Polarization Dependence of Stimulated Rayleigh-Wing Scattering and the Optical-Frequency Kerr Effect*

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A theoretical study of the polarization dependence of stimulated Rayleigh-wing scattering and the optical-frequency Kerr effect has been undertaken. When the incident light is circularly polarized, a striking difference in gain is found for the co- and counter-rotating senses of polarization of the scattered light—the former being suppressed and the latter being exceptionally favored. The analysis begins with a model for the Kerr effect, which involves the alignment of anisotropic molecules in an electric field, but the results can be immediately generalized to any light scattering process of tensor symmetry. The nonlinear problem of the propagation of an intense elliptically polarized light wave in a Kerr-active medium is shown to have a solution in which the vibrational ellipse undergoes self-precession and self-retardation. The stimulated scatterings or instabilities of such a self-precessing and self-retarding light wave are obtained for the backward and forward directions. Birefringence, optical activity, and linear and circular dichroisms are some of the phenomena which result, but the forward direction yields substantially different results from the backward direction because of Stokes–anti-Stokes coupling.

I. INTRODUCTION

It has been observed by Foltz, Cho, Rank, and Wiggins[1] that stimulated Rayleigh-wing scattering behaves quite differently when the incident light is linearly or circularly polarized. They found that excitation with linearly polarized light results in a *diffuse* Stokes wing of the same polarization, whereas circularly polarized light excites a *sharp* Stokes line with an opposite sense of circular polarization. We wish to point out that these phenomena are intimately connected with the existence or absence of an anti-Stokes channel in the forward direction due to the tensor nature of the scattering process.

The case of parallel linear polarizations for incident and scattered light has already been treated by Chiao, Kelley, and Garmire.[2] As a result of Stokes–anti-Stokes coupling, scattering in the near-forward direction was shown to be dominated by the degenerate light-by-light interaction involving no shift in scattered light frequency, up to an angle of

$$(\sqrt{\tfrac{3}{2}})\theta_{\rm opt}=(3\epsilon_2|\mathcal{E}_0|^2/4\epsilon_0)^{1/2},$$

where $\epsilon_2|\mathcal{E}_0|^2$ is the time-averaged dielectric change arising from the intensity of the light, and ϵ_0 is the linear dielectric constant of the scattering medium. Beyond that angle, however, the maximum gain occurs for light of a shifted frequency

$$\Omega=\frac{1}{\tau}\left(\frac{2\theta^2-3\theta_{\rm opt}^2}{2\theta^2-\theta_{\rm opt}^2}\right)^{1/2},\qquad(1)$$

which starts at zero and gradually approaches τ^{-1} at large angles (τ is the orientation relaxation time). Therefore, the collection of light from angles near the forward direction, as was actually done under the experimental conditions of Foltz *et al.*, would have given rise to the observed diffuse wing.[3]

The observations with circularly polarized light can be explained heuristically on the following basis: We assume that the molecules are cigar shaped and that the field-dependent index change arises from the average alignment of these molecules parallel to the time-averaged field direction.[4,5] If the scattered light has the same sense of circular polarization as that of the incident light, the molecules, being slow in response, tend to align themselves randomly in the plane swept out by the rapidly rotating electric fields of incident and scattered light. The total intensity goes through a maximum periodically with a frequency equal to the beat frequency between the incident and scattered light. The resulting periodic modulation of the molecular alignment gives rise to a symmetric production of Stokes and anti-Stokes, implying that the gain in the forward direction is zero, according to arguments similar to those in Ref. 2.

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[1] N. D. Foltz, C. W. Cho, D. H. Rank, and T. A. Wiggins, Phys. Rev. 165, 396 (1968).
[2] R. Y. Chiao, P. L. Kelley, and E. Garmire, Phys. Rev. Letters 17, 1281 (1966).
[3] See also G. I. Zaitzev, Yu. I. Kyzylasov, V. S. Starunov, I. L. Fabelinskii, Zh. Eskerim. i Teor. Fiz. Pis'ma v Redaktsiyu 6, 505 (1967) [English transl.: Soviet Phys.—JETP Letters 6, 35 (1967)].
[4] J. Frenkel, *Kinetic Theory of Liquids* (Dover Publications, Inc., New York, 1955), p. 282.
[5] N. Bloembergen and P. Lallemand, Phys. Rev. Letters 16, 81 (1966).

However, if the scattered light has the opposite sense of polarization relative to that of the incident light, the situation is quite different. In this case, if we describe the incident and scattered fields, respectively, as

$$\mathbf{E}_0 = \mathcal{E}_0(\hat{\imath}\cos\omega_0 t + \hat{\jmath}\sin\omega_0 t),$$
$$\mathbf{E}_1 = \mathcal{E}_1(\hat{\imath}\cos\omega_1 t - \hat{\jmath}\sin\omega_1 t), \quad (2)$$

where ω_0 is the incident frequency, ω_1 is the scattered Stokes frequency ($\omega_0 > \omega_1$), and $\hat{\imath}$, $\hat{\jmath}$, and $\hat{k}$ are unit vectors in the x, y, and z directions, respectively. The sum of these fields is

$$\begin{aligned}\mathbf{E}_{\text{tot}} = \mathbf{E}_0 + \mathbf{E}_1 = (\mathcal{E}_0 + \mathcal{E}_1)\cos\tfrac{1}{2}(\omega_0+\omega_1)t \\ \times[\hat{\imath}\cos\tfrac{1}{2}(\omega_0-\omega_1)t + \hat{\jmath}\sin\tfrac{1}{2}(\omega_0-\omega_1)t] \\ +(\mathcal{E}_0-\mathcal{E}_1)\sin\tfrac{1}{2}(\omega_0+\omega_1)t \\ \times[-\hat{\imath}\sin\tfrac{1}{2}(\omega_0-\omega_1)t + \hat{\jmath}\cos\tfrac{1}{2}(\omega_0-\omega_1)t]. \quad (3)\end{aligned}$$

These terms describe slowly precessing linearly polarized fields, whose directions of polarization precesses at half of the beat frequency $\Omega = \omega_0 - \omega_1$ and in the same sense as that of the light at the higher of the two frequencies ω_0 and ω_1. Provided $\Omega \lesssim \tau^{-1}$, where τ is the relaxation time of the molecular alignment, the molecular axes tend to follow this slow precession and, thus, rotate along with the time-averaged field directions, except for some phase lag. The induced dipole moments which produce scattering can be gotten by projecting a rapidly rotating $\mathbf{E}_0$ on the slowly precessing molecular axes. This gives rise to linearly polarized but precessing dipole moments, which when resolved into counter-rotating components will yield only a Stokes component in the same sense as $\mathbf{E}_1$, without any anti-Stokes component. Moreover, if ω_1 were an anti-Stokes frequency $\omega_1 > \omega_0$, then the sense of precession would be the reverse of when ω_1 is a Stokes frequency $\omega_1 < \omega_0$. Hence, the molecular motion involved in anti-Stokes scattering is orthogonal to that involved for Stokes scattering. This decoupling of the anti-Stokes channel from the Stokes channel implies an unsuppressed gain in forward scattering. A more explicit demonstration of this decoupling starts with the intensity-dependent susceptibility tensor associated with the molecular alignment

$$\begin{aligned}\Delta\chi \sim \mathbf{E}_{\text{tot}}\mathbf{E}_{\text{tot}} \sim \mathbf{E}_0\mathbf{E}_0 + (\mathbf{E}_0\mathbf{E}_1 + \mathbf{E}_1\mathbf{E}_0) \sim \tfrac{1}{2}\mathcal{E}_0^2(\hat{\imath}\hat{\imath}+\hat{\jmath}\hat{\jmath}) \\ +\mathcal{E}_0\mathcal{E}_1[(\hat{\imath}\hat{\imath}-\hat{\jmath}\hat{\jmath})\cos\Omega t + (\hat{\imath}\hat{\jmath}+\hat{\jmath}\hat{\imath})\sin\Omega t], \quad (4)\end{aligned}$$

where $\Omega = \omega_0 - \omega_1$. We neglect terms at second-harmonic frequencies and terms of the order $\mathcal{E}_1^2$, since we shall assume henceforth that $\mathcal{E}_1 \ll \mathcal{E}_0$. The induced dipole moment per unit volume is

$$\begin{aligned}\Delta\mathbf{P} = \Delta\chi\cdot(\mathbf{E}_0+\mathbf{E}_1) \sim \tfrac{1}{2}\mathcal{E}_0^2\mathcal{E}_1(\hat{\imath}\cos\omega_1 t - \hat{\jmath}\sin\omega_1 t) \\ +\mathcal{E}_0^2\mathcal{E}_1[\hat{\imath}\tfrac{1}{2}(\cos\omega_1 t+\cos\omega_2 t) - \hat{\jmath}\tfrac{1}{2}(\sin\omega_1 t+\sin\omega_2 t) \\ +\hat{\jmath}\tfrac{1}{2}(-\sin\omega_1 t+\sin\omega_2 t) + \hat{\imath}\tfrac{1}{2}(\cos\omega_1 t-\cos\omega_2 t)], \quad (5)\end{aligned}$$

where $\omega_2 \equiv \omega_0 + \Omega$ is the anti-Stokes frequency. Note, however, that all terms at ω_2 cancel and we are left with only a Stokes component

$$\Delta\mathbf{P} \sim \tfrac{3}{2}\mathcal{E}_0^2\mathcal{E}_1(\hat{\imath}\cos\omega_1 t - \hat{\jmath}\sin\omega_1 t). \quad (6)$$

These dipole moments are rotating at the same frequency and in the same sense as the scattered field $\mathbf{E}_1$, and with some phase lag they can feed energy back into $\mathbf{E}_1$ and amplify it without hindrance from anti-Stokes production. This implies that the gain curve peaks at[5] $\Omega = \tau^{-1}$ even in the forward direction, and explains the presence of the sharp Stokes line of the opposite sense of circular polarization in the observations of Foltz *et al.* An immediate consequence of such a scattering process is that an appreciable amount of angular momentum is transferred to the liquid, since each scattered photon imparts $2\hbar$ to the molecular rotation, which, because of the collisional torque exerted on the surrounding fluid, must ultimately manifest itself in a slight general rotation or vorticity of the liquid in the same sense as that of the laser polarization.[6]

Herman[7] has suggested another explanation of the polarization dependence of Rayleigh-wing scattering. His theory examines the behavior of the saturation of molecular alignment for different polarizations and the consequent influence on the gain. We believe that these effects are of higher order and are not as powerful as the ones which are discussed in this paper.

