, given by Eqs. (2.60) and (2.61) of Ref. 5, now become with the inclusion of all three-field interaction terms which are linear in the displacement $\vec{u}$,

$$\begin{aligned}
m^\mu \frac{\partial^2 y_i^\mu}{\partial t^2} = & -2m^\mu \frac{\partial u_j}{\partial t}\frac{\partial y^\mu_{i,j}}{\partial t} - m^\mu \frac{\partial^2 u_j}{\partial t^2} y^\mu_{i,j} + \sum_\nu q^{\mu\nu}\epsilon_{ijk}\frac{\partial u_j}{\partial t} y_l^\nu B_{k,l} + q^\mu E_i + q^\mu \epsilon_{ijk}\frac{\partial u_j}{\partial t} B_k + \sum_\nu q^{\mu\nu} y_j^\nu E_{i,j} \\
& + \sum_\nu q^{\mu\nu}\epsilon_{ijk}\frac{\partial u_l}{\partial t} y^\nu_{j,l} B_k + \sum_\nu q^{\mu\nu}\epsilon_{ijk}\frac{\partial y_j^\nu}{\partial t} B_k - 2\sum_\beta {}^{(2,0)}H^{\mu\beta}_{ib} y_b^\beta - {}^{(1,1)}H^\mu_{ibc} u_{b,c} \\
& - \sum_\beta {}^{(2,0)}H^{\mu\beta}_{ab}(y_b^\beta u_{i,a} + y_j^\beta u_{j,b}\delta_{ia} - y_b^\beta u_{a,i} - y_j^\beta u_{b,j}\delta_{ia}) - 3\sum_{\beta,\gamma} {}^{(3,0)}H^{\mu\beta\gamma}_{ibc} y_b^\beta y_c^\gamma - 2\sum_\beta {}^{(2,1)}H^{\mu\beta}_{ibcd} y_b^\beta u_{c,d} \\
& - 3\sum_{\beta,\gamma} {}^{(3,0)}H^{\mu\beta\gamma}_{ibc} y_b^\beta y_j^\gamma (u_{j,c} - u_{c,j}) - \frac{3}{2}\sum_{\beta,\gamma} {}^{(3,0)}H^{\mu\beta\gamma}_{abc} y_b^\beta y_c^\gamma (u_{i,a} - u_{a,i}) \\
& - 4\sum_{\beta,\gamma,\delta} {}^{(4,0)}H^{\mu\beta\gamma\delta}_{ibcd} y_b^\beta y_c^\gamma y_d^\delta - 3\sum_{\beta,\gamma} {}^{(3,1)}H^{\mu\beta\gamma}_{ibcde} y_b^\beta y_c^\gamma u_{d,e} \ ,
\end{aligned} \tag{2.9}$$

$$m^0 \frac{\partial^2 u_i}{\partial t^2} = \sum_\alpha {}^{(1,1)}H^\alpha_{agi} y^\alpha_{a,g} + 2\,{}^{(0,2)}H_{abgi} u_{a,bg} + G_i \ , \tag{2.10}$$

where

$$q^\nu = \sum_{\alpha=1}^{N} V^{\alpha\nu} q^\alpha \ , \tag{2.11}$$

$$q^{\mu\nu} = \sum_{\alpha=1}^{N} V^{\alpha\mu} q^\alpha V^{\alpha\nu} = q^{\nu\mu} \ , \tag{2.12}$$

$$\vec{u} = \vec{x} - \vec{X} \ , \tag{2.13}$$

and $\vec{G}$ represents the nonlinear terms in the acoustic equation, which we will not need. The sums in Eqs. (2.9) and (2.10) span the range $1,2,\ldots,N-1$. In Eqs. (2.11) and (2.12) q^α is the charge taken as a constant on the αth particle at position $\vec{x}^\alpha$ divided by the primitive-cell volume while q^ν is the charge associated with the coordinate vector $\vec{y}^\nu$; $q^{\mu\nu}$ is a charge associated with both the μth and νth internal coordinates. The quantity $V^{\alpha\nu}$ is the inverse of $U^{\nu\alpha}$ and $V^{\alpha 0} = 1$. Equation (2.13) defines the displacement $\vec{u}$. The independent variables in Eqs. (2.9) and (2.10) are $\vec{z}$ and t, where $\vec{z}$ is the coordinate vector in a laboratory Cartesian coordinate system. When three-field interaction terms that are linear in $\vec{u}$ are included, the electromagnetic field equations in rationalized mks units, given by Eqs. (2.18), (2.19), (2.63), and (2.64) of Ref. 5, become

$$\vec{\nabla}\times\vec{E}+\frac{\partial\vec{B}}{\partial t}=0\ , \tag{2.14}$$

$$\vec{\nabla}\cdot\vec{B}=0\ , \tag{2.15}$$

$$\epsilon_0\vec{\nabla}\cdot\vec{E}+\sum_\nu q^\nu\vec{\nabla}\cdot\vec{y}^{\,\nu}=\vec{\nabla}\cdot\left[\sum_\nu q^\nu\vec{y}^{\,\nu}\vec{\nabla}\cdot\vec{u}\right]\ , \tag{2.16}$$

$$\frac{\vec{\nabla}\times\vec{B}}{\mu_0}-\epsilon_0\frac{\partial\vec{E}}{\partial t}-\sum_\nu q^\nu\frac{\partial\vec{y}^{\,\nu}}{\partial t}=-\frac{\partial}{\partial t}\left[\sum_\nu q^\nu\vec{y}^{\,\nu}\vec{\nabla}\cdot\vec{u}\right]+\vec{\nabla}\times\left[\sum_\nu q^\nu\vec{y}^{\,\nu}\times\frac{\partial\vec{u}}{\partial t}\right]\ . \tag{2.17}$$

The $3N$ matter equations (2.9) and (2.10) and the eight electromagnetic field equations (2.14)–(2.17) are the basic equations of AIOHG. We will show, however, in Sec. III that the three matter equations (2.10) and the two electromagnetic field equations (2.15) and (2.16) are unnecessary for finding the characteristics of the output light wave in AIOHG in a dielectric.

III. DERIVATION OF THE INHOMOGENOUS WAVE EQUATION

In AIOHG an input optical wave of frequency ω_O and an input acoustic wave of frequency ω_A mix to give an output optical wave of either $2\omega_O+\omega_A$ or $2\omega_O-\omega_A$ frequency. In order to select out a particular frequency component we expand each of the fields $\vec{u}$, $\vec{y}^{\,\nu}$, $\vec{E}$, and $\vec{B}$ in a Fourier series of the form

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

where

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

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

The solutions for AIOHG are $m=2$, $n=\pm1$. Just as in the treatment of the photoelastic interaction[5] an iterative technique can be used in solving the AIOHG problem. Thus the $(m,n)=(2,\pm1)$ solutions can be obtained from a *linear* problem in terms of the (1, 0), (0, 1), (1, 1), and (2, 0) solutions which are obtained separately and independently of the $(2,\pm1)$ solutions.

The $\vec{u}$, $\vec{y}^{\,\nu}$, $\vec{E}$, and $\vec{B}$ fields in the form of Eqs. (3.1)–(3.3) are substituted into Eqs. (2.9), (2.10), and (2.14)–(2.17). For notational convenience we consider the $(m,n)=(2,1)$ problem only; the $(2,-1)$ problem can be handled analogously. We obtain

$$\vec{\nabla}\cdot\vec{B}(\vec{z};2,1)=0\ , \tag{3.4}$$

$$\epsilon_0\vec{\nabla}\cdot\vec{E}(\vec{z};2,1)+\textstyle\sum_\nu q^\nu\vec{\nabla}\cdot\vec{y}^{\,\nu}(\vec{z};2,1)=Q(\vec{z};2,1)\ , \tag{3.5}$$

$$\vec{\nabla}\times\vec{B}(\vec{z};2,1)+i\epsilon_0\omega_C\vec{E}(\vec{z};2,1)+i\omega_C\textstyle\sum_\nu q^\nu\vec{y}^{\,\nu}(\vec{z};2,1)=\vec{I}(\vec{z};2,1), \tag{3.6}$$

$$\vec{\nabla}\times\vec{E}(\vec{z};2,1)-i\omega_C\vec{B}(\vec{z};2,1)=0\ , \tag{3.7}$$

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

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

where

$$Q(\vec{z};2,1)=\tfrac{1}{2}\big[\textstyle\sum_\nu q^\nu y_{i,i}^\nu(\vec{z};2,0)\,u_{j,j}(\vec{z};0,1)+\sum_\nu q^\nu y_{i,i}^\nu(\vec{z};1,1)\,u_{j,j}(\vec{z};1,0)+\sum_\nu q^\nu y_i^\nu(\vec{z};2,0)\,u_{j,ji}(\vec{z};0,1)$$
$$+\textstyle\sum_\nu q^\nu y_i^\nu(\vec{z};1,1)\,u_{j,ji}(\vec{z};1,0)+\text{interchange of }(2,0)\text{ with }(0,1)\text{ and }(1,1)\text{ with }(1,0)\big]\ , \tag{3.10}$$

$$I_i(\vec{z};2,1)=\tfrac{1}{2}i\textstyle\sum_\nu q^\nu\{(\omega_H+\omega_A)\,y_i^\nu(\vec{z};2,0)\,u_{j,j}(\vec{z};0,1)+(\omega_B+\omega_O)\,y_i^\nu(\vec{z};1,1)\,u_{j,j}(\vec{z};1,0)$$
$$+\omega_A(\delta_{il}\delta_{jm}-\delta_{im}\delta_{jl})[y_m^\nu(\vec{z};2,0)\,u_{l,j}(\vec{z};0,1)+y_m^\nu(\vec{z};1,1)\,u_{l,j}(\vec{z};1,0)+y_{m,j}^\nu(\vec{z};2,0)\,u_l(\vec{z};0,1)$$
$$+y_{m,j}^\nu(\vec{z};1,1)\,u_l(\vec{z};1,0)]+\text{interchange of }(2,0)\text{ with }(0,1)\text{ and }(1,1)\text{ with }(1,0)\}\ , \tag{3.11}$$

$$F_i^\mu(\vec{z};2,1)=\frac{1}{2}\Big(2\omega_H\omega_Am^\mu u_j(\vec{z};0,1)\,y_{i,j}^\mu(\vec{z};2,0)+2\omega_B\omega_Om^\mu u_j(\vec{z};1,0)\,y_{i,j}^\mu(\vec{z};1,1)+\omega_A^2m^\mu u_j(\vec{z};0,1)\,y_{i,j}^\mu(\vec{z};2,0)$$
$$+\omega_O^2m^\mu u_j(\vec{z};1,0)\,y_{i,j}^\mu(\vec{z};1,1)-i\omega_Aq^\mu\epsilon_{ijk}u_j(\vec{z};0,1)B_k(\vec{z};2,0)-i\omega_Oq^\mu\epsilon_{ijk}u_j(\vec{z};1,0)B_k(\vec{z};1,1)$$
$$+\sum_\nu q^{\mu\nu}y_j^\nu(\vec{z};0,1)E_{i,j}(\vec{z};2,0)+\sum_\nu q^{\mu\nu}y_j^\nu(\vec{z};1,0)E_{i,j}(\vec{z};1,1)-i\omega_A\sum_\nu q^{\mu\nu}\epsilon_{ijk}y_j^\nu(\vec{z};0,1)B_k(\vec{z};2,0)$$
$$-i\omega_O\sum_\nu q^{\mu\nu}\epsilon_{ijk}y_j^\nu(\vec{z};1,0)B_k(\vec{z};1,1)-\sum_\beta{}^{(2,0)}H_{ab}^{\mu\beta}[y_b^\beta(\vec{z};2,0)\,u_{i,a}(\vec{z};0,1)-y_b^\beta(\vec{z};2,0)\,u_{a,i}(\vec{z};0,1)$$
$$+y_j^\beta(\vec{z};2,0)\,u_{j,b}(\vec{z};0,1)\delta_{ia}-y_j^\beta(\vec{z};2,0)\,u_{b,j}(\vec{z};0,1)\delta_{ia}]-\sum_\beta{}^{(2,0)}H_{ab}^{\mu\beta}[y_b^\beta(\vec{z};1,1)\,u_{i,a}(\vec{z};1,0)$$
$$-y_b^\beta(\vec{z};1,1)\,u_{a,i}(\vec{z};1,0)+y_j^\beta(\vec{z};1,1)\,u_{j,b}(\vec{z};1,0)\delta_{ia}-y_j^\beta(\vec{z};1,1)\,u_{b,j}(\vec{z};1,0)\delta_{ia}]$$
$$-3\sum_{\beta,\gamma}{}^{(3,0)}H_{ibc}^{\mu\beta\gamma}[y_b^\beta(\vec{z};2,0)\,y_c^\gamma(\vec{z};0,1)+y_b^\beta(\vec{z};1,1)\,y_c^\gamma(\vec{z};1,0)]-2\sum_\beta{}^{(2,1)}H_{ibcd}^{\mu\beta}[y_b^\beta(\vec{z};2,0)\,u_{c,d}(\vec{z};0,1)$$

$$+ y_b^\beta(\vec{z};1,1)\, u_{c,d}(\vec{z};1,0)] + \text{interchange of } (2.0) \text{ with } (0,1) \text{ and } (1,1) \text{ with } (1,0)\Big)$$
$$+ \frac{1}{4}\Big(- i\omega_A \epsilon_{ijk} \sum_\nu q^{\mu\nu} [u_j(\vec{z};0,1)\, y_l^\nu(\vec{z};1,0) B_{k,l}(\vec{z};1,0) + u_l(\vec{z};0,1)\, y_{j,l}^\nu(\vec{z};1,0) B_k(\vec{z};1,0)]$$
$$- \frac{3}{2} \sum_{\beta,\gamma} {}^{(3,0)}H_{abc}^{\mu\beta\gamma} \{2 y_b^\beta(\vec{z};1,0)\, y_j^\gamma(\vec{z};1,0)\, \delta_{ia}[u_{j,c}(\vec{z};0,1) - u_{c,j}(\vec{z};0,1)]$$
$$+ y_b^\beta(\vec{z};1,0)\, y_c^\gamma(\vec{z};1,0)[u_{i,a}(\vec{z};0,1) - u_{a,i}(\vec{z};0,1)]\} - 4 \sum_{\beta,\gamma,\delta} {}^{(4,0)}H_{ibcd}^{\mu\beta\gamma\delta} y_b^\beta(\vec{z};1,0)\, y_c^\gamma(\vec{z};1,0)\, y_d^\delta(\vec{z};0,1)$$
$$- 3 \sum_{\beta,\gamma} {}^{(3,1)}H_{ibcde}^{\mu\beta\gamma} y_b^\beta(\vec{z};1,0)\, y_c^\gamma(\vec{z};1,0)\, u_{d,e}(\vec{z};0,1) + \text{interchanges of } (1,0),(1,0), \text{ and } (0,1)\Big) \tag{3.12}$$

$$\omega_C = 2\omega_O + \omega_A \ , \tag{3.13}$$
$$\omega_B = \omega_O + \omega_A \tag{3.14}$$
$$\omega_H = 2\omega_O \ . \tag{3.15}$$