II. MOLECULAR-ORIENTATION KERR EFFECT

Let us consider the field-induced susceptibility change (or Kerr effect) associated with molecular alignment in more detail. We shall restrict our attention to the term in the Kerr effect which is quadratic in the electric field, since this is the lowest-order term responsible for stimulated light scattering. Furthermore, let us assume that the alignment has a single relaxation time τ (one-third the Debye time)[5] so that the susceptibility change $\Delta\chi_{ik}$ satisfies the dynamical equation

$$\frac{\partial\Delta\chi_{ik}}{\partial t} + \frac{\Delta\chi_{ik}}{\tau} = K(E_iE_k + \sigma\delta_{ik}E_jE_j). \quad (7)$$

Here, E_i is the total electric field, which is the vector sum of the incident and scattered fields (Einstein's summation convention will be used henceforth). The driving term is the most general possible second-rank tensor which can be formed from terms quadratic in the field in an isotropic medium. Specific model calculations to be carried out below will determine K and σ. These quadratic terms contain sum and difference frequencies, but Eq. (7) filters out the sum frequencies and allows the susceptibility to respond only to those difference frequencies of the order of τ^{-1}.

Now for our model, let us start with a single cigar-shaped nonpolar symmetric-top molecule oriented, as

[6] See Ref. 4, p. 286; V. N. Zwetkov, Acta Physicochim. URSS **10**, 555 (1939).
[7] R. M. Herman, Phys. Rev. **164**, 200 (1967).

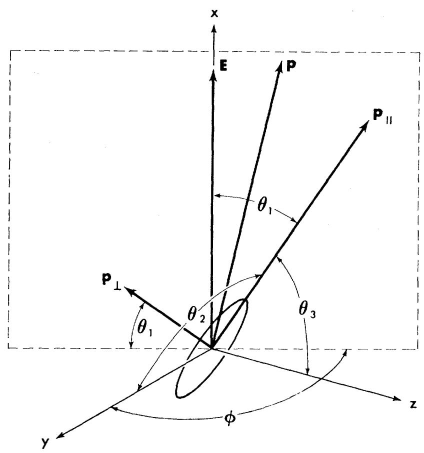

FIG. 1. Anisotropic molecule in an electric field.

shown in Fig. 1, with respect to a static electric field which is pointed along the x axis. The induced dipole moments along the symmetry axis of the molecule and perpendicular to it (in the plane determined by $\mathbf{E}$ and the symmetry axis) are, respectively,

$$\begin{aligned} p_{\parallel} &= \alpha_{\parallel} E \cos\theta_1, \\ p_{\perp} &= \alpha_{\perp} E \sin\theta_1, \end{aligned} \tag{8}$$

where $\alpha_{\parallel}$ is the polarizability of the molecule along the symmetry axis, and $\alpha_{\perp}$ is the polarizability along the two other axes. Projecting them along the three space-fixed axes (see Fig. 1), we get

$$\begin{aligned} p_x &= \alpha_{\parallel} E \cos^2\theta_1 + \alpha_{\perp} E \sin^2\theta_1 \\ &= (\alpha_{\parallel} - \alpha_{\perp}) E \cos^2\theta_1 + \alpha_{\perp} E, \\ p_y &= \alpha_{\parallel} E \cos\theta_1 \cos\theta_2 - \alpha_{\perp} E \sin\theta_1 \cos\theta_1 \cos\phi \\ &= (\alpha_{\parallel} - \alpha_{\perp}) E \cos\theta_1 \cos\theta_2, \\ p_z &= \alpha_{\parallel} E \cos\theta_1 \cos\theta_3 - \alpha_{\perp} E \sin\theta_1 \cos\theta_1 \sin\phi \\ &= (\alpha_{\parallel} - \alpha_{\perp}) E \cos\theta_1 \cos\theta_3. \end{aligned} \tag{9}$$

The energy of alignment for this molecule is

$$W = \tfrac{1}{2}\alpha_{\perp} E^2 - \tfrac{1}{2}(\alpha_{\parallel} - \alpha_{\perp}) E^2 \cos^2\theta_1 = W_0 + W_1. \tag{10}$$

Taking a dilute ensemble of these molecules at a temperature T, the susceptibility tensor of the system is

$$\begin{aligned} \chi_{ik} &= (\tfrac{1}{3}\alpha_{\parallel} + \tfrac{2}{3}\alpha_{\perp}) N \delta_{ik} + N(\alpha_{\parallel} - \alpha_{\perp}) s_{ik} \\ &= \chi_0 \delta_{ik} + \Delta\chi_{ik}, \end{aligned} \tag{11}$$

where N is the number of molecules per unit volume, and s_{ik} is the anisotropy tensor

$$s_{ik} = \langle \cos\theta_i \cos\theta_k \rangle - \tfrac{1}{3}\delta_{ik}. \tag{12}$$

When $E=0$, clearly $s_{ik}=0$, so that $\chi_0 = N(\frac{1}{3}\alpha_{\parallel} + \frac{2}{3}\alpha_{\perp})$ is the zero-field or linear optical susceptibility. The second term of s_{ik} makes it traceless, since $\sum_i \cos^2\theta_i = 1$. Hence, $\sum_i \Delta\chi_{ii} = 0$, implying that $\sigma = -\frac{1}{3}$ in Eq. (7). The brackets in (12) denote statistical mechanical averages:

$$\begin{aligned} \langle \cos\theta_i \cos\theta_k \rangle &= \int e^{-W_1/kT} \mu_i \mu_k d\Omega \Big/ \int e^{-W_1/kT} d\Omega \\ &= \left(\frac{\partial^2}{\partial E_i \partial E_k} \ln \int e^{-W_1/kT} d\Omega \right) \frac{kT}{\alpha_{\parallel} - \alpha_{\perp}}, \end{aligned} \tag{13}$$

where $\mu_i = \cos\theta_i$, and $W_1 = -\frac{1}{2}(\alpha_{\parallel} - \alpha_{\perp})(E_j \mu_j)^2$. In the particular case when the field $\mathbf{E}$ is pointing along the x axis, $\langle \cos\theta_i \cos\theta_k \rangle$ is diagonal, and expanding the Boltzmann factor into a series, we get

$$\begin{aligned} \langle \cos^2\theta_1 \rangle &\cong \frac{1}{3} + \frac{2}{45} \frac{\alpha_{\parallel} - \alpha_{\perp}}{kT} E^2, \\ \langle \cos^2\theta_2 \rangle = \langle \cos^2\theta_3 \rangle &\cong \frac{1}{3} - \frac{1}{45} \frac{\alpha_{\parallel} - \alpha_{\perp}}{kT} E^2. \end{aligned} \tag{14}$$

Evaluating (7) for this case and for a static field, we get $\Delta\chi_{11} = \frac{2}{3} K_T E^2$ and $\Delta\chi_{22} = \Delta\chi_{33} = -\frac{1}{3} K_T E^2$, so that identifying with (14) we get

$$K_T = \frac{3}{45} N \frac{(\alpha_{\parallel} - \alpha_{\perp})^2}{kT}. \tag{15}$$

To include the local-field correction, one multiplies the right-hand side of (15) by $[\frac{1}{3}(\epsilon_0 + 2)]^4$.[8] The third-order nonlinear susceptibility $\chi^{(3)}$, is $\frac{2}{3} K_T$, and the nonlinear index of refraction coefficient n_2 is $\frac{4}{3}\pi(K_T/n_0)$.

III. SELF-PRECESSION AND SELF-RETARDATION OF INCIDENT WAVE

We wish to solve the general problem of stimulated Rayleigh-wing scattering when the incident laser light is elliptically polarized with an eccentricity ϵ. Before going on to the scattering problem, however, we must first treat the problem of the propagation of the intense elliptically polarized incident light through a Kerr-active medium in the absence of scattering. As we shall see presently, this nonlinear propagation problem can be solved in closed form, and solution implies that the vibrational ellipse undergoes a self-precession of its major axis without any change of eccentricity,[9] and that the wave experiences a self-retardation. Let us assume that the incident light is propagating along the z axis and is described by the transverse wave[10]

$$E_{0j} = \mathcal{E}_{0j}(z) e^{i(k_0 z - \omega_0 t)}, \quad j = (x, y), \tag{16}$$

where $\mathcal{E}_{0j}(z)$ is assumed to be a slowly varying amplitude with $|\mathcal{E}_0|^{-1}(d|\mathcal{E}_0|/dz) \ll k_0$. The light enters the

[8] One could also include the reactive field of Onsager, in which case see Ref. 4, p. 274.

[9] P. D. Maker, R. W. Terhune, and C. M. Savage, Phys. Rev. Letters 12, 507 (1964). Their A is $\frac{1}{12} K_T$ and their B is $\frac{1}{6} K_T$.