The expression for $\vec{G}(\vec{z};2,1)$ will not be given since we will show shortly that it is unnecessary for the solution we wish. 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 either two or three 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. (3.11) and (3.12) that interchange of $(2,0) \leftrightarrow (0,1)$, $(1,1) \leftrightarrow (1,0)$, and $(1,0) \leftrightarrow (1,0) \leftrightarrow (0,1)$ requires the concurrent interchange of $\omega_H \leftrightarrow \omega_A$, $\omega_B \leftrightarrow \omega_O$, and $\omega_O \leftrightarrow \omega_O \leftrightarrow \omega_A$, respectively, wherever the frequencies appear.

Let us consider the case where the AIOH wave is phase matched to the forced wave whose wave vector is the sum of the wave vectors of the two input optical waves and that of the acoustic wave. In this case all of the terms in the nonlinear driving functions of importance to the output in Eqs. (3.10)–(3.12) will be proportional to $e^{i(2\vec{k}_O+\vec{k}_A)\cdot\vec{z}}$ for a plane-wave interaction. Here $\vec{k}_O$ and $\vec{k}_A$ are the wave vectors of the input optical and acoustic waves. Other cases where phase matching also involves one of the free waves at an intermediate frequency, for example, $\omega_O + \omega_A$, will be discussed in Sec. VI. If we further assume that there is negligible depletion of the input optical and acoustic waves, then the nonlinear driving functions can be represented as

$$Q(\vec{z};2,1) = Q(2,1)e^{i(2\vec{k}_O+\vec{k}_A)\cdot\vec{z}} \ , \tag{3.16}$$
$$\vec{I}(\vec{z};2,1) = \vec{I}(2,1)e^{i(2\vec{k}_O+\vec{k}_A)\cdot\vec{z}} \ , \tag{3.17}$$
$$\vec{F}^\mu(\vec{z};2,1) = \vec{F}^\mu(2,1)e^{i(2\vec{k}_O+\vec{k}_A)\cdot\vec{z}} \ . \tag{3.18}$$

At this point we can reduce the number of equations that we must consider. First, Eq. (3.4) is the divergence of Eq. (3.7) and hence redundant. Second, Eqs. (3.5) and (3.6) can be combined, with their time dependence retained, to yield a relation that can be called the conservation of nonlinear charge:

$$\frac{\partial Q(\vec{z},t;2,1)}{\partial t} + \vec{\nabla}\cdot\vec{I}(\vec{z},t;2,1) = 0 \ . \tag{3.19}$$

Because of this relation it is not surprising that, of the two nonlinear driving functions Q and $\vec{I}$, only one, $\vec{I}$, contributes to the nonlinear susceptibility governing AIOHG and that of the two Eqs. (3.5) and (3.6) only one, Eq. (3.6), is needed in the development. Third, 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 the velocities of acoustic and optical waves in the medium). The neglect of this term uncouples the equation for $\vec{u}$ from the remainder of the equations. This means that the $\vec{u}$ equation can be ignored in solving for the output optical wave and hence that $\vec{G}(\vec{z};2,1)$ plays an insignificant role in driving the output optical field.

The remaining equations can be solved by using Eq. (3.8) to eliminate $\vec{y}^\nu$ from Eq. (3.6), then using Eqs. (3.6) and (3.7) to form an inhomogeneous wave equation for $\vec{E}(\vec{z};2,1)$, and solving it for the electric field of the output light wave.

Define $\Upsilon_{ab}^{\alpha\beta}(\omega)$ by

$$\epsilon_0 \textstyle\sum_\beta \Upsilon_{ab}^{\alpha\beta}(\omega)(2\,{}^{(2,0)}H_{bc}^{\beta\gamma} - \omega^2 m^\beta \delta^{\beta\gamma}\delta_{bc}) = \delta^{\alpha\gamma}\delta_{ac} \ . \tag{3.20}$$

With the use of this equation, Eq. (3.8) yields

$$y_j^\nu(\vec{z};2,1) = \epsilon_0 \textstyle\sum_\mu \Upsilon_{ji}^{\nu\mu}(\omega_C)[q^\mu E_i(\vec{z};2,1) + F_i^\mu(\vec{z};2,1)] \ . \tag{3.21}$$

From Eqs. (3.6) and (3.20) it is seen that the linear susceptibility is given by

$$\chi_{ij}(\omega) = \sum_{\nu,\mu} q^\nu \Upsilon_{ij}^{\nu\mu}(\omega) q^\mu \ . \tag{3.22}$$

Equations (3.6), (3.7), and (3.20) combine to yield the driven wave equation

$$(c/\omega_C)^2[E_{j,ij}(\vec{z};2,1) - E_{i,jj}(\vec{z};2,1)] - \kappa_{ij}(\omega_C)E_j(\vec{z};2,1) = \mathcal{P}_i(\vec{z};2,1)/\epsilon_0 \ , \tag{3.23}$$

where the dielectric tensor $\kappa_{ij}(\omega)$ and the nonlinear driving polarization are given by

$$\kappa_{ij}(\omega) = \delta_{ij} + \chi_{ij}(\omega) \ , \tag{3.24}$$
$$\mathcal{P}_i(\vec{z};2,1) = \epsilon_0 \sum_{\nu,\mu} q^\nu \Upsilon_{ij}^{\nu\mu}(\omega_c) F_j^\mu(\vec{z};2,1) + iI_i(\vec{z};2,1)/\omega_c \ . \tag{3.25}$$

With Eqs. (3.17) and (3.18) the latter equation can be written as

$$\vec{\mathcal{P}}(\vec{z};2,1)=\vec{\mathcal{P}}(2,1)e^{i(2\vec{k}_O+\vec{k}_A)\cdot\vec{z}} . \qquad (3.26)$$

IV. SOLUTION OF WAVE EQUATION FOR SINGLE PHASE MATCHING

The general solution of the inhomogeneous wave equation (3.23) is the sum of the general solution of the homogeneous equation, called the free wave, and a particular solution of the inhomogeneous equation, call the forced wave. Each of these waves will contain parts corresponding to the ordinary and extraordinary waves. We are interested in the general solution near the condition of phase matching of the output wave. Usually only one wave, either the ordinary or the extraordinary, can be phase matched in a given geometry and so we wish to consider only that wave. It must be proportional to one of the free-wave eigenvectors and only the projection of the forced wave onto this eigenvector will contribute to the phase-matched output.

To express the output wave we use the biorthogonal set[5] of plane-wave eigenvectors of the free-wave electric field $\vec{\mathcal{E}}^{(\theta)}$ and the free-wave electric displacement $\vec{\mathfrak{D}}^{(\theta)}$. Here θ refers to either the ordinary wave, extraordinary wave, or longitudinal nonpropagating solution (infinite refractive index). They can be expressed in the principal coordinate system of the dielectric tensor (diagonal elements κ_1, κ_2, κ_3) as

$$\mathcal{E}_i^{(\theta)}=s_i/(n_\theta^2-\kappa_i)N_\theta , \qquad (4.1)$$

$$\mathfrak{D}_i^{(\theta)}=\kappa_i s_i/(n_\theta^2-\kappa_i)N_\theta , \qquad (4.2)$$

where

$$\vec{s}=\vec{k}^\theta/|\vec{k}^\theta| , \qquad (4.3)$$

$$n_\theta=c|\vec{k}^\theta|/\omega , \qquad (4.4)$$

$$N_\theta=\left(\sum_{i=1}^{3} s_i^2\kappa_i/(n_\theta^2-\kappa_i)^2\right)^{1/2} , \qquad (4.5)$$

$\vec{k}^\theta$ being the wave vector of a free wave. Their normalization has been chosen so that

$$\vec{\mathcal{E}}^{(\theta)}\cdot\vec{\mathfrak{D}}^{(\varphi)}=\delta^{\theta\varphi} . \qquad (4.6)$$

Denote the free wave that is phase matchable by ξ and consider the direction of propagation, the frequency, and the polarization state as independent variables of $\vec{\mathcal{E}}$ and $\vec{\mathfrak{D}}$. Therefore

$$\vec{E}(\vec{z};2,1)_{\text{free}}=C^{(\xi)}\vec{\mathcal{E}}^{(\xi)}(\vec{s}_C,\omega_C)e^{i\vec{k}_C\cdot\vec{z}} , \qquad (4.7)$$

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

$$\vec{s}_C=\vec{k}_C/|\vec{k}_C| . \qquad (4.8)$$

The forced wave can be expressed[3] in terms of the eigenvectors of the free-wave electric field that propagates in the direction of the forced wave as

$$\vec{E}(\vec{z};2,1)_{\text{forced}}=\sum_{\varphi=1}^{3}\frac{\vec{\mathcal{E}}^{(\varphi)}(\vec{s}_D,\omega_C)\vec{\mathcal{E}}^{(\varphi)}(\vec{s}_D,\omega_C)\cdot\vec{P}(2,1)}{\epsilon_0(|2\vec{k}_O+\vec{k}_A|^2/|\vec{k}_C|^2-1)}e^{i(2\vec{k}_O+\vec{k}_A)\cdot\vec{z}} , \qquad (4.9)$$

where

$$\vec{s}_D=(2\vec{k}_O+\vec{k}_A)/|2\vec{k}_O+\vec{k}_A| . \qquad (4.10)$$

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

Near the condition of phase matching where a relatively intense output occurs, the small "reflected" electric field at the frequency ω_C can be neglected in the boundary condition at the input surface of the material medium. Thus, the projection of $\vec{E}(\vec{z};2,1)$ on $\vec{\mathcal{E}}^{(\xi)}(\vec{s}_D,\omega_C)$, which is the scalar product of $\vec{E}(\vec{z};2,1)$ with $\vec{\mathfrak{D}}^{(\xi)}(\vec{s}_D,\omega_C)$, is taken as zero at the plane input surface,

$$\vec{E}(\vec{z}_P;2,1)\cdot\vec{\mathfrak{D}}^{(\xi)}(\vec{s}_D,\omega_C)=0$$

$$=C^{(\xi)}\vec{\mathfrak{D}}^{(\xi)}(\vec{s}_D,\omega_C)\cdot\vec{\mathcal{E}}^{(\xi)}(\vec{s}_C,\omega_C)e^{i\vec{k}_C\cdot\vec{z}_P}+\frac{\vec{\mathcal{E}}^{(\xi)}(\vec{s}_D,\omega_C)\cdot\vec{\mathcal{P}}(2,1)e^{i(2\vec{k}_O+\vec{k}_A)\cdot\vec{z}_P}}{\epsilon_0(|2\vec{k}_O+\vec{k}_A|/|\vec{k}_C|^2-1)} , \qquad (4.11)$$

where $\vec{z}_P$ are the coordinates of the input surface, the origin of them being taken to lie in the plane. Since Eq. (4.11) is good only near phase matching, it is a good approximation to take

$$\vec{\mathfrak{D}}^{(\xi)}(\vec{s}_D,\omega_C)\cdot\vec{\mathcal{E}}^{(\xi)}(\vec{s}_C,\omega_C)=1$$

even though the propagation directions are slightly different. Two conditions result from Eq. (4.11):

$$(\Delta\vec{k}_C)_t=(\vec{k}_C-2\vec{k}_O-\vec{k}_A)_t=0 , \qquad (4.12)$$

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

$$C^{(\xi)}=-\frac{\vec{\mathcal{E}}^{(\xi)}(\vec{s}_D,\omega_C)\cdot\vec{\mathcal{P}}(2,1)}{\epsilon_0(|2\vec{k}_O+\vec{k}_A|^2/|\vec{k}_C|^2-1)} . \qquad (4.13)$$

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

$$\vec{E}(\vec{z};2,1)=(-2i/\epsilon_0)\vec{\mathcal{E}}^{(\xi)}\vec{\mathcal{E}}^{(\xi)}\cdot\vec{\mathcal{P}}(2,1)\times(|2\vec{k}_O+\vec{k}_A|^2/|\vec{k}_C|^2-1)^{-1}\times\sin(\Delta\vec{k}_{Cn}\cdot\tfrac{1}{2}\vec{z})e^{i(\vec{k}_C+\vec{k}_O+\vec{k}_A)\cdot\vec{z}/2} , \qquad (4.14)$$

$$\vec{H}(\vec{z};2,1)=(2ic^2/\omega_C)\vec{k}_C\times\vec{\mathcal{E}}^{(\xi)}\vec{\mathcal{E}}^{(\xi)}\cdot\vec{\mathcal{P}}(2,1)\times(|2\vec{k}_O+\vec{k}_A|^2/|\vec{k}_C|^2-1)^{-1}\times\sin(\Delta\vec{k}_{Cn}\cdot\tfrac{1}{2}\vec{z})e^{i(\vec{k}_C+\vec{k}_O+\vec{k}_A)\cdot\vec{z}/2} , \qquad (4.15)$$

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

$$\underline{\vec{S}}(\vec{z};2,1)=\frac{2c^2k_C^4}{\epsilon_0\omega_C}[(\vec{\mathcal{E}}^{(\xi)})^2\vec{k}_C-(\vec{\mathcal{E}}^{(\xi)}\cdot\vec{k}_C)\vec{\mathcal{E}}^{(\xi)}]$$

$$\times \left|\frac{\vec{\mathcal{E}}^{(t)}\cdot\vec{\mathcal{P}}(2,1)}{(\vec{k}_C+\vec{k}_O+\vec{k}_A)\cdot\Delta\vec{k}_{Cn}}\right|^2 \times \sin^2(\Delta\vec{k}_{Cn}\cdot\tfrac{1}{2}\vec{z})\ . \qquad (4.16)$$

This expression applies within the material medium at reasonable distances into the medium but where pump-wave depletion is negligible and at directions and frequencies close to those needed for phase matching of the output AIOH. Except for the latter condition, Eq. (4.16) applies to input waves having arbitrary orientations with respect to the anisotropic medium. Phase matching occurs in this plane-wave geometry when

$$(\Delta\vec{k}_C)_n=(\vec{k}_C-2\vec{k}_O-\vec{k}_A)_n=0 \qquad (4.17)$$

since the boundary condition has required the tangential component to vanish already. At exact phase matching Eq. (4.16) becomes

$$\vec{S}(l;2,1)=(c^2k_C^4l^2/8\epsilon_0\omega_Ck_{Cn}^2)\left|\vec{\mathcal{E}}^{(t)}\cdot\vec{\mathcal{P}}(2,1)\right|^2 \times[(\vec{\mathcal{E}}^{(t)})^2\vec{k}_C-(\vec{\mathcal{E}}^{(t)}\cdot\vec{k}_C)\vec{\mathcal{E}}^{(t)}]\ , \qquad (4.18)$$

where l is the distance into the crystal normal to the entrance surface.

V. NONLINEAR POLARIZATION GOVERNING AIOHG

The nonlinear polarization which drives the AIOHG process is given by Eq. (3.25) in conjunction with Eqs. (3.11) and (3.12). Estimates of magnitudes of the various terms show many of them to be of negligible size under typical conditions. Assuming that there is no phase matching of an intermediate step in any of the indirect contributions and that only the forced wave whose wave vector is the sum of the two input optical wave vectors and the input acoustic wave vector is important for the phase matching of the AIOH output, we find that the following terms in $\vec{\mathcal{P}}(2,1)$ are significant and of comparable size (see Appendix A):

$$\begin{aligned}\mathcal{P}_i(2,1)=&-\epsilon_0\sum_{\nu,\mu}q^\nu\Upsilon_{ij}^{\nu\mu}(\omega_C)\Big(\frac{1}{2}\sum_\beta{}^{(2,0)}H_{ab}^{\mu\beta}[y_b^\beta(2,0)u_{j,a}(0,1)-y_b^\beta(2,0)u_{a,j}(0,1)\\&+y_k^\beta(2,0)u_{k,b}(0,1)\delta_{ja}-y_k^\beta(2,0)u_{b,k}(0,1)\delta_{ja}]+3\sum_{\beta,\gamma}{}^{(3,0)}H_{jbc}^{\mu\beta\gamma}[y_b^\beta(2,0)y_c^\gamma(0,1)+y_b^\beta(1,1)y_c^\gamma(1,0)]\\&+\sum_\beta{}^{(2,1)}H_{jbcd}^{\mu\beta}y_b^\beta(2,0)u_{c,d}(0,1)+\frac{3}{8}\sum_{\beta,\gamma}{}^{(3,0)}H_{abc}^{\mu\beta\gamma}\{2y_b^\beta(1,0)y_k^\gamma(1,0)\delta_{ja}[u_{k,c}(0,1)-u_{c,k}(0,1)]\\&+y_b^\beta(1,0)y_c^\gamma(1,0)[u_{j,a}(0,1)-u_{a,j}(0,1)]\}+3\sum_{\beta,\gamma,\delta}{}^{(4,0)}H_{jbcd}^{\mu\beta\gamma\delta}y_b^\beta(1,0)y_c^\gamma(1,0)y_d^\delta(0,1)\\&+\frac{3}{4}\sum_{\beta,\gamma}{}^{(3,1)}H_{jbcde}^{\mu\beta\gamma}y_b^\beta(1,0)y_c^\gamma(1,0)u_{d,e}(0,1)\Big)-\frac{1}{2}\sum_\alpha q^\alpha y_i^\alpha(2,0)u_{k,k}(0,1)\ ,\end{aligned} \qquad (5.1)$$

where, since the spatial dependence has been removed, the derivative notation now stands for multiplication by the imaginary unit times the appropriate wave vector.

This equation must be reexpressed in terms of a nonlinear susceptibility summed over the input electric field $E_i(1,0)$ and displacement gradient $u_{k,l}(0,1)$ amplitudes. Expressions from the linear optical problem[3] $(m,n)=(1,0)$, the linear acoustic problem[3] $(m,n)=(0,1)$, the acousto-optic scattering problem[5] $(m,n)=(1,1)$, and the second-harmonic generation problem[3] $(m,n)=(2,0)$ are needed. They are

$$y_b^\beta(1,0)=\epsilon_0\sum_\nu\Upsilon_{bj}^{\beta\nu}(\omega_O)q^\nu E_j(1,0)\ , \qquad (5.2)$$

$$y_c^\gamma(0,1)=-\epsilon_0\sum_\varphi\Upsilon_{cr}^{\gamma\varphi}(\omega_A)[{}^{(1,1)}H_{rkl}^\varphi+q^\varphi a_ra_se_{skl}^{\omega_A}/\epsilon_0a_p\kappa_{pq}(\omega_A)a_q]u_{k,l}(0,1)\ , \qquad (5.3)$$

$$y_j^\nu(1,1)=\epsilon_0\sum_\nu\Upsilon_{jk}^{\nu\mu}(\omega_B)[q^\mu E_k(1,1)+F_k^\mu(1,1)]\ , \qquad (5.4)$$

$$y_j^\nu(2,0)=\epsilon_0\sum_\nu\Upsilon_{jk}^{\nu\mu}(\omega_H)[q^\mu E_k(2,0)+F_k^\mu(2,0)]\ , \qquad (5.5)$$

$$\vec{E}(1,1)=\sum_{\varphi=1}^{3}\frac{\vec{\mathcal{E}}^{(\varphi)}(\vec{s}_F,\omega_B)\vec{\mathcal{E}}^{(\varphi)}(\vec{s}_F,\omega_B)\cdot\vec{\mathcal{P}}(1,1)}{\epsilon_0(|\vec{k}_O+\vec{k}_A|^2/|\vec{k}_B|^2-1)}\ , \qquad (5.6)$$

$$\vec{E}(2,0)=\sum_{\varphi=1}^{3}\frac{\vec{\mathcal{E}}^{(\varphi)}(\vec{s}_I,\omega_H)\vec{\mathcal{E}}^{(\varphi)}(\vec{s}_I,\omega_H)\cdot\vec{\mathcal{P}}(2,0)}{\epsilon_0(|2\vec{k}_O|^2/|\vec{k}_H|^2-1)}\ , \qquad (5.7)$$

$$\begin{aligned}F_i^\mu(1,1)=\epsilon_0\Big[&\frac{1}{2}\sum_{\rho,\xi}\Upsilon_{hg}^{\rho\xi}(\omega_O)q^\xi[{}^{(2,0)}H_{ln}^{\mu\rho}\delta_{ik}-{}^{(2,0)}H_{il}^{\mu\rho}\delta_{nk}\\&-{}^{(2,0)}H_{kn}^{\mu\rho}\delta_{il}+{}^{(2,0)}H_{ik}^{\mu\rho}\delta_{nl}+2\,{}^{(2,1)}H_{inkl}^{\mu\rho}]\\&+3\epsilon_0\sum_{\rho,\epsilon,\xi,\varphi}\Upsilon_{eg}^{\rho\epsilon}(\omega_O)\Upsilon_{hr}^{\xi\varphi}(\omega_A)q^\epsilon\,{}^{(3,0)}H_{ieh}^{\mu\rho\xi}\\&\times\Big({}^{(1,1)}H_{rkl}^\varphi+\frac{q^\varphi a_ra_se_{skl}^{\omega_A}}{\epsilon_0a_p\kappa_{pq}(\omega_A)a_q}\Big)\Big]\\&\times E_g(1,0)u_{k,l}(0,1)\ ,\end{aligned} \qquad (5.8)$$

$$F_i^\mu(2,0)=-\tfrac{3}{2}\epsilon_0^2\sum_{\rho,\zeta,\xi,\epsilon}\Upsilon_{df}^{\rho\xi}(\omega_O)\Upsilon_{eg}^{\zeta\epsilon}(\omega_O)\times q^\epsilon q^\xi\,{}^{(3,0)}H_{ide}^{\mu\rho\zeta}E_f(1,0)E_g(1,0)\ , \qquad (5.9)$$

$$\mathcal{P}_i(1,1)=\epsilon_0\left(\chi_i{}^{\omega_B}{}_j{}^{\omega_O}{}_{kl}^{\omega_A}-\frac{2d_i{}^{\omega_B}{}_j{}^{\omega_O}{}_r^{\omega_A}a_ra_se_{skl}^{\omega_A}}{\epsilon_0a_p\kappa_{pq}(\omega_A)a_q}\right)\times E_j(1,0)u_{k,l}(0,1)\ , \qquad (5.10)$$

$$\mathcal{P}_i(2,0)=\epsilon_0d_i{}^{\omega_H}{}_j{}^{\omega_O}{}_k^{\omega_O}E_j(1,0)E_k(1,0)\ . \qquad (5.11)$$

In these equations we have used the unit vectors $\vec{a}$, $\vec{s}_F$, and $\vec{s}_I$ defined by

$$\vec{a}=\vec{k}_A/|\vec{k}_A| \ , \tag{5.12}$$

$$\vec{s}_F=(\vec{k}_O+\vec{k}_A)/|\vec{k}_O+\vec{k}_A| \ , \tag{5.13}$$

$$\vec{s}_I=2\vec{k}_O/|2\vec{k}_O| \ . \tag{5.14}$$

We have also used the piezoelectric tensor $e_{skl}^{\omega_A}$ given by

$$e_{skl}^{\omega_A}=-\epsilon_0\sum_{\theta,\varphi} q^\theta \Upsilon_{se}^{\theta\varphi}(\omega_A)\,^{(1,1)}H_{ekl}^{\varphi} \ , \tag{5.15}$$

which relates the infinitesimal strain S_{kl} defined by

$$S_{kl}=\tfrac{1}{2}(u_{k,l}+u_{l,k}) \ , \tag{5.16}$$

to the linear polarization according to

$$P_j^{\omega_A}=\epsilon_0\chi_{jk}(\omega_A)E_k^{\omega_A}+e_{jkl}^{\omega_A}S_{kl}^{\omega_A} \ . \tag{5.17}$$

The quantity $d_{i\,j\,k}^{\omega_B\omega_O\omega_A}$, defined by

$$d_{i\,j\,k}^{\omega_B\omega_O\omega_A}=-\frac{3\epsilon_0^2}{2}\sum_{\substack{\rho,\mu,\beta\\ \nu,\eta,\gamma}}\Upsilon_{im}^{\rho\mu}(\omega_B)\Upsilon_{bj}^{\beta\nu}(\omega_O)\Upsilon_{ck}^{\gamma\eta}(\omega_A) \times q^\nu q^\eta q^\rho\,^{(3,0)}H_{mbc}^{\mu\beta\gamma} \ , \tag{5.18}$$

is the *direct* electro-optic (clamped) tensor expressed in the form of the optical mixing tensor. It relates two input electric field amplitudes to a nonlinear polarization[7] by

$$\mathrm{P}_i^{\omega_B}=D\epsilon_0 d_{i\,j\,k}^{\omega_B\omega_O\omega_A}E_j^{\omega_O}E_k^{\omega_A} \ , \tag{5.19}$$

where

$D=1$ if $\omega_O=\omega_A$
$\ \ =2$ if $\omega_O\neq\omega_A$.