[10] We shall use complex notation throughout this paper. One useful rule is that if $[x]$ is the complex representation of a real quantity x then $[xy] = \frac{1}{2}([x][y] + [x][y]^*)$.

medium at $z=0$ with

$$\begin{pmatrix}\mathcal{E}_{0x}(0)\\ \mathcal{E}_{0y}(0)\end{pmatrix}=\mathcal{E}_{00}\begin{pmatrix}1\\ i\epsilon\end{pmatrix}. \tag{17}$$

This describes an initial state of elliptical polarization with the principal axes along the x and y directions. The nonlinear wave equation reduces to

$$e^{-i(k_0z-\omega_0t)}\left(\frac{\partial^2}{\partial z^2}E_{0i}-\frac{n_0^2}{c^2}\frac{\partial^2}{\partial t^2}E_{0i}\right)$$
$$\cong 2ik_0\frac{d\mathcal{E}_{0i}}{dz}=-4\pi\frac{\omega_0^2}{c^2}\chi_{ik}^0\mathcal{E}_{0k}, \tag{18}$$

where, because the molecules cannot respond to the second harmonic of the optical frequency,

$$\chi_{ik}^0=\tfrac{1}{4}K\tau(\mathcal{E}_{0i}^*\mathcal{E}_{0k}-\tfrac{1}{3}\delta_{ik}\mathcal{E}_{oj}^*\mathcal{E}_{0j}+\text{c.c.}), \tag{19}$$

which is the susceptibility change deduced from (7). Let us define a propagation tensor

$$\Gamma_{ik}=-2\pi(k_0/n_0^2)\chi_{ik}^0, \tag{20}$$

where $k_0=n_0\omega_0/c$. Since Γ_{ik} is a tensor, in another coordinate system (x',y') rotated at an angle ϕ with respect to the original system (x,y), its components Γ_{ik}' are given by

$$\Gamma_{ik}=R_{il}R_{km}\Gamma_{lm}'=R_{il}\Gamma_{lm}'R^{-1}{}_{mk}, \tag{21}$$

where R_{ik} is the orthogonal matrix

$$(R_{ik})=\begin{pmatrix}\cos\phi & -\sin\phi\\ \sin\phi & \cos\phi\end{pmatrix}, \tag{22}$$

transforming a vector from the primed to the unprimed system. For complete generality, let us allow $\phi(z)$ to be an arbitrary function of z, except for the boundary condition $\phi(z=0)=0$. It is convenient to transform (18) into this rotating coordinate system, because our ansatz will describe a uniform precession of the vibrational ellipse. Substituting (21) into (18) we obtain

$$id\mathcal{E}_{0i}/dz=R_{il}\Gamma_{lm}'R^{-1}{}_{mk}\mathcal{E}_{0k}. \tag{23}$$

Multiplying on both sides by $R^{-1}{}_{ji}$ and using the relation

$$dR^{-1}{}_{ji}/dz=\epsilon_{jk}R^{-1}{}_{ki}(d\phi/dz), \tag{24}$$

where

$$(\epsilon_{jk})=\begin{pmatrix}0 & 1\\ -1 & 0\end{pmatrix}$$

is the unit antisymmetric tensor, we deduce that

$$i\left(\frac{d\mathcal{E}_{0j}'}{dz}-\frac{d\phi}{dz}\epsilon_{jk}\mathcal{E}_{0k}'\right)=\Gamma_{jm}'\mathcal{E}_{0m}'$$
$$=i\left(\frac{d\mathcal{E}_{0j}'}{dz}+(\mathbf{\Gamma}\times\boldsymbol{\mathcal{E}}_0')_j\right), \tag{25}$$

where $\mathcal{E}_{0j}'=R^{-1}{}_{jk}\mathcal{E}_{0k}$ is the electric field as viewed in the rotating frame, and where $\mathbf{\Gamma}=\hat{k}d\phi/dz$ is the instantaneous rate of precession of this frame.

Our ansatz can now be stated as follows:

$$\begin{pmatrix}\mathcal{E}_{0x'}(z)\\ \mathcal{E}_{0y'}(z)\end{pmatrix}=\mathcal{E}_{00}\begin{pmatrix}1\\ i\epsilon\end{pmatrix}e^{i\kappa z}. \tag{26}$$

It states that the vibrational ellipse as viewed in the rotating frame is unchanged from that at $z=0$, except for a phase factor $e^{i\kappa z}$ arising from some nonlinear self-retardation. Evaluating Γ_{jm}' from (26) and the primed version of (19),

$$(\Gamma_{jm}')=-\tfrac{1}{2}g_0\begin{pmatrix}2-\epsilon^2 & 0\\ 0 & 2\epsilon^2-1\end{pmatrix}, \tag{27}$$

where

$$\tfrac{1}{2}g_0=\tfrac{1}{3}\pi\frac{k_0}{n_0^2}K\tau|\mathcal{E}_{00}|^2=k_0\frac{n_2|\mathcal{E}_{00}|^2}{4n_0}=\frac{2\pi}{\lambda_0}\frac{\langle\Delta n\rangle}{n_0}\times\frac{1}{2}, \tag{28}$$

which is also equal to half the maximum stimulated Rayleigh-wing gain for linear polarization.[2] Note that Γ_{jm}' is independent of z and is equal to $\Gamma_{jm}(z=0)$. Substitution of (26) and (27) into (25) yields the self-consistency condition (or nonlinear eigenvalue equation)[11]

$$\kappa\begin{pmatrix}1\\ i\epsilon\end{pmatrix}=\tfrac{1}{2}g_0\begin{pmatrix}2-\epsilon^2 & -i(2/g_0)d\phi/dz\\ i(2/g_0)d\phi/dz & 2\epsilon^2-1\end{pmatrix}\begin{pmatrix}1\\ i\epsilon\end{pmatrix}, \tag{29}$$

the solution of which is

$$\kappa=g_0(1+\epsilon^2)=k_0\times\frac{n_2}{2n_0}[|\mathcal{E}_{0x}(0)|^2+|\mathcal{E}_{0y}(0)|^2]$$
$$=\tfrac{2}{3}\pi\frac{k_0}{n_0^2}K\tau\mathcal{E}_{0j}^*\mathcal{E}_{0j}, \tag{30a}$$

$$\Gamma=\tfrac{3}{2}g_0\epsilon=k_0\times\frac{3}{4}\frac{n_2}{n_0}|\mathcal{E}_{0x}(0)||\mathcal{E}_{oy}(0)|$$
$$=\tfrac{1}{2}\pi\frac{k_0}{n_0^2}K\tau|\mathrm{Im}\,\epsilon_{3jk}\mathcal{E}_{0j}^*\mathcal{E}_{0k}|. \tag{30b}$$

These quantities represent the retardation and the rate of precession (both of which are constant) of the incident light and its vibrational ellipse, respectively. The sense of the precession is the same as that of the polarization of the light and the retardation produces an actual slowing down of the wave provided $n_2>0$, as is the case for the molecular-orientation Kerr effect. Furthermore, these quantities are to be considered small compared with k_0, since we have assumed $n_2|\mathcal{E}_0|^2\ll n_0$ in order to restrict our attention to the lowest-order (quadratic) Kerr effect.

[11] Cf. R. Y. Chiao, E. Garmire, and C. H. Townes, Phys. Rev. Letters 13, 479 (1964).

It should be remarked that photon angular momentum conservation forbids any change in the eccentricity of the vibrational ellipse, since the medium is isotropic and the propagation involves no inelastic scattering. These considerations are still valid for the case of nonlinear propagation.[12]

IV. BACKWARD SCATTERING

Let us proceed to treat stimulated Rayleigh-wing scattering in the backward direction first, since this case does not involve an anti-Stokes channel, due to momentum-conservation restrictions.[2] The intense incident laser wave was shown in Sec. III to propagate as

$$\mathbf{E}_0=\mathcal{E}_{00}e^{i(k_0z+\kappa z-\omega_0t)}\begin{pmatrix}\cos\Gamma z & -\sin\Gamma z\\ \sin\Gamma z & \cos\Gamma z\end{pmatrix}\begin{pmatrix}1\\ i\epsilon\end{pmatrix}. \quad (31)$$

Let us assume that the back scattered wave has the form

$$E_1=\boldsymbol{\varepsilon}_1(z)e^{i(-k_1z-\kappa z-\omega_1t)}, \quad (32)$$

where $\boldsymbol{\varepsilon}_1(z)$ is slowly varying in the sense

$$\frac{1}{|\mathcal{E}_1|}\frac{d|\mathcal{E}_1|}{dz}\ll k_1,$$

and $|\mathcal{E}_1|\ll|\mathcal{E}_0|$, i.e., the scattered wave is much weaker than the incident wave. The sum of these fields will produce a susceptibility change obeying Eq. (7). Neglecting sum frequencies and terms of the order $\mathcal{E}_1^2$, we obtain the solution (in complex form)

$$\begin{aligned}\Delta\chi_{ik}=\tfrac{1}{2}K\tau[(E_{0i}^*E_{0k}-\tfrac{1}{3}\delta_{ik}E_{0j}^*E_{0j})\\ +(1+i\Omega\tau)^{-1}(E_{0i}^*E_{1k}+E_{0k}^*E_{1i}-\tfrac{2}{3}\delta_{ik}E_{0j}^*E_{1j})],\end{aligned} \quad (33)$$

where $\Omega=\omega_0-\omega_1$. The first two terms are constant in time and the last three terms vary as $e^{+i\Omega t}$. It is convenient to define a molecular response function as

$$D=(1+i\Omega\tau)^{-1}. \quad (34)$$

The induced nonlinear polarization, again neglecting terms of the order $\mathcal{E}_1^2$ and neglecting anti-Stokes terms varying with frequency $\omega_0+\Omega$, is

$$\begin{aligned}\Delta P_i&=\tfrac{1}{2}(\Delta\chi_{ik}E_k+\Delta\chi_{ik}^*E_k)\\ &=\chi_{ik}^0(E_{0k}+E_{1k})+\chi_{ik}^{\mathrm{I}}E_{0k},\end{aligned} \quad (35)$$

where

$$\begin{aligned}\chi_{ik}^0&=\tfrac{1}{4}K\tau(E_{0i}^*E_{0k}+E_{0i}E_{0k}^*-\tfrac{2}{3}\delta_{ik}E_{0j}^*E_{0j}),\\ \chi_{ik}^{\mathrm{I}}&=D\cdot\tfrac{1}{4}K\tau(E_{0i}^*E_{1k}+E_{0k}^*E_{1i}-\tfrac{2}{3}\delta_{ik}E_{0j}^*E_{1j}).\end{aligned} \quad (36)$$

Substituting into the wave equation and remembering that $\mathcal{E}_1e^{-i\kappa z}$ is slowly varying, we get

$$-2ik_1\left(\frac{1}{\mathcal{E}_{1i}}\frac{d\mathcal{E}_{1i}}{dz}-i\kappa\right)E_{1i}=-4\pi\frac{\omega_1^2}{c^2}(\chi_{ik}^0E_{1k}+\chi_{ik}^{\mathrm{I}}E_{0k}). \quad (37)$$

Let us assume $\omega_1\cong\omega_0$ and $k_1\cong k_0$, since $\Omega\ll\omega_0$. Then the last term on the left-hand side can be subtracted from the first term of the right-hand side of this equation by introducing

$$\chi_{ik}^{10}=\tfrac{1}{4}K\tau(E_{0i}^*E_{0k}+E_{0i}E_{0k}^*-2\delta_{ik}E_{0j}^*E_{0j}) \quad (38)$$

in place of χ_{ik}^0. This eliminates the contribution to "strong-wave retardation[2]" due to the isotropic index change caused by the strong incident wave from the propagation of the weak scattered wave. What is left describes birefringent effects of the strong wave upon the weak wave. Hence we shall refer to χ_{ik}^{10} as the "strong-wave susceptibility tensor."

To express (37) as a differential equation for the scattered field E_{1k} we need to rewrite the last term on the right-hand side as

$$\chi_{ik}^{\mathrm{I}}E_{0k}=\chi_{ik}^{11}E_{1k}, \quad (39)$$

where, by juggling the order of the tensor product of the fields, we determine that

$$\chi_{ik}^{11}=D\times\tfrac{1}{4}K\tau(E_{0i}^*E_{0k}-\tfrac{2}{3}E_{0k}^*E_{0i}+\delta_{ik}E_{0j}^*E_{0j}), \quad (40)$$

which is a known quantity determinable from (31). The real part of this susceptibility causes "weak-wave retardation,[2]" and the imaginary part causes stimulated light scattering by reacting the scattered field back upon itself via the molecular alignment. Hence, we shall refer to χ_{ik}^{11} as the "self-coupling susceptibility tensor." The wave equation becomes

$$i\frac{d\mathcal{E}_{1i}}{dz}=2\pi\frac{k_0}{n_0^2}(\chi_{ik}^{10}+\chi_{ik}^{11})\mathcal{E}_{1k}. \quad (41)$$

Note that χ_{ik}^{10} and χ_{ik}^{11} are slowly varying functions of z due to the precession of (31), and hence this differential equation in its present form is not solvable by the conventional linear-eigenvalue method. However, let us introduce the quantities $\chi_{jl}^{10\prime}$ and $\chi_{jl}^{11\prime}$ through

$$\begin{aligned}\chi_{ik}^{10}&=R_{ij}R_{kl}\chi_{jl}^{10\prime},\\ \chi_{ik}^{11}&=R_{ij}R_{kl}\chi_{jl}^{11\prime},\end{aligned} \quad (42)$$

where

$$(R_{ij})=\begin{pmatrix}\cos\Gamma z & -\sin\Gamma z\\ \sin\Gamma z & \cos\Gamma z\end{pmatrix}, \quad (43)$$

so that $\chi_{jl}^{10\prime}$ and $\chi_{jl}^{11\prime}$ are the strong-wave and self-coupling susceptibilities, respectively, as viewed in the rotating frame or, equivalently, these tensors evaluated at $z=0$

$$\begin{aligned}\chi_{ik}^{10\prime}&=\tfrac{1}{4}K\tau[\mathcal{E}_{0i}^*(0)\mathcal{E}_{0k}(0)\\ &\quad+\mathcal{E}_{0k}^*(0)\mathcal{E}_{0i}(0)-2\delta_{ik}\mathcal{E}_{0j}^*(0)\mathcal{E}_{0j}(0)],\\ \chi_{ik}^{11\prime}&=D\times\tfrac{1}{4}K\tau[\mathcal{E}_{0i}^*(0)\mathcal{E}_{0k}(0)\\ &\quad-\tfrac{2}{3}\mathcal{E}_{0k}^*(0)\mathcal{E}_{0i}(0)+\delta_{ik}\mathcal{E}_{0j}^*(0)\mathcal{E}_{0j}(0)],\end{aligned} \quad (44)$$

[12] This argument can be generalized to any order of the Kerr effect, as can be seen by writing (18) in covariant form.

or evaluated using (17)

$$(\chi_{ik}^{10'})=\tfrac{1}{4}K\tau|\mathcal{E}_{00}|^2\begin{pmatrix}-2\epsilon^2 & 0\\ 0 & -2\end{pmatrix},$$
$$(\chi_{ik}^{11'})=D\times\tfrac{1}{4}K\tau|\mathcal{E}_{00}|^2\begin{pmatrix}\frac{4}{3}+\epsilon^2 & \frac{5}{3}i\epsilon\\ -\frac{5}{3}i\epsilon & \frac{4}{3}\epsilon^2+1\end{pmatrix}. \qquad (45)$$

Let us introduce

$$\mathcal{E}_{1i}'=R^{-1}{}_{ik}\mathcal{E}_{1k}. \qquad (46)$$

Then (41) becomes, in the rotating frame,

$$i\left(\frac{d}{dz}\mathcal{E}_{1i}'-\Gamma\epsilon_{ik}\mathcal{E}_{1k}'\right)=\gamma_{ik}'\mathcal{E}_{1k}', \qquad (47)$$

where γ_{ik}' is z-independent and given by

$$\gamma_{ik}'=2\pi(k_0/n_0^2)(\chi_{ik}^{10'}+\chi_{ik}^{11'}). \qquad (48)$$

We now introduce our ansatz

$$\begin{pmatrix}\mathcal{E}_{1x}'(z)\\ \mathcal{E}_{1y}'(z)\end{pmatrix}=\mathcal{E}_{10}\begin{pmatrix}1\\ i\delta\end{pmatrix}e^{-i\gamma z}, \qquad (49a)$$

or

$$\begin{pmatrix}E_{1x}(z)\\ E_{1y}(z)\end{pmatrix}=\begin{pmatrix}\cos\Gamma z & -\sin\Gamma z\\ \sin\Gamma z & \cos\Gamma z\end{pmatrix}\begin{pmatrix}1\\ i\delta\end{pmatrix}\times\mathcal{E}_{10}e^{i[(-k_1-\kappa-\gamma)z-\omega_1]t}, \qquad (49b)$$

where γ is a complex propagative eigenvalue of the scattered wave, whose imaginary part gives gain and whose real part gives the (weak-wave) retardation of the scattered wave; δ is also in general a complex number indicating that the vibrational ellipse of the scattered light need not coincide with that of the laser. Substituting (49) in (47), we obtain the linear-eigenvalue equation

$$\gamma\begin{pmatrix}1\\ i\delta\end{pmatrix}=\tfrac{1}{2}g_0\begin{pmatrix}-3\epsilon^2+D(2+\frac{3}{2}\epsilon^2) & 3i\epsilon+D\times\frac{5}{2}i\epsilon\\ -3i\epsilon-D\times\frac{5}{2}i\epsilon & -3+D(2\epsilon^2+\frac{3}{2})\end{pmatrix}\begin{pmatrix}1\\ i\delta\end{pmatrix}, \qquad (50)$$

where g_0 is given by (28). We shall refer to the matrix on the right-hand side of (50) as the net propagation tensor $\Lambda_{ik}=(\gamma_{ik}'+i\Gamma\epsilon_{ik})(2/g_0)$. The two eigenroots are

$$\gamma^{(\pm)}=\tfrac{3}{4}g_0(1+\epsilon^2)\times\left\{-1+\frac{7D}{6}\pm\left[1+\tfrac{1}{3}D\left(1+\frac{16\epsilon^2}{(1+\epsilon^2)^2}\right)+\frac{D^2}{36}\left(1+\frac{96\epsilon^2}{(1+\epsilon^2)^2}\right)\right]^{1/2}\right\}, \qquad (51)$$

whose two corresponding eigenpolarizations are

$$\delta^{(\pm)}=-\frac{\epsilon^2+1}{\epsilon}\left(2+\frac{5D}{3}\right)^{-1}\times\left\{\frac{\epsilon^2-1}{\epsilon^2+1}(1+\tfrac{1}{6}D)\pm\left[1+\tfrac{1}{3}D\left(1+\frac{16\epsilon^2}{(1+\epsilon^2)^2}\right)+\frac{D^2}{36}\left(1+\frac{96\epsilon^2}{(1+\epsilon^2)^2}\right)\right]^{1/2}\right\}. \qquad (52)$$

The gain of these roots is gotten from

$$g^{(\pm)}=-2\,\mathrm{Im}\gamma^{(\pm)}. \qquad (53a)$$

Let us also define a gain normalized to unit intensity (except for a constant factor) as

$$G^{(\pm)}=(g^{(\pm)}/g_0)(1+\epsilon^2)^{-1}. \qquad (53b)$$

Since the gain of the (+) root turns out to be greater than the (−) root, we shall refer to these solutions as the "major" and "minor" eigenmodes, respectively. A table of $G^{(\pm)}$ for various values of ϵ and $\Omega\tau$ is given in Table I. For a given eccentricity ϵ of the incident vibrational ellipse, there is a frequency shift $\Omega_{\mathrm{opt}}^{(\pm)}$ of the scattered light, which will maximize the gain for the two roots. This optimum frequency shift is found by computation from (51) and is plotted against ϵ in Fig. 2 as the dashed curves. The maximized gain per unit intensity $G(\Omega_{\mathrm{opt}}^{(\pm)})$ is plotted against ϵ in Fig. 3 as the dashed curves. If we express $\delta=\rho e^{i\psi}$, then the angle ϕ_1 that the major axis of the vibrational ellipse of the

TABLE I. Backward gain versus ϵ and $\Omega\tau$ for major and minor roots. The gain is normalized with respect to incident intensity (53b) and is expressed in units of $\frac{3}{4}g_0$. [See (51).]