It is related to the Pockels electro-optic tensor r_{lmk} by

$$d_{i\,j\,k}^{\omega_B\omega_O\omega_A}=-\tfrac{1}{4}\kappa_{il}(\omega_B)r_{lmk}\kappa_{mj}(\omega_O) \ , \tag{5.20}$$

where r_{ijk} is defined in terms of the change of the inverse dielectric tensor caused by a low-frequency electric field E_k

$$(\delta\kappa^{-1})_{ij}=r_{ijk}E_k \ . \tag{5.21}$$

The factor of 4 in Eq. (5.20) arises from $D=2$ and the fact that $d_{i\,j\,k}^{\omega_B\omega_O\omega_A}$ refers to one of the two frequency components $\omega_O\pm\omega_A$ produced, while r_{lmk} is quoted as if for a static value ($\omega_A=0$). The *direct* photoelastic susceptibility[5] $\chi_{i\,j\,kl}^{\omega_B\omega_O\omega_A}$ consists of a part symmetric in its elastic indices, indicated by parentheses, and a part antisymmetric,[4–6] indicated by brackets,

$$\chi_{i\,j\,kl}^{\omega_B\omega_O\omega_A}=\chi_{i\,j\,(kl)}^{\omega_B\omega_O\omega_A}+\chi_{i\,j\,[kl]}^{\omega_B\omega_O\omega_A} \ , \tag{5.22}$$

where

$$\chi_{i\,j\,(kl)}^{\omega_B\omega_O\omega_A}=-\tfrac{1}{2}\chi_{ij}(\omega_O)\delta_{kl}-\epsilon_0\sum_{\substack{\rho,\mu\\ \beta,\nu}}\Upsilon_{im}^{\rho\mu}(\omega_B)\Upsilon_{bj}^{\beta\nu}(\omega_O) \times q^\nu q^\rho\,^{(2,1)}H_{mbkl}^{\mu\beta}+3\epsilon_0^2\sum_{\substack{\rho,\mu,\beta\\ \gamma,\varphi,\nu}}\Upsilon_{im}^{\rho\mu}(\omega_B) \times\Upsilon_{bj}^{\beta\nu}(\omega_O)\Upsilon_{ce}^{\gamma\varphi}(\omega_A)q^\nu q^\rho\,^{(3,0)}H_{mbc}^{\mu\beta\gamma}\,^{(1,1)}H_{ekl}^{\varphi} \ , \tag{5.23}$$

$$\chi_{i\,j\,[kl]}^{\omega_B\omega_O\omega_A}=\tfrac{1}{4}[\kappa_{il}(\omega_B)\delta_{jk}-\kappa_{ik}(\omega_B)\delta_{jl} +\kappa_{lj}(\omega_O)\delta_{ik}-\kappa_{kj}(\omega_O)\delta_{il}] \ . \tag{5.24}$$

The symmetric part is related to the Pockels photoelastic tensor p_{mnkl} by

$$\chi_{i\,j\,(kl)}^{\omega_B\omega_O\omega_A}=-\tfrac{1}{2}\kappa_{im}(\omega_B)p_{mnkl}\kappa_{nj}(\omega_O) \ , \tag{5.25}$$

where the factor of 2 arises because p_{mnkl} is quoted as if a static value as done above for r_{lmk}. The Pockels photoelastic tensor relates the change in the inverse dielectric tensor to the strain by

$$(\delta\kappa^{-1})_{ij}=p_{ijkl}S_{kl} \ . \tag{5.26}$$

For the simplification of Eq. (5.1) we must also define the direct third-order optical mixing tensor by

$$e_{i\,f\,g\,h}^{\omega_4\omega_3\omega_2\omega_1}=-\epsilon_0^3\sum_{\substack{\nu,\eta,\xi,\epsilon\\ \mu,\beta,\gamma,\delta}}q^\nu q^\eta q^\xi q^\epsilon\Upsilon_{ij}^{\nu\mu}(\omega_4)\Upsilon_{bf}^{\beta\eta}(\omega_3)\Upsilon_{cg}^{\gamma\xi}(\omega_2)\Upsilon_{dh}^{\delta\epsilon}(\omega_1)\,^{(4,0)}H_{jbcd}^{\mu\beta\gamma\delta}+\tfrac{3}{2}\epsilon_0^4\sum_{\substack{\nu,\mu,\gamma,\eta,\epsilon\\ \beta,\delta,\rho,\xi,\zeta}}q^\nu q^\eta q^\xi q^\epsilon\Upsilon_{ij}^{\nu\mu}(\omega_4)$$
$$\times[\Upsilon_{ch}^{\gamma\eta}(\omega_1)\Upsilon_{bd}^{\beta\delta}(\omega_{23})\Upsilon_{af}^{\rho\xi}(\omega_3)\Upsilon_{eg}^{\zeta\epsilon}(\omega_2)+\Upsilon_{cf}^{\gamma\eta}(\omega_3)\Upsilon_{bd}^{\beta\delta}(\omega_{12})\Upsilon_{ag}^{\rho\xi}(\omega_2)\Upsilon_{eh}^{\zeta\epsilon}(\omega_1)$$
$$+\Upsilon_{cg}^{\gamma\eta}(\omega_2)\Upsilon_{bd}^{\beta\delta}(\omega_{31})\Upsilon_{ah}^{\rho\xi}(\omega_1)\Upsilon_{ef}^{\zeta\epsilon}(\omega_3)]\,^{(3,0)}H_{jbc}^{\mu\beta\gamma}\,^{(3,0)}H_{dae}^{\delta\rho\zeta} \ , \tag{5.27}$$

where

$$\omega_4=\omega_1+\omega_2+\omega_3 \ , \tag{5.28}$$

$$\omega_{12}=\omega_1+\omega_2 \ , \tag{5.29}$$

$$\omega_{23}=\omega_2+\omega_3 \ , \tag{5.30}$$

$$\omega_{31}=\omega_3+\omega_1 \ . \tag{5.31}$$

It relates three input electric field amplitudes to a nonlinear polarization by

$$\mathcal{P}_i^{\omega_4}+D\epsilon_0 e_{i\,f\,g\,h}^{\omega_4\omega_3\omega_2\omega_1}E_f^{\omega_3}E_g^{\omega_2}E_h^{\omega_1} \ , \tag{5.32}$$

where

$D=1$ if all of ω_1, ω_2, ω_3 are equal
$\ \ =3$ if two of ω_1, ω_2, ω_3 are equal
$\ \ =6$ if none of ω_1, ω_2, ω_3 are equal .

By substituting Eqs. (5.2)–(5.11) into Eq. (5.1) and using the definitions in Eqs. (5.15), (5.18), (5.22)–(5.24), and (5.27) we are led to define a susceptibility $\chi_{i\,f\,g\,kl}^{\omega_C\omega_1\omega_2\omega_A}$ responsible for the *direct* mixing of three optical waves with one acoustic wave by

$$\chi_{i\,f\,g\,kl}^{\omega_C\omega_1\omega_2\omega_A}=\chi_{i\,f\,g\,(kl)}^{\omega_C\omega_1\omega_2\omega_A}+\chi_{i\,f\,g\,[kl]}^{\omega_C\omega_1\omega_2\omega_A} \ , \tag{5.33}$$

$$\chi_{i\,f\,g\,(kl)}^{\omega_C\omega_1\omega_2\omega_A} = \frac{-3\epsilon_0^2}{4}\sum_{\substack{\nu,\xi,\eta\\ \mu,\beta,\gamma}} q^\nu q^\xi q^\eta \Upsilon_{ij}^{\nu\mu}(\omega_C)\Upsilon_{bf}^{\beta\xi}(\omega_1)\Upsilon_{cg}^{\gamma\eta}(\omega_2)\Big({}^{(3,1)}H_{jbckl}^{\mu\beta\gamma} - 4\epsilon_0\sum_{\zeta,\varphi}{}^{(4,0)}H_{jbcp}^{\mu\beta\gamma\zeta}\Upsilon_{pq}^{\zeta\varphi}(\omega_A)\,{}^{(1,1)}H_{qkl}^{\varphi}$$

$$-2\epsilon_0\sum_{\rho,\delta}\big[{}^{(3,0)}H_{dbc}^{\rho\beta\gamma}\Upsilon_{de}^{\rho\delta}(\omega_{12})\,{}^{(2,1)}H_{ejkl}^{\delta\mu} + {}^{(3,0)}H_{jdc}^{\mu\rho\gamma}\Upsilon_{de}^{\rho\delta}(\omega_{1A})\,{}^{(2,1)}H_{ebkl}^{\delta\beta} + {}^{(3,0)}H_{jbd}^{\mu\beta\rho}\Upsilon_{de}^{\rho\delta}(\omega_{2A})\,{}^{(2,1)}H_{eckl}^{\delta\gamma}$$

$$+6\epsilon_0^2\sum_{\rho,\delta,\zeta,\varphi}\big[{}^{(3,0)}H_{dbc}^{\rho\beta\gamma}\Upsilon_{de}^{\rho\delta}(\omega_{12})\,{}^{(3,0)}H_{jep}^{\mu\delta\zeta}\Upsilon_{pq}^{\zeta\varphi}(\omega_A)\,{}^{(1,1)}H_{qkl}^{\varphi} + {}^{(3,0)}H_{jdc}^{\mu\rho\gamma}\Upsilon_{de}^{\rho\delta}(\omega_{1A})\,{}^{(3,0)}H_{ebp}^{\delta\beta\zeta}\Upsilon_{pq}^{\zeta\varphi}(\omega_A)\,{}^{(1,1)}H_{qkl}^{\varphi}$$

$$+{}^{(3,0)}H_{jdb}^{\mu\rho\beta}\Upsilon_{de}^{\rho\delta}(\omega_{2A})\,{}^{(3,0)}H_{epc}^{\delta\zeta\gamma}\Upsilon_{pq}^{\zeta\varphi}(\omega_A)\,{}^{(1,1)}H_{qkl}^{\varphi}\big]\Big) - \tfrac{1}{2}d_{i\,f\,g}^{\omega_{12}\omega_1\omega_2}\delta_{kl} \qquad (5.34)$$

and

$$\chi_{i\,f\,g\,[kl]}^{\omega_C\omega_1\omega_2\omega_A} = -\frac{3\epsilon_0^2}{8}\sum_{\substack{\nu,\xi,\eta\\ \mu,\beta,\gamma}} q^\nu q^\xi q^\eta \Upsilon_{ij}^{\nu\mu}(\omega_C)\Upsilon_{bf}^{\beta\xi}(\omega_1)\Upsilon_{cg}^{\gamma\eta}(\omega_2)\Big({}^{(3,0)}H_{lbc}^{\mu\beta\gamma}\delta_{jk} - {}^{(3,0)}H_{kbc}^{\mu\beta\gamma}\delta_{jl} + {}^{(3,0)}H_{jlc}^{\mu\beta\gamma}\delta_{bk} - {}^{(3,0)}H_{jkc}^{\mu\beta\gamma}\delta_{bl}$$

$$+{}^{(3,0)}H_{jbl}^{\mu\beta\gamma}\delta_{ck} - {}^{(3,0)}H_{jbk}^{\mu\beta\gamma}\delta_{cl} - 2\epsilon_0\sum_{\rho,\delta}\big[{}^{(3,0)}H_{dbc}^{\rho\beta\gamma}\Upsilon_{de}^{\rho\delta}(\omega_{12})\,{}^{(2,0)}H_{el}^{\delta\mu}\delta_{kj} - {}^{(3,0)}H_{dbc}^{\rho\beta\gamma}\Upsilon_{de}^{\rho\delta}(\omega_{12})\,{}^{(2,0)}H_{ek}^{\delta\mu}\delta_{lj}$$

$$+{}^{(3,0)}H_{jdc}^{\mu\rho\gamma}\Upsilon_{de}^{\rho\delta}(\omega_{1A})\,{}^{(2,0)}H_{el}^{\delta\beta}\delta_{kb} - {}^{(3,0)}H_{jdc}^{\mu\rho\gamma}\Upsilon_{de}^{\rho\delta}(\omega_{1A})\,{}^{(2,0)}H_{ek}^{\delta\beta}\delta_{lb} + {}^{(3,0)}H_{jbd}^{\mu\beta\rho}\Upsilon_{de}^{\rho\delta}(\omega_{2A})\,{}^{(2,0)}H_{el}^{\delta\gamma}\delta_{kc}$$

$$-{}^{(3,0)}H_{jbd}^{\mu\beta\rho}\Upsilon_{de}^{\rho\delta}(\omega_{2A})\,{}^{(2,0)}H_{ek}^{\delta\gamma}\delta_{lc} + {}^{(3,0)}H_{dbc}^{\rho\beta\gamma}\Upsilon_{dk}^{\rho\delta}(\omega_{12})\,{}^{(2,0)}H_{jl}^{\mu\delta} - {}^{(3,0)}H_{dbc}^{\rho\beta\gamma}\Upsilon_{dl}^{\rho\delta}(\omega_{12})\,{}^{(2,0)}H_{jk}^{\mu\delta}$$

$$+{}^{(3,0)}H_{jdc}^{\mu\rho\gamma}\Upsilon_{dk}^{\rho\delta}(\omega_{1A})\,{}^{(2,0)}H_{lb}^{\delta\beta} - {}^{(3,0)}H_{jdc}^{\mu\rho\gamma}\Upsilon_{dl}^{\rho\delta}(\omega_{1A})\,{}^{(2,0)}H_{kb}^{\delta\beta}$$

$$+{}^{(3,0)}H_{jbd}^{\mu\beta\rho}\Upsilon_{dk}^{\rho\delta}(\omega_{2A})\,{}^{(2,0)}H_{lc}^{\delta\gamma} - {}^{(3,0)}H_{jbd}^{\mu\beta\rho}\Upsilon_{dl}^{\rho\delta}(\omega_{2A})\,{}^{(2,0)}H_{kc}^{\delta\gamma}\big]\Big)\,, \qquad (5.35)$$