A. Major root

$\epsilon\downarrow$ \ $\Omega\tau\rightarrow$	0.97	0.98	0.99	1.00	1.01
0.0	1.33271	1.33306	1.33327	1.33333	1.33327
0.1	1.36660	1.36689	1.36704	1.36705	1.36692
0.2	1.45618	1.45638	1.45641	1.45630	1.45605
0.3	1.57394	1.57408	1.57406	1.57388	1.57354
0.4	1.69320	1.69338	1.69338	1.69320	1.69286
0.5	1.79713	1.79739	1.79746	1.79735	1.79705
0.6	1.87859	1.87895	1.87910	1.87906	1.87883
0.7	1.93671	1.93714	1.93736	1.93738	1.93721
0.8	1.97377	1.97426	1.97453	1.97460	1.97447
0.9	1.99333	1.99384	1.99414	1.99423	1.99413
1.0	1.99907	1.99959	1.99990	2.00000	1.99990

B. Minor root

$\epsilon\downarrow$ \ $\Omega\tau\rightarrow$	0.98	1.00	1.02	1.04	1.06
0.0	0.99980	1.00000	0.99980	0.99923	0.99830
0.1	0.96596	0.96628	0.96622	0.96579	0.96501
0.2	0.87648	0.87703	0.87723	0.87709	0.87663
0.3	0.75877	0.75945	0.75983	0.75991	0.75972
0.4	0.63948	0.64013	0.64052	0.64067	0.64059
0.5	0.53547	0.53599	0.53629	0.53640	0.53631
0.6	0.45391	0.45428	0.45446	0.45447	0.45432
0.7	0.39572	0.39595	0.39602	0.39595	0.39573
0.8	0.35860	0.35874	0.35873	0.35859	0.35832
0.9	0.33902	0.33910	0.33905	0.33887	0.33857
1.0	0.33327	0.33333	0.33327	0.33308	0.33277

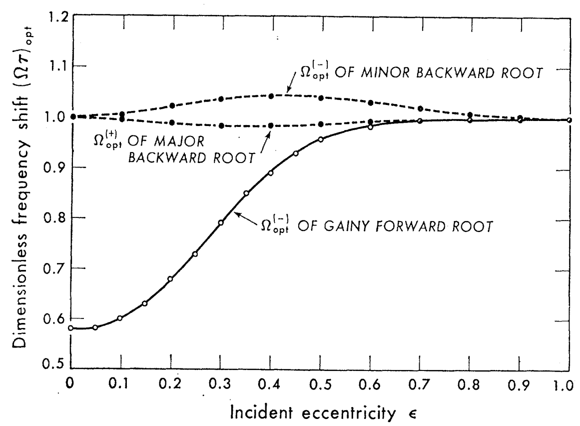

FIG. 2. Frequency shift which maximizes the gain of stimulated Rayleigh-wing scattering for various elliptical polarizations. τ is the molecular-alignment relaxation time. The limiting value of the frequency shift for linear polarization ($\epsilon=0$) in forward scattering is $(\Omega\tau)_{opt}=3^{-1/2}$. (In this figure and all following figures, dots represent computed points.)

scattered light makes with respect to that of the incident light and the eccentricity ϵ_1 of this ellipse are

$$-\phi_1=\tfrac{1}{2}\arctan[2\rho\sin\psi/(1-\rho^2)],$$

$$\epsilon_1=\frac{2\rho\cos\psi}{1+\rho^2+(1-2\rho^2\cos 2\psi+\rho^4)^{1/2}}. \tag{54}$$

$\phi_1(\Omega_{opt}{}^{(\pm)})$ and $\epsilon_1(\Omega_{opt}{}^{(\pm)})$ are plotted against ϵ as the dashed curves in Figs. 4 and 5, respectively. (Note that since Λ_{ik} is non-Hermitian, the eigenpolarizations need not be orthogonal.) Since the eigenroot with the highest gain exponentially dominates the scattering which starts from zero point or thermal fluctuations, a frequency shift of $\Omega_{opt}{}^{(+)}$ will usually be experimentally observed. However, amplifier-type experiments in which the scattered beam is artificially introduced into

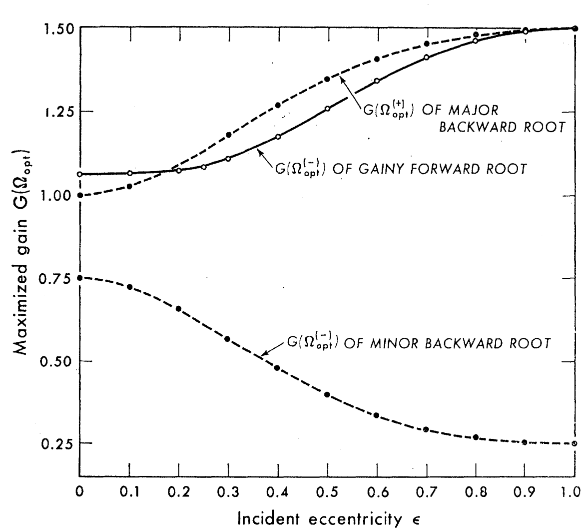

FIG. 3. Maximized normalized gain [Eqs. (52), (53), and (93)]. The unit of gain is g_0 given by (28) and is the maximum stimulated-Rayleigh-wing scattering gain for linear polarization in the backward direction. The limiting value of the gain for linear polarization ($\epsilon=0$) in forward scattering is $\frac{3}{4}\sqrt{2}$.

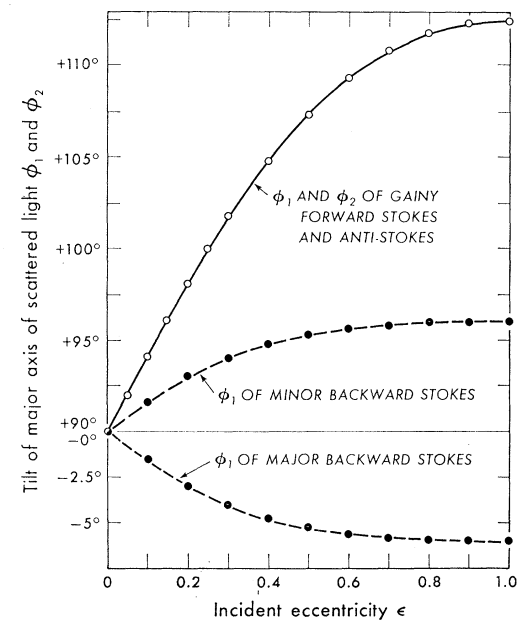

FIG. 4. Angle of tilt between the major axis of incident and the major axis of scattered elliptical polarizations. The limiting value for circular polarization ($\epsilon=1$) of this angle for the backward scattered light is $\pm\frac{1}{2}\arctan(24/113)$, whereas that for the forward scattered light is $-\frac{1}{2}\arctan 1$. Note that the forward Stokes and anti-Stokes ellipses are tilted the same amount.

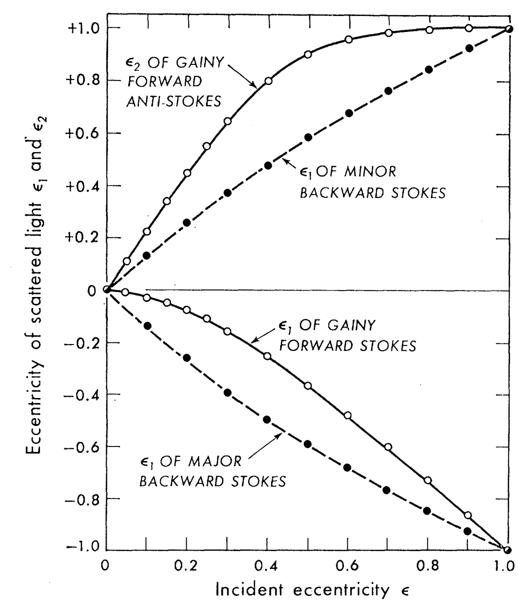

FIG. 5. Eccentricity of the scattered elliptical polarization. Positive and negative eccentricities denote, respectively, co- and counter-rotating senses of polarization relative to that of the incident light. Note that in forward scattering the Stokes is more elongated than the incident polarization, whereas the anti-Stokes is rounder than the incident polarization.

the scattering region must be analyzed as a boundary-value problem involving mixtures of the eigenmodes of propagation.[13]

To obtain a physical insight into these solutions, let us first consider the special cases when $\epsilon=0$ (linearly polarized laser) and $\epsilon=1$ (circularly polarized laser). When $\epsilon=0$, the net propagation tensor Λ_{ik} of (50) becomes

$$\begin{pmatrix} 2D & 0 \\ 0 & -3+\frac{3}{2}D \end{pmatrix}, \tag{55}$$

which has the obvious eigenvalues and eigenpolarizations [also obtainable from (51) and (52)],

$$\begin{aligned} \gamma^{(+)}&=g_0D \quad &\text{and}\quad \begin{pmatrix} 1 \\ i\delta^{(+)} \end{pmatrix}=\begin{pmatrix} 1 \\ 0 \end{pmatrix}, \\ \gamma^{(-)}&=g_0\tfrac{3}{4}(-2+D) \quad &\text{and}\quad \begin{pmatrix} 1 \\ i\delta^{(-)} \end{pmatrix}=\begin{pmatrix} 0 \\ 1 \end{pmatrix}, \end{aligned} \tag{56}$$

which correspond to parallel and perpendicular orientations of the scattered polarization relative to that of the laser, respectively. Taking the imaginary part of $\gamma^{(\pm)}$ and hence of D gives a frequency response factor of $\Omega\tau(1+\Omega^2\tau^2)^{-1}$, which has a maximum at $\Omega_{\text{opt}}{}^{(\pm)}\tau=1$ for both roots. This also implies that Stokes shifts yield gain and anti-Stokes shifts yield loss. For a Stokes shift of $\Omega_{\text{opt}}{}^{(\pm)}=\tau^{-1}$, we therefore have gains of

$$g^{(+)}=g_0 \quad\text{and}\quad g^{(-)}=\tfrac{3}{4}g_0. \tag{57}$$

When $\epsilon=1$, the net propagation tensor becomes

$$\begin{pmatrix} -3+\frac{7}{2}D & i(3+\frac{5}{2}D) \\ -i(3+\frac{5}{2}D) & -3+\frac{7}{2}D \end{pmatrix}, \tag{58}$$

which has the eigenmodes [also obtainable from (51) and (52)]

$$\begin{aligned} \gamma^{(+)}&=3g_0D \quad &\text{and}\quad \begin{pmatrix} 1 \\ i\delta^{(+)} \end{pmatrix}=\begin{pmatrix} 1 \\ -i \end{pmatrix}, \\ \gamma^{(-)}&=g_0(-3+\tfrac{1}{2}D) \quad &\text{and}\quad \begin{pmatrix} 1 \\ i\delta^{(-)} \end{pmatrix}=\begin{pmatrix} 1 \\ +i \end{pmatrix}, \end{aligned} \tag{59}$$

which correspond to counter-rotating and co-rotating scattered polarizations relative to that of the laser, respectively. As was the case when $\epsilon=0$, the gain is optimized at $\Omega_{\text{opt}}{}^{(\pm)}\tau=1$ with

$$g^{(+)}=3g_0, \quad g^{(-)}=\tfrac{1}{2}g_0. \tag{60}$$

Thus, the ratio of the gains per unit intensity of incident light [i.e., $G=(g/g_0)(1+\epsilon^2)^{-1}$] are 4:3:6:1 for parallel-linear, perpendicular-linear, counter-rotating–circular, and co-rotating–circular scattered polarizations, respectively. This ratio is the same as that of spontaneous cross sections for the same polarizations.[14] Since the eigenpolarization of highest gain switches from parallel linear to counter-rotating circular in the extreme limits of $\epsilon=0$ and 1, one expects that for the in-between cases, the elliptical eigenpolarizations are neither the same as nor orthogonal to that of the laser, but somewhere in between, as is verified from Figs. 4 and 5.

An important special case of backward scattering occurs when $\Omega=0$ (i.e., the weak wave is introduced into the medium with an unshifted frequency). Although there is no exponential growth in this case, since $D=1$ and $\gamma^{(\pm)}$ is pure real, there is a weak-wave retardation which affects all backward-going unshifted waves:

$$\gamma^{(\pm)}=\tfrac{3}{4}g_0(1+\epsilon^2)\left\{\frac{1}{6}\pm\left[\left(\frac{7}{6}\right)^2+\frac{8\epsilon^2}{(1+\epsilon^2)^2}\right]^{1/2}\right\}. \tag{61}$$

The eigenpolarizations corresponding to these eigenvalues are

$$\delta^{(\pm)}=-\frac{3}{11}\frac{\epsilon^2+1}{\epsilon}\left\{\frac{7}{6}\frac{\epsilon^2-1}{\epsilon^2+1}\pm\left[\left(\frac{7}{6}\right)^2+\frac{8\epsilon^2}{(1+\epsilon^2)^2}\right]^{1/2}\right\}. \tag{62}$$

In addition, it must be remembered that these eigenmodes of propagation undergo strong-wave retardation and precession (49b). Since the net propagation tensor (50) with $D=1$ is Hermitian, the eigenpolarizations are orthogonal, as can be verified directly from (62). In general, an arbitrarily polarized backward-going wave must be decomposed into a linear combination of these orthogonal eigenpolarizations, and since these eigenpolarizations travel with different speeds, a combination of birefringence and optical activity results. When the forward-going strong wave is linearly polarized ($\epsilon=0$), $\gamma^{(+)}=g_0$, and $\gamma^{(-)}=-\frac{3}{4}g_0$, which produces birefringence[15]; when the incident wave is circularly polarized ($\epsilon=1$), $\gamma^{(+)}=3g_0$, and $\gamma^{(-)}=-\frac{5}{2}g_0$, which produces optical activity. The corresponding eigenpolarizations are given by (56) and (59). For the circular eigenpolarizations, the over-all precession of (49b) becomes a retardative effect and combines with the eigenroots $\gamma^{(\pm)}$ to give total propagation constants $\kappa+\gamma^{(+)}-\Gamma=\frac{7}{2}g_0$ and $\kappa+\gamma^{(-)}+\Gamma=g_0$ for the major and minor eigenmodes, respectively. Hence, the ratio of speeds relative to that of the strong wave and normalized to unit intensity of the incident wave is 4:−3:6:1 for parallel-linear, perpendicular-linear, counter-rotating–circular, and co-rotating–circular

[13] R. L. Carman, R. Y. Chiao, and P. L. Kelley, Phys. Rev. Letters **17**, 1281 (1966).

[14] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon Publishing Corp., New York, 1960), p. 383. This ratio was also obtained by R. W. Minck, E. E. Hagenlocker, and W. G. Rado, Phys. Rev. Letters **17**, 229 (1966) for stimulated pure rotational Raman scattering, which is also a traceless symmetric tensor scattering process.

[15] This birefringence implies angular momentum transfer between the forward-going strong wave and the backward-going weak wave and produces small modification of eccentricity of the vibrational ellipse of the strong wave as a function of z.

polarizations of the weak backward-going unshifted wave, respectively.

Another simple special case occurs when $\Omega=\infty$, or more precisely $\Omega\tau\gg 1$, in which case the molecular alignment fails to respond to the weak backward wave (i.e., $\chi_{ik}^{11}=0$ and there is no self-coupling). Again $\gamma^{(\pm)}$ is pure real, since now $D=0$, and we expect no weak-wave retardation:

$$\gamma^{(+)}=0\,, \qquad \delta^{(+)}=-\epsilon\,,$$
$$\gamma^{(-)}=\tfrac{3}{2}g_0(1+\epsilon^2)\,, \quad \delta^{(-)}=+1/\epsilon. \tag{63}$$

$\gamma^{(-)}$ is nonzero not because of weak-wave retardation, but simply from the birefringence induced solely by the strong wave. Note that the eigenpolarization corresponding to the zero root describes a vibrational ellipse identical to that of the strong forward wave, except it is of the opposite sense, and the other eigenpolarization is orthogonal. Further discussion of this case will be deferred to the end of Sec. V.

V. FORWARD SCATTERING

In the case of forward scattering, we must allow for the possibility of an anti-Stokes channel as well as a Stokes channel, since momentum conservation permits the simultaneous coupling into both channels.[2] We take this consideration into account by introducing the two weak scattered waves

$$\mathbf{E}_1=\boldsymbol{\varepsilon}_1(z)e^{i(k_1z+\kappa z-\omega_1t)}\,,$$
$$\mathbf{E}_2=\boldsymbol{\varepsilon}_2(z)e^{i(k_2z+\kappa z-\omega_2t)}\,, \tag{64}$$

where $\omega_1=\omega_0-\Omega$, $\omega_2=\omega_0+\Omega$, $k_1=n_0\omega_1/c$, and $k_2=n_0\omega_2/c$ (we neglect dispersion); κ is given by (30a) and $\kappa\ll k_0$; $\Omega\ll\omega_0$; $|\mathcal{E}_{1,2}|\ll\mathcal{E}_0$; and $\boldsymbol{\varepsilon}_{1,2}(z)$ are slowly varying over distances of the order of a wavelength. The intense incident laser wave $\mathbf{E}_0$ is given previously by (31). The sum field $\mathbf{E}=\mathbf{E}_0+\mathbf{E}_1+\mathbf{E}_2$ interacts with the medium through Eq. (7) to produce a susceptibility change:

$$\begin{aligned}\Delta\chi_{ik}=\tfrac{1}{2}K\tau[&(E_{0i}^*E_{0k}-\tfrac{1}{3}\delta_{ik}E_{0j}^*E_{0j})\\ &+D(E_{0i}^*E_{1k}+E_{0k}^*E_{1i}-\tfrac{2}{3}\delta_{ik}E_{0j}^*E_{1j})\\ &+D(E_{0i}E_{2k}^*+E_{0k}E_{2i}^*-\tfrac{2}{3}\delta_{ik}E_{0j}E_{2j}^*)],\end{aligned} \tag{65}$$

where D is given by (34), and where we have neglected terms of the order of $\mathcal{E}_1^2$, $\mathcal{E}_2^2$, and $\mathcal{E}_1\mathcal{E}_2$. All time-varying terms in (65) vary as $e^{+i\Omega t}$.

The induced polarization, again neglecting terms of the order $\mathcal{E}_1^2$, $\mathcal{E}_2^2$, and $\mathcal{E}_1\mathcal{E}_2$, is

$$\begin{aligned}\Delta P_i&=\tfrac{1}{2}(\Delta\chi_{ik}E_k+\Delta\chi_{ik}^*E_k)\\ &=\chi_{ik}^0(E_{0k}+E_{1k}+E_{2k}^*)\\ &\qquad+(\chi_{ik}^{\mathrm{I}}+\chi_{ik}^{\mathrm{II}})(E_{0k}+E_{0k}^*)\,,\end{aligned} \tag{66}$$

where χ_{ik}^0 and χ_{ik}^{I} are given by (36) and χ_{ik}^{II} is given by

$$\chi_{ik}^{\mathrm{II}}=D\tfrac{1}{4}K\tau(E_{0i}E_{2k}^*+E_{0k}E_{2i}^*-\tfrac{2}{3}\delta_{ik}E_{0j}E_{2j}^*). \tag{67}$$

Substituting into the wave equation, we get

$$\begin{aligned}&2ik_0\left(\frac{1}{\mathcal{E}_{1i}}\frac{d\mathcal{E}_{1i}}{dz}+i\kappa\right)E_{1i}\\ &\qquad=-4\pi\frac{\omega_1^2}{c^2}(\chi_{ik}^0E_{1k}+\chi_{ik}^{\mathrm{I}}E_{0k}+\chi_{ik}^{\mathrm{II}}E_{0k})\,,\\ &-2ik_0\left(\frac{1}{\mathcal{E}_{2i}^*}\frac{d\mathcal{E}_{2i}^*}{dz}-i\kappa\right)E_{2i}^*\\ &\qquad=-4\pi\frac{\omega_2^2}{c^2}(\chi_{ik}^0E_{2k}^*+\chi_{ik}^{\mathrm{I}}E_{0k}^*+\chi_{ik}^{\mathrm{II}}E_{0k}^*).\end{aligned} \tag{68}$$

Let us assume $\omega_1\cong\omega_0\cong\omega_2$ and $k_1\cong k_0\cong k_2$. Then the last term on the left-hand side can be subtracted from the first term of the right-hand side of both equations in (64) by introducing $\chi_{ik}^{10}=\chi_{ik}^{20}$ given by (38) in place of χ_{ik}^0. This eliminates the isotropic contribution of strong-wave retardation from the propagation of both scattered waves.