where

$$\omega_C = \omega_1 + \omega_2 + \omega_A\,, \qquad (5.36)$$

$$\omega_{12} = \omega_1 + \omega_2\,, \qquad (5.37)$$

$$\omega_{1A} = \omega_1 + \omega_A\,, \qquad (5.38)$$

$$\omega_{2A} = \omega_2 + \omega_A\,. \qquad (5.39)$$

Equation (5.1) now can be expressed as

$$\mathcal{P}_i(2,1) = \epsilon_0\Bigg[\chi_{i\,f\,g\,kl}^{\omega_C\omega_O\omega_O\omega_A} + 3e_{i\,f\,g\,h}^{\omega_C\omega_O\omega_O\omega_A}\frac{a_h a_s e_{skl}^{\omega_A}}{\epsilon_0 a_p \kappa_{pq}(\omega_A)a_q} + 2d_{i\,f\,j}^{\omega_C\omega_O\omega_B}\sum_{\varphi=1}^{3}\frac{\mathcal{E}_j^{(\varphi)}(\vec{s}_F,\omega_B)\mathcal{E}_m^{(\varphi)}(\vec{s}_F,\omega_B)}{(|\vec{k}_O+\vec{k}_A|^2/|\vec{k}_B|^2-1)}$$

$$\times\left(\chi_{m\,g\,kl}^{\omega_B\omega_O\omega_A} - \frac{2d_{m\,g\,r}^{\omega_B\omega_O\omega_A}a_r a_s e_{skl}^{\omega_A}}{\epsilon_0 a_p\kappa_{pq}(\omega_A)a_q}\right) + \left(\chi_{i\,j\,kl}^{\omega_C\omega_H\omega_A} - \frac{2d_{i\,j\,r}^{\omega_C\omega_H\omega_A}a_r a_s e_{skl}^{\omega_A}}{\epsilon_0 a_p\kappa_{pq}(\omega_A)a_q}\right)$$

$$\times\sum_{\varphi=1}^{3}\frac{\mathcal{E}_j^{(\varphi)}(\vec{s}_I,\omega_H)\mathcal{E}_m^{(\varphi)}(\vec{s}_I,\omega_H)}{(|2\vec{k}_O|^2/|k_H|^2-1)}\,d_{m\,f\,g}^{\omega_H\omega_O\omega_O}\Bigg]E_f(1,0)E_g(1,0)u_{k,l}(0,1) \qquad (5.40)$$

for the case $\omega_1=\omega_2=\omega_O$.

Equation (5.40) in conjunction with Eqs. (5.33)–(5.35) is one of the two major results of this work, the concept of double phase matching to be discussed in Sec. VI being the other. The effective susceptibility controlling the AIOHG process is contained in the braces of Eq. (5.40). The direct-effect term and the many indirect-effect terms appearing there have been described already in the Introduction in the order written. We will not repeat that description here.

The form of the denominators of four of the indirect processes—direct and indirect acousto-optic scattering followed by second-order optical mixing and harmonic generation followed by direct and indirect acousto-optic scattering—suggests the possibility of phase matching the fields at the intermediate step. When this is done the free wave at this intermediate step must be introduced into the development as well as the forced wave, which was the only one used in the derivation of Eq. (5.40). These highly interesting cases will be considered in Sec. VI.

If the two input optical frequencies are different, the analog to Eq. (5.40) is

$$\mathcal{P}_i^{\omega_C\omega_1\omega_2\omega_A} = \epsilon_0\Bigg[2\chi_{i\,f\,g\,kl}^{\omega_C\omega_1\omega_2\omega_A} + 6\frac{e_{i\,f\,g\,h}^{\omega_C\omega_1\omega_2\omega_A}a_h a_s e_{skl}^{\omega_A}}{\epsilon_0 a_p\kappa_{pq}(\omega_A)a_q} + 2d_{i\,g\,j}^{\omega_C\omega_2\omega_{1A}}\sum_{\varphi=1}^{3}\frac{\mathcal{E}_j^{(\varphi)}(\vec{s}_{F1},\omega_{1A})\mathcal{E}_m^{(\varphi)}(\vec{s}_{F1},\omega_{1A})}{(|\vec{k}_1+\vec{k}_A|^2/|\vec{k}_{1A}|^2-1)}$$

$$\times \left(\chi_{m\,f\,kl}^{\omega_{1A}\omega_1\omega_A} - \frac{2 d_{m\,f\,r}^{\omega_{1A}\omega_1\omega_A} a_r a_s e_{skl}^{\omega_A}}{\epsilon_0 a_p \kappa_{pq}(\omega_A) a_q} \right) + 2 d_{i\,f\,j}^{\omega_C\omega_1\omega_{2A}} \sum_{\varphi=1}^{3} \frac{\mathcal{E}_j^{(\varphi)}(\vec{s}_{F2}, \omega_{2A}) \mathcal{E}_m^{(\varphi)}(\vec{s}_{F2}, \omega_{2A})}{(|\vec{k}_2 + \vec{k}_A|^2 / |\vec{k}_{2A}|^2 - 1)}$$

$$\times \left(\chi_{m\,g\,kl}^{\omega_{2A}\omega_2\omega_A} - \frac{2 d_{m\,g\,r}^{\omega_{2A}\omega_2\omega_A} a_r a_s e_{skl}^{\omega_A}}{\epsilon_0 a_p \kappa_{pq}(\omega_A) a_q} \right) + 2 \left(\chi_{i\,j\,kl}^{\omega_C\omega_{12}\omega_A} - \frac{2 d_{i\,j\,r}^{\omega_C\omega_{12}\omega_A} a_r a_s e_{skl}^{\omega_A}}{\epsilon_0 a_p \kappa_{pq}(\omega_A) a_q} \right)$$

$$\times \sum_{\varphi=1}^{3} \frac{\mathcal{E}_j^{(\varphi)}(\vec{s}_M, \omega_{12}) \mathcal{E}_m^{(\varphi)}(\vec{s}_M, \omega_{12})}{(|\vec{k}_1 + \vec{k}_2|^2 / |\vec{k}_{12}|^2 - 1)} d_{m\,f\,g}^{\omega_{12}\omega_1\omega_2} \Big] E_f^{\omega_1} E_g^{\omega_2} u_{k,l}^{\omega_A} \quad , \tag{5.41}$$

where the frequencies are defined in Eqs. (5.36)–(5.39), [the frequency notation is such that $\vec{\mathcal{P}}_i^{\omega_C\omega_O\omega_O\omega_A} = \mathcal{P}_i(2,1)$ of Eq. (5.40)], $\vec{k}_{1A}$, $\vec{k}_{2A}$, and $\vec{k}_{12}$ are the wave vectors of the free waves at frequencies ω_{1A}, ω_{2A}, and ω_{12}, and the unit vectors $\vec{s}_{F1}$, $\vec{s}_{F2}$, and $\vec{s}_M$ are defined by

$$\vec{s}_{F1} = (\vec{k}_1 + \vec{k}_A)/|\vec{k}_1 + \vec{k}_A| \quad , \tag{5.42}$$

$$\vec{s}_{F2} = (\vec{k}_2 + \vec{k}_A)/|\vec{k}_2 + \vec{k}_A| \quad , \tag{5.43}$$

$$\vec{s}_M = (\vec{k}_1 + \vec{k}_2)/|\vec{k}_1 + \vec{k}_2| \quad . \tag{5.44}$$

The antisymmetric part of the susceptibility given by Eq. (5.35) can be simplified by the use of Eq. (3.20). If terms of order (ω_A/ω_O) are then dropped, we find simply that

$$\chi_{i\,f\,g\,[kl]}^{\omega_C\omega_1\omega_2\omega_A} = \tfrac{1}{4} (d_{l\,f\,g}^{\omega_{12}\omega_1\omega_2} \delta_{ik} - d_{k\,f\,g}^{\omega_{12}\omega_1\omega_2} \delta_{il} + d_{i\,l\,g}^{\omega_C\omega_{1A}\omega_2} \delta_{fk} - d_{i\,k\,g}^{\omega_C\omega_{1A}\omega_2} \delta_{fl} + d_{i\,f\,l}^{\omega_C\omega_1\omega_{2A}} \delta_{gk} - d_{i\,f\,k}^{\omega_C\omega_1\omega_{2A}} \delta_{gl}). \tag{5.45}$$

Concerning Eq. (5.45) we note the following: (a) $\chi_{i\,f\,g\,[kl]}^{\omega_C\omega_1\omega_2\omega_A}$ contains only shear components in the elastic indices; (b) any shear distortion causes a rotation; (c) rotation of a lower-order optical anisotropy should contribute to a higher-order optical effect; and (d) Eq. (5.45) depends only on the second-order optical mixing tensor. We thus interpret Eq. (5.45) as the contribution to the total susceptibility governing AIOHG caused by rotation of the optical mixing tensor in the presence of an elastic shear distortion. Equation (5.45) can, in fact, be derived on this basis from a very simple kinematic argument given in Appendix B. It is completely analogous to the rotational effects recently predicted[4,5] and observed[6] to occur in ordinary acousto-optic and Brillouin scattering. The presence of the tensor asymmetric in the elastic indices in Eqs. (5.33) and (5.35) once again[4,5] demonstrates the fact that the measure of elastic deformation relevant to acousto-optic effects is not just the strain but rather the strain and the rotation, or more simply, just the displacement gradient as indicated in Eq. (5.40).

The antisymmetric susceptibility of Eq. (5.45) will be zero for all centrosymmetric crystals since $d_{ijk} = 0$ for them. It will be nonzero for all acentric crystal classes except the cubic 432 class, regardless of the input optical frequencies. For class 432 it will be nonzero only when the input optical frequencies are different. It would also be zero for an acentric isotropic noncrystalline medium.

The antisymmetric susceptibility of Eq. (5.45) can be comparable in magnitude to the symmetric part for many crystals. For instance, for GaAs we predict

$$\chi_{122[23]} = + \tfrac{1}{2} d_{123} = +0.65 \times 10^{-10}\ \mathrm{m/V}$$

on the basis of a recent measurement[8] of d_{123}, while measurements[1] yielded

$$\chi_{12223} = \chi_{122(23)} + \chi_{122[23]} = (+0.8 \text{ or } +1.1) \times 10^{-10}\ \mathrm{m/V} \quad .$$

Thus it is seen that rotations in shear waves can give effects in AIOHG comparable to those of strains.

The symmetric part of the *direct* susceptibility representing AIOHG, given in Eq. (5.34), cannot be expressed in terms of directly measurable lower-order tensors (except for the last term). As such, it represents a new measurable tensor that characterizes the material medium. We will term the various contributions to the *direct* susceptibility as *internal* contributions. The first one in Eq. (5.34) represents a one-step mixing of the two input optical fields and the one input acoustic field via the ${}^{(3,1)}\overleftrightarrow{H}$ material descriptor. The second term corresponds to a two-step mixing of the polarizations induced by the input optical fields via ${}^{(4,0)}\overleftrightarrow{H}$ with the internal displacement produced via ${}^{(1,1)}\overleftrightarrow{H}$ by the input acoustic field. The third group of terms represents a two-step mixing process: First, the polarization induced by one of the input optical fields is mixed with the displacement gradient of the input acoustic field via ${}^{(2,1)}\overleftrightarrow{H}$; the resultant internal displacement mixes with the polarization induced by the other input optical field via ${}^{(3,0)}\overleftrightarrow{H}$. The fourth group of terms corresponds to a three-step mixing process: First, the displacement gradient of the acoustic wave produces an internal displacement via ${}^{(1,1)}\overleftrightarrow{H}$; the resultant internal displacement mixes via ${}^{(3,0)}\overleftrightarrow{H}$ with the polarization induced by an input optical field; the internal displacement resulting from the latter mixing then mixes with the polarization induced by the other input optical field via ${}^{(3,0)}\overleftrightarrow{H}$ again. The last contribution in Eq. (5.34) is expressible in terms of a lower-order directly measurable tensor and represents simply the change in density of nonlinear oscillators arising from compressional or dilatational components of the strain.

Equation (5.34) predicts the form of the frequency dispersion of $\chi_{i\ f\ g\ kl}^{\omega_C\omega_1\omega_2\omega_A}$. All frequency dependence is seen to enter via the $\Upsilon_{ij}^{\nu\mu}(\omega)$ susceptibilitylike factors which have arisen from the solution of the dynamics of the problem. The $^{(m,n)}\overleftrightarrow{\mathrm{H}}$ material descriptors by their nature are frequency-independent quantities. Note particularly that contributions to the susceptibility having different symmetry have different dispersion. A sufficiently complete and accurate dispersion analysis could be used to determine $^{(3,1)}\overleftrightarrow{\mathrm{H}}$. For the analysis it would be necessary to have found $^{(1,1)}\overleftrightarrow{\mathrm{H}}$ from the piezoelectric tensor, $\overleftrightarrow{\Upsilon}$ from the dielectric tensor, $^{(3,0)}\overleftrightarrow{\mathrm{H}}$ from the electro-optic and the second-order optical mixing tensors, $^{(2,1)}\overleftrightarrow{\mathrm{H}}$ from the photoelastic tensor, and $^{(4,0)}\overleftrightarrow{\mathrm{H}}$ from the third-order optical mixing tensor. To perform such an analysis it is necessary to decide how many internal coordinates (number of oscillators) are important to the experimental results. This determines the range of the summations over Greek-letter superscripts. Even if a single electronic and a single ionic degree of freedom are assumed for GaAs, $^{(3,1)}\overleftrightarrow{\mathrm{H}}$ values cannot be deduced from the measured AIOHG susceptibility for GaAs[1] because one of the interacting waves (the acoustic wave) in that experiment is a low-frequency wave and because for many of the nonlinear interactions there are no data for GaAs for interacting waves, one of which has a frequency below the ionic (infrared) resonance which could be used to distinguish between ionic and electronic parts of the various $^{(m,n)}\overleftrightarrow{\mathrm{H}}$.

The symmetry of the susceptibility of Eq. (5.34) for a given crystal class can be determined from the symmetry of the individual $^{(m,n)}\overleftrightarrow{\mathrm{H}}$. Alternatively, and more simply, its symmetry can be found by applying the point-group operations to a fifth-rank tensor which is symmetric upon interchange of its last two indices. Being an odd-rank tensor, it will be nonzero only in acentric crystals. If we take the input frequencies identical, $\omega_1=\omega_2$, then the tensor will also be symmetric upon interchange of the second and third indices. As this is likely to be the most common experimental situation, we list the symmetry of the *symmetric direct* susceptibility expected for this case in Table I. If $\omega_1\neq\omega_2$, but they are close enough that there is no significant dispersion of the optical properties between these frequencies, then the symmetry of Table I will be applicable to high accuracy. If ω_1, ω_2, and ω_C all lie in an essentially dispersionless region, then it is apparent from Eq. (5.34) that

$$\chi_{i\ f\ g\ (kl)}^{\omega_C\omega_1\omega_2\omega_A}=\chi^{\omega_C\omega_1\omega_2\omega_A}\ . \tag{5.46}$$

Conceptually this is the same as the Kleinman sym-

TABLE I. Form of a fifth-rank material tensor which is symmetric upon interchange of the second with the third index and upon interchange of the fourth with the fifth index for each of the 21 accentric point groups is presented. All elements of the tensor are zero for the centrosymmetric point groups (triclinic $\bar{1}$, monoclinic $2/m$, orthorhombic mmm, trigonal $\bar{3}$ and $\bar{3}\,m$, tetragonal $4/m$ and $4/mmm$, hexagonal $6/m$ and $6/mmm$, and cubic $m3$ and $m3m$). The tensor represents the elastically symmetric direct susceptibility governing AIOHG, given in Eq. (5.34) for the case that the two input electric fields are of the same wavelength (or close enough that dispersion is negligible—see text). In the table $\chi_{i(fg)(kl)}$ is contracted to χ_{iab}, where $a=1, 2, 3, 4, 5, 6$ for $(f,g)=(1,1)$, $(2,2)$, $(3,3)$, $(2,3)$ or $(3,2)$, $(3,1)$ or $(1,3)$, $(1,2)$ or $(2,1)$, respectively, and b represents (k,l), similarly. Zero elements are indicated by a dot; nonzero elements by letters; related elements are expressed in terms of the same letters except when only X appears, for which cases no relation exists between the nonzero elements.

	$i=1$						$i=2$						$i=3$					
	$a=1$	2	3	4	5	6	1	2	3	4	5	6	1	2	3	4	5	6
Triclinic 1																		
$b=1$	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
2	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
3	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
4	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
5	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
6	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
Monoclinic 2																		
$b=1$	·	·	·	X	·	X	X	X	X	·	X	·	·	·	·	X	·	X
2	·	·	·	X	·	X	X	X	X	·	X	·	·	·	·	X	·	X
3	·	·	·	X	·	X	X	X	X	·	X	·	·	·	·	X	·	X
4	X	X	X	·	X	·	·	·	·	X	·	X	X	X	X	·	X	·
5	·	·	·	X	·	X	X	X	X	·	X	·	·	·	·	X	·	X
6	X	X	X	·	X	·	·	·	·	X	·	X	X	X	X	·	X	·

TABLE I. (*continued*)

Monoclinic *m*

b																		
b=1	X	X	X	·	X	·	·	·	·	X	·	X	X	X	X	·	X	·
2	X	X	X	·	X	·	·	·	·	X	·	X	X	X	X	·	X	·
3	X	X	X	·	X	·	·	·	·	X	·	X	X	X	X	·	X	·
4	·	·	·	X	·	X	X	X	X	·	X	·	·	·	·	X	·	X
5	X	X	X	·	X	·	·	·	·	X	·	X	X	X	X	·	X	·
6	·	·	·	X	·	X	X	X	X	·	X	·	·	·	·	X	·	X

Orthorhombic 222

b																		
b=1	·	·	·	X	·	·	·	·	·	·	X	·	·	·	·	·	·	X
2	·	·	·	X	·	·	·	·	·	·	X	·	·	·	·	·	·	X
3	·	·	·	X	·	·	·	·	·	·	X	·	·	·	·	·	·	X
4	X	X	X	·	·	·	·	·	·	·	·	X	·	·	·	·	X	·
5	·	·	·	·	·	X	X	X	X	·	·	·	·	·	·	X	·	·
6	·	·	·	·	X	·	·	·	·	X	·	·	X	X	X	·	·	·

Orthorhomic *mm*2

b																		
b=1	·	·	·	·	X	·	·	·	·	X	·	·	X	X	X	·	·	·
2	·	·	·	·	X	·	·	·	·	X	·	·	X	X	X	·	·	·
3	·	·	·	·	X	·	·	·	·	X	·	·	X	X	X	·	·	·
4	·	·	·	·	·	X	X	X	X	·	·	·	·	·	·	X	·	·
5	X	X	X	·	·	·	·	·	·	·	·	X	·	·	·	·	X	·
6	·	·	·	X	·	·	·	·	·	·	X	·	·	·	·	·	·	X

Trigonal 3$[D \equiv -(A+B+C);\ m = -(j+k+l)]$

b																		
b=1	$2A$	$2B$	$-E$	$2d$	$2I$	$l-m$	$2j$	$2k$	n	$2J$	$-2e$	$D-C$	$2N$	$2P$	Q	r	$-U$	t
2	$2C$	$2D$	E	$2e$	$2J$	$j-k$	$2l$	$2m$	$-n$	$2I$	$-2d$	$B-A$	$2P$	$2N$	Q	$-r$	U	$-t$
3	$-F$	F	·	f	K	o	o	$-o$	·	K	$-f$	F	R	R	S	·	·	·
4	$2a$	$2b$	c	M	p	$G-H$	$2H$	$2G$	L	$-p$	M	$a-b$	q	$-q$	·	T	s	O
5	$2G$	$2H$	L	p	$-M$	$b-a$	$-2b$	$-2a$	$-c$	M	p	$G-H$	$-O$	O	·	$-s$	T	q
6	$k-m$	$j-l$	n	$I-J$	$e-d$	$B+C$	$D-B$	$C-A$	E	$d-e$	$I-J$	$k+l$	$-t$	t	·	U	r	$N-P$

Trigonal 3*m*$[D \equiv -(A+B+C)]$

b																		
b=1	$2A$	$2B$	$-E$	·	$2I$	·	·	·	·	$2J$	·	$D-C$	$2N$	$2P$	Q	·	$-U$	·
2	$2C$	$2D$	E	·	$2J$	·	·	·	·	$2I$	·	$B-A$	$2P$	$2N$	Q	·	U	·
3	$-F$	F	·	·	K	·	·	·	·	K	·	F	R	R	S	·	·	·
4	·	·	·	M	·	$G-H$	$2H$	$2G$	L	·	M	·	·	·	·	T	·	Z
5	$2G$	$2H$	L	·	$-M$	·	·	·	·	M	·	$G-H$	$-Z$	Z	·	·	T	·
6	·	·	·	$I-J$	·	$B+C$	$D-B$	$C-A$	E	·	$I-J$	·	·	·	·	U	·	$N-P$

Trigonal 32 $[m \equiv -(j+k+l)]$

b																		
b=1	·	·	·	$2d$	·	$l-m$	$2j$	$2k$	n	·	$-2e$	·	·	·	·	r	·	t
2	·	·	·	$2e$	·	$j-k$	$2l$	$2m$	$-n$	·	$-2d$	·	·	·	·	$-r$	·	$-t$
3	·	·	·	f	·	z	z	$-z$	·	·	$-f$	·	·	·	·	·	·	·
4	$2a$	$2b$	c	·	p	·	·	·	·	$-p$	·	$a-b$	q	$-q$	·	·	s	·
5	·	·	·	p	·	$b-a$	$-2b$	$-2a$	$-c$	·	p	·	·	·	·	$-s$	·	q
6	$k-m$	$j-l$	n	·	$e-d$	·	·	·	·	$d-e$	·	$k+l$	$-t$	t	·	·	r	·

Tetragonal 4

b																		
b=1	·	·	·	I	M	·	·	·	·	N	$-J$	·	Q	R	S	·	·	Z
2	·	·	·	J	N	·	·	·	·	M	$-I$	·	R	Q	S	·	·	$-Z$
3	·	·	·	K	O	·	·	·	·	O	$-K$	·	T	T	U	·	·	·
4	A	B	C	·	·	D	F	E	G	·	·	$-H$	·	·	·	V	W	·
5	E	F	G	·	·	H	$-B$	$-A$	$-C$	·	·	D	·	·	·	$-W$	V	·
6	·	·	·	L	P	·	·	·	·	$-P$	L	·	Y	$-Y$	·	·	·	X

TABLE I. (*continued*)

Tetragonal $\bar{4}$

$b=1$	·	·	·	I	M	·	·	·	·	$-N$	J	·	Q	R	S	·	·	Z
2	·	·	·	J	N	·	·	·	·	$-M$	I	·	$-R$	$-Q$	$-S$	·	·	Z
3	·	·	·	K	W	·	·	·	·	$-W$	K	·	T	$-T$	·	·	·	X
4	A	B	C	·	·	D	$-F$	$-E$	$-G$	·	·	H	·	·	·	V	·	·
5	E	F	G	·	·	H	B	A	C	·	·	$-D$	·	·	·	·	$-V$	·
6	·	·	·	L	P	·	·	·	·	P	$-L$	·	Y	Y	U	·	·	·

Tetragonal 422

$b=1$	·	·	·	I	·	·	·	·	·	·	$-J$	·	·	·	·	·	·	Z
2	·	·	·	J	·	·	·	·	·	·	$-I$	·	·	·	·	·	·	$-Z$
3	·	·	·	K	·	·	·	·	·	·	$-K$	·	·	·	·	·	·	·
4	A	B	C	·	·	·	·	·	·	·	·	$-H$	·	·	·	·	W	·
5	·	·	·	·	·	H	$-B$	$-A$	$-C$	·	·	·	·	·	·	$-W$	·	·
6	·	·	·	·	P	·	·	·	·	$-P$	·	·	Y	$-Y$	·	·	·	·

Tetragonal $4mm$

$b=1$	·	·	·	·	M	·	·	·	·	N	·	·	Q	R	S	·	·	·
2	·	·	·	·	N	·	·	·	·	M	·	·	R	Q	S	·	·	·
3	·	·	·	·	W	·	·	·	·	W	·	·	T	T	U	·	·	·
4	·	·	·	·	·	D	F	E	G	·	·	·	·	·	·	V	·	·
5	E	F	G	·	·	·	·	·	·	·	·	D	·	·	·	·	V	·
6	·	·	·	L	·	·	·	·	·	·	L	·	·	·	·	·	·	X

Tetragonal $\bar{4}2m$

$b=1$	·	·	·	I	·	·	·	·	·	·	J	·	·	·	·	·	·	Z
2	·	·	·	J	·	·	·	·	·	·	I	·	·	·	·	·	·	Z
3	·	·	·	K	·	·	·	·	·	·	K	·	·	·	·	·	·	X
4	A	B	C	·	·	·	·	·	·	·	·	H	·	·	·	·	W	·
5	·	·	·	·	·	H	B	A	C	·	·	·	·	·	·	W	·	·
6	·	·	·	·	P	·	·	·	·	P	·	·	Y	Y	U	·	·	·

Hexagonal 6

$b=1$	·	·	·	$2D$	$2J$	·	·	·	·	$2K$	$-2E$	·	$2M$	$2N$	O	·	·	T
2	·	·	·	$2E$	$2K$	·	·	·	·	$2J$	$-2D$	·	$2N$	$2M$	O	·	·	$-T$
3	·	·	·	F	L	·	·	·	·	L	$-F$	·	P	P	Q	·	·	·
4	$2A$	$2B$	C	·	·	$G-H$	$2H$	$2G$	T	·	·	$A-B$	·	·	·	R	S	·
5	$2G$	$2H$	I	·	·	$B-A$	$-2B$	$-2A$	$-C$	·	·	$G-H$	·	·	·	$-S$	R	·
6	·	·	·	$J-K$	$E-D$	·	·	·	·	$D-E$	$J-K$	·	$-T$	T	·	·	·	$M-N$

Hexagonal $\bar{6}$ [$D\equiv-(A+B+C)$; $M\equiv-(J+K+L)$]

$b=1$	$2A$	$2B$	E	·	·	$L-M$	$2J$	$2K$	N	·	·	$D-C$	·	·	·	R	$-I$	·
2	$2C$	$2D$	$-E$	·	·	$J-K$	$2L$	$2M$	$-N$	·	·	$B-A$	·	·	·	$-R$	I	·
3	F	$-F$	·	·	·	O	O	$-O$	·	·	·	$-F$	·	·	·	·	·	·
4	·	·	·	G	P	·	·	·	·	$-P$	G	·	Q	$-Q$	·	·	·	H
5	·	·	·	P	$-G$	·	·	·	·	G	P	·	$-H$	H	·	·	·	Q
6	$K-M$	$J-L$	N	·	·	$B+C$	$D-B$	$C-A$	$-E$	·	·	$K+L$	·	·	·	I	R	·