To express (68) as differential equations for the scattered fields E_{1k} and E_{2k}^*, we juggle the order of the tensor products of the last two terms so that

$$\begin{aligned}\chi_{ik}^{\mathrm{I}}E_{0k}&=\chi_{ik}^{11}E_{1k}\,,\\ \chi_{ik}^{\mathrm{II}}E_{0k}^*&=\chi_{ik}^{22}E_{2k}^*\,,\\ \chi_{ik}^{\mathrm{II}}E_{0k}&=X_{ik}^{12}E_{2k}^*\,,\\ \chi_{ik}^{\mathrm{I}}E_{0k}^*&=X_{ik}^{21}E_{1k}\,,\end{aligned} \tag{69}$$

where it can be shown that

$$\begin{aligned}\chi_{ik}^{11}&=D\tfrac{1}{4}K\tau(E_{0i}^*E_{0k}-\tfrac{2}{3}E_{0k}^*E_{0i}+\delta_{ik}E_{0j}^*E_{0j})\,,\\ \chi_{ik}^{22}&=D\tfrac{1}{4}K\tau(E_{0i}E_{0k}^*-\tfrac{2}{3}E_{0k}E_{0i}^*+\delta_{ik}E_{0j}E_{0j}^*)\,,\\ X_{ik}^{12}&=D\tfrac{1}{4}K\tau(\tfrac{1}{3}E_{0i}E_{0k}+\delta_{ik}E_{0j}E_{0j})\,,\\ X_{ik}^{21}&=D\tfrac{1}{4}K\tau(\tfrac{1}{3}E_{0i}^*E_{0k}^*+\delta_{ik}E_{0j}^*E_{0j}^*).\end{aligned} \tag{70}$$

We shall refer to χ_{ik}^{11} and χ_{ik}^{22} as "self-coupling" and X_{ik}^{12} and X_{ik}^{21} as "cross-coupling" susceptibilities since they self-couple and cross-couple the Stokes and anti-Stokes waves, respectively. Since X_{ik}^{12} and X_{ik}^{21} vary as $e^{-2i\omega_0t}$ and $e^{+2i\omega_0t}$, respectively, it is convenient to define the time-independent quantities as

$$\begin{aligned}\chi_{ik}^{12}&=X_{ik}^{12}\exp[-2i(k_0z+\kappa z-\omega_0t)]\,,\\ \chi_{ik}^{21}&=X_{ik}^{21}\exp[+2i(k_0z+\kappa z-\omega_0t)].\end{aligned} \tag{71}$$

The quantities χ_{ik}^{11}, χ_{ik}^{22}, χ_{ik}^{12}, and χ_{ik}^{21} are now all z-independent as well as time-independent, except for a slowly varying dependence on z due to the precession of the incident vibrational ellipse. The wave equation, in terms of these susceptibilities, is

$$\begin{aligned}-i\frac{d\mathcal{E}_{1i}}{dz}&=2\pi\frac{k_0}{n_0^2}[(\chi_{ik}^{10}+\chi_{ik}^{11})\mathcal{E}_{1k}+\chi_{ik}^{12}\mathcal{E}_{2k}^*]\,,\\ +i\frac{d\mathcal{E}_{2i}^*}{dz}&=2\pi\frac{k_0}{n_0^2}[(\chi_{ik}^{20}+\chi_{ik}^{22})\mathcal{E}_{2k}^*+\chi_{ik}^{21}\mathcal{E}_{1k}].\end{aligned} \tag{72}$$

To express these equations in terms of a conventional linear-eigenvalue problem, let us transform these equations into the rotating frame by introducing

$$\chi_{ik}{}^{10}=R_{ij}R_{kl}\chi_{jl}{}^{10'}, \quad \text{etc.}, \tag{73}$$

and

$$\mathcal{E}_{1i}=R_{ik}\mathcal{E}_{1k}{}' \quad \text{and} \quad \mathcal{E}_{2i}{}^{*}=R_{ik}\mathcal{E}_{2k}{}^{*\prime}, \tag{74}$$

where R_{ij} is given by (43). Since in the rotating frame the strong-wave, self-coupling, and cross-coupling susceptibility tensors all involve tensor products of the primed incident field (26) which are z-independent, these susceptibilities are constants evaluated at $z=0$

$$\begin{aligned}
\chi_{ik}{}^{10'}=\chi_{ik}{}^{20'}&=\tfrac{1}{4}K\tau[\mathcal{E}_{0i}{}^{*}(0)\mathcal{E}_{0k}(0)\\
&\quad+\mathcal{E}_{0k}{}^{*}(0)\mathcal{E}_{0i}(0)-2\delta_{ik}\mathcal{E}_{0j}{}^{*}(0)\mathcal{E}_{0j}(0)],\\
\chi_{ik}{}^{12'}D^{-1}&=\chi_{ik}{}^{21'*}D^{-1*}\\
&=\tfrac{1}{4}K\tau[\tfrac{1}{3}\mathcal{E}_{0i}(0)\mathcal{E}_{0k}(0)+\delta_{ik}\mathcal{E}_{0j}(0)\mathcal{E}_{0j}(0)],\\
\chi_{ik}{}^{11'}=\chi_{ki}{}^{22'}&=D\tfrac{1}{4}K\tau[\mathcal{E}_{0i}{}^{*}(0)\mathcal{E}_{0k}(0)\\
&\quad-\tfrac{2}{3}\mathcal{E}_{0k}{}^{*}(0)\mathcal{E}_{0i}(0)+\delta_{ik}\mathcal{E}_{0j}{}^{*}(0)\mathcal{E}_{0j}(0)],
\end{aligned} \tag{75}$$

or, evaluated using (17),

$$\begin{aligned}
(\chi_{ik}{}^{10'})=(\chi_{ik}{}^{20'})&=\tfrac{1}{4}K\tau|\mathcal{E}_{00}|^2\begin{pmatrix}-2\epsilon^2 & 0\\ 0 & -2\end{pmatrix},\\
(\chi_{ik}{}^{12'})D^{-1}=(\chi_{ik}{}^{21'*})D^{-1*}&=\tfrac{1}{4}K\tau\mathcal{E}_{00}{}^2\begin{pmatrix}\tfrac{4}{3}-\epsilon^2 & \tfrac{1}{3}i\epsilon\\ \tfrac{1}{3}i\epsilon & -\tfrac{4}{3}\epsilon^2+1\end{pmatrix},\\
(\chi_{ik}{}^{11'})D^{-1}=(\chi_{ik}{}^{22'})^TD^{-1}&=\tfrac{1}{4}K\tau|\mathcal{E}_{00}|^2\begin{pmatrix}\tfrac{4}{3}+\epsilon^2 & \tfrac{5}{3}i\epsilon\\ -\tfrac{5}{3}i\epsilon & \tfrac{4}{3}\epsilon^2+1\end{pmatrix}.
\end{aligned} \tag{76}$$

We shall take $\mathcal{E}_{00}$ real henceforth, so that $|\mathcal{E}_{00}|^2=\mathcal{E}_{00}{}^2$. Let us introduce the 4×4 matrices

$$(\chi'_{\alpha\beta})=\begin{pmatrix}(\chi_{ik}{}^{10'}+\chi_{ik}{}^{11'}) & (\chi_{ik}{}^{12'})\\ -(\chi_{ik}{}^{21'}) & -(\chi_{ik}{}^{20'}+\chi_{ik}{}^{22'})\end{pmatrix}, \tag{77}$$

$$(R_{\alpha\beta})=\begin{pmatrix}(R_{ik}) & (0)\\ (0) & (R_{ik})\end{pmatrix}, \tag{78}$$

$$(\epsilon_{\alpha\beta})=\begin{pmatrix}(\epsilon_{ik}) & (0)\\ (0) & (\epsilon_{ik})\end{pmatrix}, \tag{79}$$

and the 4-dimensional column vector

$$(\mathcal{E}'_{\alpha})=\begin{pmatrix}(\mathcal{E}'_{1i})\\ (\mathcal{E}'^{*}{}_{2i})\end{pmatrix}=(R^{-1}{}_{\alpha\beta}\mathcal{E}_{\beta}). \tag{80}$$

Then the coupled wave equations (72) in the rotating coordinate frame become the single equation

$$i\left(\frac{d}{dz}\mathcal{E}'_{\alpha}-\Gamma\epsilon_{\alpha\beta}\mathcal{E}'_{\beta}\right)=\gamma'_{\alpha\beta}\mathcal{E}'_{\beta}, \tag{81}$$

where Γ is given by (30b), and

$$\gamma'_{\alpha\beta}=-2\pi(k_0/n_0{}^2)\chi'_{\alpha\beta} \tag{82}$$

is a z-independent propagation matrix. We shall call $(\gamma'_{\alpha\beta}+i\Gamma\epsilon_{\alpha\beta})2/g_0=\Lambda_{\alpha\beta}$ the *net* propagation matrix, which is to be diagonalized. We introduce the ansatz

$$(\mathcal{E}_{\alpha}{}'(z))=\begin{bmatrix}\mathcal{E}_{1x}{}'(z)\\ \mathcal{E}_{1y}{}'(z)\\ \mathcal{E}_{2x}{}'^{*}(z)\\ \mathcal{E}_{2y}{}'^{*}(z)\end{bmatrix}=\begin{bmatrix}\mathcal{E}_{1x}(0)\\ \mathcal{E}_{1y}(0)\\ \mathcal{E}_{2x}{}^{*}(0)\\ \mathcal{E}_{2y}{}^{*}(0)\end{bmatrix}e^{i\gamma z}
=\begin{bmatrix}\mathcal{E}_{10}\begin{pmatrix}1\\ i\delta_1\end{pmatrix}\\ \mathcal{E}_{20}{}^{*}\begin{pmatrix}1\\ -i\delta_2{}^{*}\end{pmatrix}\end{bmatrix}e^{i\gamma z} \tag{83a}$$

or

$$\begin{aligned}
\begin{pmatrix}E_{1x}(z)\\ E_{1y}(z)\end{pmatrix}&=\begin{pmatrix}\cos\Gamma z & -\sin\Gamma z\\ \sin\Gamma z & \cos\Gamma z\end{pmatrix}\begin{pmatrix}1\\ i\delta_1\end{pmatrix}\times\mathcal{E}_{10}e^{i[(k_1+\kappa+\gamma)z-\omega_1 t]},\\
\begin{pmatrix}E_{2x}(z)\\ E_{2y}(z)\end{pmatrix}&=\begin{pmatrix}\cos\Gamma z & -\sin\Gamma z\\ \sin\Gamma z & \cos\Gamma z\end{pmatrix}\begin{pmatrix}1\\ i\delta_2\end{pmatrix}\times\mathcal{E}_{20}e^{i[(k_2+\kappa-\gamma^{*})z-\omega_2 t]}.
\end{aligned} \tag{83b}$$