Hexagonal 622

$b=1$	·	·	·	$2D$	·	·	·	·	·	·	$-2E$	·	·	·	·	·	·	T
2	·	·	·	$2E$	·	·	·	·	·	·	$-2D$	·	·	·	·	·	·	$-T$
3	·	·	·	F	·	·	·	·	·	·	$-F$	·	·	·	·	·	·	·
4	$2A$	$2B$	C	·	·	·	·	·	·	·	·	$A-B$	·	·	·	·	S	·
5	·	·	·	·	·	$B-A$	$-2B$	$-2A$	$-C$	·	·	·	·	·	·	$-S$	·	·
6	·	·	·	·	$E-D$	·	·	·	·	$D-E$	·	·	$-T$	T	·	·	·	·

TABLE I. (*continued*)

Hexagonal 6 *mm*

$b=1$	•	∘	∘	∘	$2J$	∘	∘	∘	∘	$2K$	∘	∘	$2M$	$2N$	O	∘	∘	•
2	∘	∘	•	∘	$2K$	•	•	∘	•	$2J$	∘	∘	$2N$	$2M$	O	•	∘	•
3	•	•	•	•	L	•	•	•	•	L	∘	∘	P	P	Q	•	•	•
4	•	∘	∘	•	∘	$G-H$	$2H$	$2G$	I	∘	•	∘	∘	∘	•	R	•	•
5	$2G$	$2H$	I	•	∘	•	•	•	•	∘	•	$G-H$	∘	•	•	•	R	•
6	•	∘	∘	$J-K$	•	•	•	•	•	•	$J-K$	∘	•	•	•	•	∘	$M-N$

Hexagonal $\bar{6}\,m2$ $[M \equiv -(J+K+L)]$

$b=1$	•	∘	•	•	•	$L-M$	$2J$	$2K$	N	•	•	•	•	•	∘	R	•	∘
2	•	∘	•	•	∘	$J-K$	$2L$	$2M$	$-N$	∘	•	•	∘	•	∘	$-R$	•	•
3	•	∘	•	•	•	O	O	$-O$	∘	•	•	∘	•	•	•	∘	•	∘
4	•	•	•	•	P	•	•	•	•	$-P$	•	•	Q	$-Q$	•	∘	•	∘
5	∘	•	•	P	•	•	∘	•	∘	•	P	∘	•	∘	∘	∘	∘	Q
6	$K-M$	$J-L$	N	•	•	∘	∘	•	∘	•	∘	$K+L$	•	∘	∘	∘	R	∘

Cubic 23

$b=1$	∘	•	∘	I	∘	•	∘	•	•	∘	K	•	∘	•	∘	∘	∘	J
2	∘	∘	∘	J	•	•	•	•	∘	∘	I	•	∘	•	∘	∘	∘	K
3	∘	∘	•	K	•	∘	∘	•	∘	∘	J	•	∘	•	∘	∘	∘	I
4	A	B	C	∘	∘	∘	•	∘	∘	•	•	P	∘	∘	•	•	H	∘
5	•	∘	∘	∘	•	H	C	A	B	∘	•	•	•	∘	∘	P	∘	∘
6	∘	∘	∘	∘	P	•	•	•	•	H	∘	∘	B	C	A	•	•	∘

Cubic 432

$b=1$	•	•	•	∘	•	∘	•	•	•	∘	$-J$	∘	•	•	•	∘	•	J
2	•	•	•	J	∘	•	•	•	•	∘	•	•	•	•	•	•	•	$-J$
3	∘	•	∘	$-J$	∘	•	•	•	∘	∘	J	•	∘	∘	•	•	∘	•
4	∘	B	$-B$	•	∘	•	∘	∘	•	•	•	$-H$	∘	•	•	•	H	•
5	•	∘	•	•	∘	H	$-B$	•	B	•	•	•	∘	•	∘	$-H$	•	∘
6	•	∘	∘	•	$-H$	•	∘	•	•	H	∘	∘	B	$-B$	•	•	•	•

Cubic $\bar{4}3\,m$

$b=1$	∘	•	•	I	∘	•	∘	•	•	•	J	∘	∘	•	∘	•	∘	J
2	∘	•	∘	J	•	∘	∘	•	∘	•	I	•	•	•	•	•	•	J
3	•	∘	•	J	•	•	•	∘	•	∘	J	•	•	∘	•	•	•	I
4	A	B	B	∘	•	∘	∘	•	•	•	∘	H	•	•	∘	•	H	∘
5	∘	•	∘	∘	•	H	B	A	B	•	∘	∘	∘	∘	∘	H	∘	∘
6	•	•	•	•	H	•	∘	•	∘	H	•	•	B	B	A	•	•	∘

metry condition for the second-order optical mixing coefficient.[9] The forms of the susceptibility tensor in Table I can also be used for the direct acoustically induced electro-optical effect if the first index is used for the low-frequency electric field and the second and third indices for the output and input optical electric fields.

VI. SOLUTION OF WAVE EQUATION FOR DOUBLE PHASE MATCHING

Double phase matching, as discussed in the Introduction, refers to phase matching both individual steps of an *indirect* contribution to a third-order (four-wave) interaction such as AIOHG.[10] In this case there are two types of indirect contributions which can be doubly phase matched under appropriate conditions: (a) acousto-optic scattering (direct and indirect) followed by second-order optical mixing, and (b) second-harmonic generation followed by acousto-optic scattering (direct and indirect). We will consider the former of these first.

When acousto-optic scattering is phase matched according to Eq. (1.1), the free wave must also be included in the nonlinear driving polarization of Eq. (3.25). This will cause this particular indirect contribution to dominate the nonlinear susceptibility of AIOHG for sufficiently large but reasonable interaction lengths. For this reason we will drop the remaining terms in the nonlinear driving polarization. Only the term in Eq. (3.12), proportional to $y_b^\beta(\vec{z};1,1)\,y_c^\gamma(\vec{z};1,0)$, need be retained. Into this term we substitute Eqs. (5.2) and (5.4), dropping the $\vec{F}^\mu(\vec{z};1,1)$ term from Eq. (5.4) because it cannot be phase matched. For $\vec{E}(\vec{z};1,1)$ in Eq. (5.4) we use

$$\vec{E}(\vec{z};1,1)=\frac{\vec{\mathcal{E}}^{(\eta)}(\vec{s}_F,\omega_B)\vec{\mathcal{E}}^{(\eta)}(\vec{s}_F,\omega_B)\cdot\vec{\mathcal{P}}(1,1)}{\epsilon_0(|\vec{k}_O+\vec{k}_A|^2/|\vec{k}_B|^2-1)}\times[e^{i(\vec{k}_O+\vec{k}_A)\cdot\vec{z}}-e^{i\vec{k}_B\cdot\vec{z}}] \quad (6.1)$$

from Ref. 5. Here η refers to the particular one of the eigenvectors that is phase matchable and $\vec{s}_F$ is given by Eq. (5.13). We obtain for the nonlinear driving polarization for this case

$$\vec{\mathcal{P}}^{DB}(\vec{z};(2,1)=\frac{\vec{P}^{DB}(2,1)}{(|\vec{k}_O+\vec{k}_A|^2/|\vec{k}_B|^2-1)}\times[e^{i(2\vec{k}_O+\vec{k}_A)\cdot\vec{z}}-e^{i(\vec{k}_O+\vec{k}_B)\cdot\vec{z}}], \quad (6.2)$$

where

$$\vec{P}_i^{DB}(2,1)=2\epsilon_0 d_{i\,f\,j}^{\omega_C\omega_O\omega_B}\mathcal{E}_j^{(\eta)}(\vec{s}_F,\omega_B)\mathcal{E}_m^{(\eta)}(\vec{s}_F,\omega_B)\times\left(\chi_{m\,g\,kl}^{\omega_B\omega_O\omega_A}-\frac{2d_{m\,g\,r}^{\omega_B\omega_O\omega_A}a_r a_s e_{skl}^{\omega_A}}{\epsilon_0 a_p\kappa_{pq}(\omega_A)a_q}\right)\times E_f(1,0)E_g(1,0)u_{k,l}(0,1). \quad (6.3)$$

Here the superscript DB stands for double phase matching of the intermediate step at frequency ω_B and denotes that part of the total $\vec{\mathcal{P}}(\vec{z};2,1)$ given in Eq. (3.25) that is important in the present situation.

The nonlinear polarization in Eq. (6.2) is now used on the right-hand side of Eq. (3.23). The general solution of the wave equation in the medium consists this time of one free and two forced waves according to

$$\vec{E}(\vec{z};2,1)=C^{(\xi)}\vec{\mathcal{E}}^{(\xi)}(\vec{s}_C,\omega_C)e^{i\vec{k}_C\cdot\vec{z}}+\frac{\vec{\mathcal{E}}^{(\xi)}(\vec{s}_D,\omega_C)\vec{\mathcal{E}}^{(\xi)}(\vec{s}_D,\omega_C)\cdot\vec{P}^{DB}(2,1)}{\epsilon_0(|2\vec{k}_O+\vec{k}_A|^2/|\vec{k}_C|^2-1)(|\vec{k}_O+\vec{k}_A|^2/|\vec{k}_B|^2-1)}e^{i(2\vec{k}_O+\vec{k}_A)\cdot\vec{z}}+\frac{\vec{\mathcal{E}}^{(\xi)}(\vec{s}_G,\omega_C)\vec{\mathcal{E}}^{(\xi)}(\vec{s}_G,\omega_C)\cdot\vec{P}^{DB}(2,1)}{\epsilon_0(|\vec{k}_O+\vec{k}_B|^2/|\vec{k}_C|^2-1)(|\vec{k}_O+\vec{k}_A|^2/|\vec{k}_B|^2-1)}e^{i(\vec{k}_O+\vec{k}_B)\cdot\vec{z}}. \quad (6.4)$$

Here ξ denotes the particular eigenvector which is phase matchable, $\vec{s}_C$ and $\vec{s}_D$ are defined by Eqs. (4.8) and (4.10), respectively, and $\vec{s}_G$ is defined by

$$\vec{s}_G=(\vec{k}_O+\vec{k}_B)/|\vec{k}_O+\vec{k}_B|. \quad (6.5)$$

By using the same input boundary condition as in Sec. III, the conditions

$$\vec{\mathfrak{D}}^{(\xi)}(\vec{s}_G,\omega_C)\cdot\vec{\mathcal{E}}^{(\xi)}(\vec{s}_C,\omega_C)\cong 1$$

and

$$\vec{\mathfrak{D}}^{(\xi)}(\vec{s}_G,\omega_C)\cdot\vec{\mathcal{E}}^{(\xi)}(\vec{s}_D,\omega_C)\cong 1,$$

which are good near phase matching, and an origin of coordinates in the input plane surface, we obtain by the procedure of Sec. III, three conditions

$$(\vec{k}_C-2\vec{k}_O-\vec{k}_A)_t=(\vec{k}_C-\vec{k}_O-\vec{k}_B)_t=0, \quad (6.6)$$

$$C^{(\xi)}=-\left[\frac{1}{|2\vec{k}_O+\vec{k}_A|^2/|\vec{k}_C|^2-1}-\frac{1}{|\vec{k}_O+\vec{k}_B|^2/|\vec{k}_C|^2-1}\right]\frac{\vec{\mathcal{E}}^{(\xi)}\cdot\vec{P}^{DB}(2,1)}{\epsilon_0(|\vec{k}_O+\vec{k}_A|^2/|\vec{k}_B|^2-1)}. \quad (6.7)$$

This leads to an output electrical field of

$$\vec{E}(\vec{z};2,1)=-\frac{2i\vec{\mathcal{E}}^{(\xi)}\vec{\mathcal{E}}^{(\xi)}\cdot\vec{P}^{DB}(2,1)k_C^2k_B^2}{\epsilon_0\Delta\vec{k}_{Bn}\cdot(\vec{k}_B+\vec{k}_O+\vec{k}_A)}\left(\frac{\sin(\frac{1}{2}\Delta\vec{k}_{Cn}\cdot\vec{z})e^{-(1/4)i\Delta\vec{k}_{Bn}\cdot\vec{z}}}{\Delta\vec{k}_{Cn}\cdot(\vec{k}_C+2\vec{k}_O+\vec{k}_A)}-\frac{\sin(\frac{1}{2}\Delta\vec{k}_{Dn}\cdot\vec{z})e^{(1/4)i\Delta\vec{k}_{Bn}\cdot\vec{z}}}{\Delta\vec{k}_{Dn}\cdot(\vec{k}_C+\vec{k}_O+\vec{k}_B}\right)\times\exp[\tfrac{1}{4}i(\vec{k}_C+\vec{k}_B+3\vec{k}_O+\vec{k}_A)\cdot\vec{z}], \quad (6.8)$$

where

$$\Delta\vec{k}_B=\vec{k}_B-\vec{k}_O-\vec{k}_A, \quad (6.9)$$

$$\Delta\vec{k}_C=\vec{k}_C-2\vec{k}_O-\vec{k}_A, \quad (6.10)$$

$$\Delta\vec{k}_D=\vec{k}_C-\vec{k}_O-\vec{k}_B. \quad (6.11)$$

The time-averaged Poynting vector then is

$$\vec{S}(\vec{z};2,1)=\frac{2c^2k_C^4k_B^4|\vec{\mathcal{E}}^{(\xi)}\circ\vec{P}^{DB}(2,1)|^2[(\vec{\mathcal{E}}^{(\xi)})^2\vec{k}_C\cdot-(\vec{k}_C\cdot\vec{\mathcal{E}}^{(\xi)})\vec{\mathcal{E}}^{(\xi)}]}{\epsilon_0\omega_C[\Delta\vec{k}_{Bn}\cdot(\vec{k}_B+\vec{k}_O+\vec{k}_A)]^2}\left(\frac{\sin^2(\frac{1}{2}\Delta\vec{k}_{Cn}\cdot\vec{z})}{[\Delta\vec{k}_{Cn}\cdot(\vec{k}_C+2\vec{k}_O+\vec{k}_A)]^2}+\frac{\sin^2(\frac{1}{2}\Delta\vec{k}_{Dn}\cdot\vec{z})}{[\Delta\vec{k}_{Dn}\cdot(\vec{k}_C+\vec{k}_O+\vec{k}_B)]^2}-\frac{2\sin(\frac{1}{2}\Delta\vec{k}_{Cn}\cdot\vec{z})\sin(\frac{1}{2}\Delta\vec{k}_{Dn}\cdot\vec{z})\cos(\frac{1}{2}\Delta\vec{k}_{Bn}\cdot\vec{z})}{\Delta\vec{k}_{Cn}\cdot(\vec{k}_C+2\vec{k}_O+\vec{k}_A)\Delta\vec{k}_{Dn}\cdot(\vec{k}_C+\vec{k}_O+\vec{k}_B)}\right). \quad (6.12)$$

This equation is valid under conditions similar to those stated for Eq. (4.16).

Double phase matching occurs when

$$\Delta\vec{k}_{Bn}=0, \quad (6.13)$$

$$\Delta\vec{k}_{Dn}=0, \quad (6.14)$$

which then also requires,

$$\Delta\vec{k}_{Cn}=0. \tag{6.15}$$

The input boundary condition in the plane-wave geometry assumed here has required the tangential components of these quantities to vanish already. At exact double phase matching Eq. (6.12) becomes

$$\vec{S}(l;2,1)=\frac{c^2k_C^4k_B^4l^4|\vec{\mathcal{E}}^{(\xi)}\cdot\vec{P}^{DB}(2,1)|^2}{128\epsilon_0\omega_Ck_{Cn}^2k_{Bn}^2}\times[(\vec{\mathcal{E}}^{(\xi)})^2\vec{k}_C-(\vec{k}_C\cdot\vec{\mathcal{E}}^{(\xi)})\vec{\mathcal{E}}^{(\xi)}], \tag{6.16}$$

where l once again is the distance into the crystal normal to the entrance surface. Note that doubly phase-matched AIOHG grows as l^4 compared to the usual l^2 characteristic of singly phase-matched nonlinear processes such as given in Eq. (4.18). Neglecting the ratio $[\vec{\mathcal{E}}^{(\xi)}\cdot\vec{P}^{DB}(2,1)/\vec{\mathcal{E}}^{(\xi)}\cdot\vec{\mathcal{P}}(2,1)]^2$ which can be either greater than or less than unity in different materials and geometries, and neglecting cosine projection factors of the normal components of various wave vectors, we see that the ratio of the intensity of the doubly phase-matched AIOH to the intensity of the singly phase-matched AIOH is $[\frac{1}{4}k_Bl]^2=6\times10^8$ for a crystal thickness of 1 cm, a wavelength in vacuum of 1 μ, and a refractive index of two. This spectacular enhancement factor offers hope that AIOHG under doubly phase-matched conditions may find practical uses.

By a similar development the time-averaged Poynting vector can be derived for AIOHG with double phase matching of the other two-step indirect contribution, second-harmonic generation followed by acousto-optic scattering (denoted by DH on the effective nonlinear polarization). We find that

$$\vec{S}(\vec{z};2,1)=\frac{2c^2k_C^4k_H^4|\vec{\mathcal{E}}^{(\xi)}\cdot\vec{P}^{DH}(2,1)|^2[(\vec{\mathcal{E}}^{(\xi)})^2\vec{k}_C-(\vec{k}_C\cdot\vec{\mathcal{E}}^{(\xi)})\vec{\mathcal{E}}^{(\xi)}]}{\epsilon_0\omega_C[\Delta\vec{k}_{Hn}\cdot(\vec{k}_H+2k_O)]^2}\left(\frac{\sin^2(\frac{1}{2}\Delta\vec{k}_{Cn}\cdot\vec{z})}{[\Delta k_{Cn}\cdot(\vec{k}_C+2\vec{k}_O+\vec{k}_A)]^2}+\frac{\sin^2(\frac{1}{2}\Delta\vec{k}_{En}\cdot\vec{z})}{[\Delta\vec{k}_{En}\cdot(\vec{k}_C+\vec{k}_H+\vec{k}_A)]^2}-\frac{2\sin(\frac{1}{2}\Delta\vec{k}_{Cn}\cdot\vec{z})\sin(\frac{1}{2}\Delta\vec{k}_{En}\cdot\vec{z})\cos(\frac{1}{2}\Delta\vec{k}_{Hn}\cdot\vec{z})}{\Delta\vec{k}_{Cn}\cdot(\vec{k}_C+2\vec{k}_O+\vec{k}_A)\Delta\vec{k}_{En}\cdot(\vec{k}_C+\vec{k}_H+\vec{k}_A)}\right), \tag{6.17}$$

where

$$\Delta\vec{k}_H=\vec{k}_H-2\vec{k}_O, \tag{6.18}$$

$$\Delta\vec{k}_E=\vec{k}_C-\vec{k}_H-\vec{k}_A, \tag{6.19}$$

$$P_i^{DH}(2,1)=\epsilon_0\left(\chi_{i\ j\ kl}^{\omega_C\omega_H\omega_A}-\frac{2d_{i\ j\ r}^{\omega_C\omega_H\omega_A}a_ra_se_{skl}^{\omega_A}}{\epsilon_0a_p\kappa_{pq}(\omega_A)a_q}\right)\times\mathcal{E}_j^{(\eta)}(\vec{s}_I,\omega_H)\mathcal{E}_m^{(\eta)}(\vec{s}_I,\omega_H)\times d_{m\ f\ g}^{\omega_H\omega_O\omega_O}E_f(1,0)E_g(1,0)u_{k,l}(0,1), \tag{6.20}$$

and $\vec{s}_I$ is given by Eq. (5.14).

Double phase matching in this case corresponds to

$$\Delta\vec{k}_{Hn}=0, \tag{6.21}$$

$$\Delta\vec{k}_{En}=0, \tag{6.22}$$

which then also requires

$$\Delta\vec{k}_{Cn}=0. \tag{6.23}$$

Here again the input boundary condition in the plane-wave geometry assumed has required the tangential components of these quantities to vanish already. At exact double phase matching Eq. (6.17) becomes

$$\vec{S}(l;2,1)=\frac{c^2k_C^4k_H^4l^4|\vec{\mathcal{E}}^{(\xi)}\cdot\vec{P}^{DH}(2,1)|^2}{128\epsilon_0\omega_Ck_{Cn}^2k_{Hn}^2}\times[(\vec{\mathcal{E}}^{(\xi)})^2\vec{k}_C-(\vec{k}_C\cdot\vec{\mathcal{E}}^{(\xi)})\vec{\mathcal{E}}^{(\xi)}], \tag{6.24}$$

which is analogous to Eq. (6.16).

APPENDIX A

We wish to make an order of magnitude estimate of the various terms in $\vec{F}^{\mu}(2,1)$ of Eq. (3.12) and in $\vec{I}(2,1)$ of Eq. (3.11) in order to determine which should be kept in the nonlinear polarization $\vec{P}(2,1)$ of Eq. (3.25). From Eqs. (3.8) and (3.9) with the nonlinear terms set to zero it is possible to make the following estimates:

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

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

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

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 that

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

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

$${}^{(3,1)}\overleftrightarrow{H}\approx{}^{(3,0)}\overleftrightarrow{H}, \tag{A6}$$

$${}^{(4,0)}\overleftrightarrow{H}\approx{}^{(3,0)}\overleftrightarrow{H}/a, \tag{A7}$$

where a is a typical primitive unit-cell dimension. From the lower-order problems, $(m,n)=(0,1)$, $(1,0)$, $(1,1)$, and $(2,0)$, the following estimates are possible:

$$y(0,1)\approx[\omega_A(m^0)^{1/2}/\omega_R(m^R)^{1/2}]u(0,1), \tag{A8}$$

$$u(1,0)\approx[v_A\omega_R(m^R)^{1/2}/v_O\omega_O(m^0)^{1/2}]y(1,0), \tag{A9}$$

$$E(1,0)\approx(\omega_R^2m^R/q^R)y(1,0), \tag{A10}$$

$$E(0,1)\approx[\omega_Aq^R(m^0)^{1/2}/\omega_R\epsilon_0(m^R)^{1/2}]u(0,1), \tag{A11}$$

$$B(1,0)\approx(\omega_R^2 m^R/q^R v_O)y(1,0)\,, \tag{A12}$$

$$B(0,1)\approx[v_A\omega_A q^R(m^0)^{1/2}/c^2\omega_R\epsilon_0(m^R)^{1/2}]u(0,1), \tag{A13}$$

$$y(1,1)\approx(\omega_A/v_A)y(1,0)u(0,1), \tag{A14}$$

$$E(1,1)\approx(\omega_A m^R\omega_R^2/v_A q^R)y(1,0)u(0,1), \tag{A15}$$

$$B(1,1)\approx(\omega_A m^R\omega_R^2/v_A v_O q^R)y(1,0)u(0,1), \tag{A16}$$

$$u(1,1)\approx[\omega_A\omega^R(m^R)^{1/2}/v_O\,\omega_O(m^0)^{1/2}]\times y(1,0)u(0,1), \tag{A17}$$

$$y(2,0)\approx(1/a)y(1,0)y(1,0), \tag{A18}$$

$$E(2,0)\approx(m^R\omega_R^2/q^R a)y(1,0)y(1,0), \tag{A19}$$

$$B(2,0)\approx(m^R\omega_R^2/q^R a v_O)y(1,0)y(1,0), \tag{A20}$$

$$u(2,0)\approx[v_A\omega_R(m^R)^{1/2}/v_O\,\omega_O a(m^0)^{1/2}]\times y(1,0)y(1,0)\,. \tag{A21}$$

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

$$\Delta\kappa\approx(q^R)^2/\epsilon_0 m^R\omega_R^2\,. \tag{A22}$$

Substitution of Eqs. (A1)–(A22) into each of the 40 terms of $\vec{F}(2,1)$ in Eq. (3.12) and into each of the 20 terms of $\vec{I}(2,1)$ in Eq. (3.11) leads to the conclusion that only 13 of those in $\vec{F}(2,1)$ and one of those in $\vec{I}(2,1)$ yield significant contributions to $P_i(2,1)$. These terms are given in Eq. (5.1).

APPENDIX B

Consider the effect of an infinitesimal body rotation on the second-order optical mixing tensor of the material body. An infinitesimal body rotation carries a body point at x_i to the new position x_i' given by

$$x_i'=x_i+u_i=x_i+\epsilon_{ijk}\,\delta\theta_j\,x_k\equiv-\frac{\partial\chi_i'}{\partial\chi_j}x_j\,, \tag{B1}$$

where

$$\frac{\partial\chi_i'}{\partial\chi_j}=\delta_{ij}+u_{i,j}=\delta_{ij}+\tilde{R}_{ij}\,, \tag{B2}$$

$$\tilde{R}_{ij}\equiv\tfrac{1}{2}(u_{i,j}-u_{j,i})=-\tilde{R}_{ji}=-\epsilon_{ijk}\,\delta\theta_k\,. \tag{B3}$$

Here $\vec{u}$ is the displacement vector and $\overleftrightarrow{R}$ is the mean rotation tensor that describes the counterclockwise infinitesimal body rotation through the angle $|\delta\vec{\theta}|$ about the direction of $\delta\vec{\theta}$. The second-order optical mixing tensor d_{ijk} must transform on each index i, j, k under the body rotation exactly as the vector x_i transforms under the same rotation:

$$\delta d_{ifg}=\frac{\partial x_i'}{\partial x_l}\frac{\partial x_f'}{\partial x_m}\frac{\partial x_g'}{\partial x_n}d_{lmn}-d_{ifg}\,, \tag{B4}$$

$$\delta d_{ifg}=d_{lfg}\tilde{R}_{il}+d_{img}\tilde{R}_{fm}+d_{ifn}\tilde{R}_{gn}\,, \tag{B5}$$

$$\delta d_{ifg}=\tfrac{1}{2}(d_{lfg}\,\delta_{ik}-d_{kfg}\,\delta_{il}+d_{ilg}\,\delta_{fk}-d_{ikg}\,\delta_{fl}+d_{ifl}\,\delta_{gk}-d_{ifk}\,\delta_{gl})u_{k,l}\,. \tag{B6}$$

If $\chi_{ifg[kl]}$ represents one of the two frequency components of the antisymmetric part of the AIOHG susceptibility which arise if the displacement gradient oscillates at a particular acoustic frequency, then

$$\delta d_{ifg}=2\chi_{ifg[kl]}u_{k,l}\,. \tag{B7}$$

Thus we obtain Eq. (5.45) of the text

$$\chi_{ifg[kl]}=\tfrac{1}{4}(d_{lfg}\,\delta_{ik}-d_{kfg}\,\delta_{il}+d_{ilg}\,\delta_{fk}-d_{ikg}\,\delta_{fl}+d_{ifl}\,\delta_{gk}-d_{ifk}\,\delta_{gl}). \tag{B8}$$

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