Substituting (83) into (81), we obtain the eigenvalue equation

$$\gamma\begin{bmatrix}\mathcal{E}_{1x}(0)\\ \mathcal{E}_{1y}(0)\\ \mathcal{E}_{2x}{}^{*}(0)\\ \mathcal{E}_{2y}{}^{*}(0)\end{bmatrix}=\tfrac{1}{2}g_0\begin{bmatrix}-3\epsilon^2+D(2+\tfrac{3}{2}\epsilon^2) & -3i\epsilon+D\times\tfrac{5}{2}i\epsilon & D(2-\tfrac{3}{2}\epsilon^2) & D\times\tfrac{1}{2}i\epsilon\\ +3i\epsilon-D\times\tfrac{5}{2}i\epsilon & -3+D(2\epsilon^2+\tfrac{3}{2}) & D\times\tfrac{1}{2}i\epsilon & D(-2\epsilon^2+\tfrac{3}{2})\\ D(-2+\tfrac{3}{2}\epsilon^2) & D\times\tfrac{1}{2}i\epsilon & 3\epsilon^2-D(2+\tfrac{3}{2}\epsilon^2) & -3i\epsilon+D\times\tfrac{5}{2}i\epsilon\\ D\times\tfrac{1}{2}i\epsilon & D(2\epsilon^2-\tfrac{3}{2}) & 3i\epsilon-D\times\tfrac{5}{2}i\epsilon & 3-D(2\epsilon^2+\tfrac{3}{2})\end{bmatrix}\begin{bmatrix}\mathcal{E}_{1x}(0)\\ \mathcal{E}_{1y}(0)\\ \mathcal{E}_{2x}{}^{*}(0)\\ \mathcal{E}_{2y}{}^{*}(0)\end{bmatrix}, \tag{84}$$

where g_0 is given by (28). The net propagation matrix has the form

$$\begin{bmatrix} a & c & d & f \\ -c & b & f & e \\ -d & f & -a & c \\ f & -e & -c & -b \end{bmatrix} \equiv (\Lambda_{\alpha\beta}), \tag{85}$$

where

$$\begin{aligned} a&=-3\epsilon^2+D(2+\tfrac{3}{2}\epsilon^2), \\ b&=-3+D(2\epsilon^2+\tfrac{3}{2}), \\ c&=-3i\epsilon+D\times\tfrac{5}{2}i\epsilon, \\ d&=D\times(2-\tfrac{3}{2}\epsilon^2), \\ e&=D\times(-2\epsilon^2+\tfrac{3}{2}), \\ f&=D\times\tfrac{1}{2}i\epsilon. \end{aligned} \tag{86}$$

The eigenvalues of (85) are

$$\lambda=\pm\{-A\pm[A^2-(\det\Lambda)^2]^{1/2}\}^{1/2}, \tag{87}$$

where $\gamma=\frac{1}{2}g_0\lambda$ and

$$A=-\tfrac{1}{2}(a^2+b^2-2c^2-d^2-e^2+2f^2), \tag{88}$$

$$\det\Lambda=(a^2-d^2)(b^2-e^2)+2(ab-de)(c^2+f^2) -4cf(ae-bd)+(f^2-c^2)^2. \tag{89}$$

Calculation of $\det\Lambda$ from (86) gives

$$\det\Lambda=0, \tag{90}$$

which simplifies the eigenvalues to

$$\lambda=\pm(-2A)^{1/2},\ 0. \tag{91}$$

Calculation from (86) and (88) gives

$$\begin{aligned} A&=9\{-2\epsilon^2D^2+\tfrac{1}{2}[(1+\epsilon^2)^2+4\epsilon^2]D-\tfrac{1}{2}(1+\epsilon^2)^2\} \\ &=-[\tfrac{9}{2}(1+\epsilon^2)^2-18\epsilon^2D](1-D). \end{aligned} \tag{92}$$

Hence the nonzero eigenvalues of (84) are

$$\begin{aligned} \gamma^{(\pm)}&=\pm\tfrac{3}{2}g_0(1+\epsilon^2) \\ &\quad\times\left[1-D\left(1+\frac{4\epsilon^2}{(1+\epsilon^2)^2}\right)+D^2\left(\frac{4\epsilon^2}{(1+\epsilon^2)^2}\right)\right]^{1/2} \\ &=\pm\tfrac{3}{2}g_0(1+\epsilon^2)(1-D)^{1/2}\left(1-\frac{4\epsilon^2}{(1+\epsilon^2)^2}D\right)^{1/2}. \end{aligned} \tag{93}$$

To obtain the eigenpolarizations associated with these nonzero eigenvalues, let us first notice that, using (86),

$$\begin{bmatrix} a & c & d & f \\ -c & b & f & e \\ -d & f & -a & c \\ f & -e & -c & -b \end{bmatrix} \begin{bmatrix} 1 \\ i\epsilon \\ -1 \\ i\epsilon \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \end{bmatrix}, \tag{94}$$

so that $(1, i\epsilon, -1, i\epsilon)^T$ is an eigenpolarization of the zero eigenvalue.[16] (This implies immediately that $\det\Lambda=0$.) It can be easily verified that this is its only eigenpolarization ($\Lambda_{\alpha\beta}$ is non-Hermitian). Physically, this states that if we introduce Stokes and anti-Stokes polarized the same way as the strong wave, with the anti-Stokes amplitude equal to but 180° out of phase with respect to the Stokes amplitude, then these waves propagate with canceling cross and self-couplings (i.e., no exponential growth and no weak-wave retardation). This result is independent of frequency shift Ω of the weak waves. The underlying reason for this behavior is that the anti-Stokes photon annihilates the "phonons" created by the Stokes photons, with equal probabilities of annihilation and creation for this choice of polarization and relative phase. However, it must be remembered that this eigenpolarization still undergoes the strong-wave precession and retardation (83b).

Having found one eigenvector, it is natural to construct a unitary matrix U with this eigenvector as the first column, with three mutually orthogonal vectors as the next three columns, and have $\Lambda_{\alpha\beta}$ undergo a unitary transformation using U. A natural choice for such a matrix is

$$(U_{\alpha\beta})=\begin{bmatrix} 1 & i\epsilon & 1 & i\epsilon \\ i\epsilon & 1 & i\epsilon & 1 \\ -1 & i\epsilon & 1 & -i\epsilon \\ i\epsilon & -1 & -i\epsilon & 1 \end{bmatrix}\times 2^{-1/2}(1+\epsilon^2)^{-1/2}. \tag{95}$$

It can be shown that

$$(\Lambda_{\alpha\delta}{}^U)=(U_{\alpha\beta}{}^\dagger\Lambda_{\beta\gamma}U_{\gamma\delta})=\begin{bmatrix} 0 & c_{12} & c_{13} & 0 \\ 0 & 0 & 0 & c_{24} \\ 0 & 0 & 0 & 0 \\ 0 & c_{42} & c_{43} & 0 \end{bmatrix}, \tag{96}$$

where

$$\begin{aligned} c_{12}&=6Di\epsilon[(1-\epsilon^2)/(1+\epsilon^2)], \\ c_{13}&=4D[(1-\epsilon^2+\epsilon^4)/(1+\epsilon^2)], \\ c_{24}&=-3(1+\epsilon^2)(1-D), \\ c_{42}&=-3(1+\epsilon^2)+[12\epsilon^2D/(1+\epsilon^2)], \\ c_{43}&=-6Di\epsilon[(1-\epsilon^2)/(1+\epsilon^2)]=-c_{12}. \end{aligned} \tag{97}$$

Then $\Lambda_{\alpha\beta}{}^U$ possesses a characteristic equation $\lambda^2(\lambda^2-c_{24}c_{42})=0$ which reduces to (91), as it must. The eigenvectors in the new representation are

$$\mathcal{E}_\alpha{}^U=U_{\alpha\beta}{}^{-1}\mathcal{E}_\beta. \tag{98}$$

So if

$$(\mathcal{E}_\alpha{}^U)=\begin{bmatrix} 1 \\ u \\ v \\ w \end{bmatrix}, \tag{99}$$

[16] Another solution associated with linear growth (as is usual with degenerate zero roots) is

$$(\mathcal{E}_{1x},\mathcal{E}_{1y},\mathcal{E}_{2x}^*,\mathcal{E}_{2y}^*)^T=[(1,i\delta_0,1,-i\delta_0)^T-i\gamma_0z(1,i\epsilon,-1,i\epsilon)^T],$$

where

$$\delta_0=\epsilon[3(\epsilon^2+1)-6D]/[3(\epsilon^2+1)-6\epsilon^2D],$$

and

$$\gamma_0=12D[\epsilon^4+1-\epsilon^2(1+D)]/[3(\epsilon^2+1)-6\epsilon^2D].$$

This along with the other three eigenpolarizations (94) and (103) can satisfy arbitrary boundary conditions at $z=0$.

then

$$(\mathcal{E}_\beta)=1\begin{bmatrix}1\\ i\epsilon\\ -1\\ i\epsilon\end{bmatrix}+u\begin{bmatrix}i\epsilon\\ 1\\ i\epsilon\\ -1\end{bmatrix}+v\begin{bmatrix}1\\ i\epsilon\\ 1\\ -i\epsilon\end{bmatrix}+w\begin{bmatrix}i\epsilon\\ 1\\ -i\epsilon\\ 1\end{bmatrix}, \quad (100)$$

so that the basis vectors of the new representation are the columns of U as usual. It has already been noted that the first basis vector is already an eigenvector of the zero eigenvalue. The other eigenvectors, however, must also in general contain this basis vector because $\Lambda_{\alpha\beta}$ is non-Hermitian and eigenvectors need not be orthogonal. In this new representation, the eigenvectors satisfy

$$\begin{bmatrix}0 & c_{12} & c_{13} & 0\\ 0 & 0 & 0 & c_{24}\\ 0 & 0 & 0 & 0\\ 0 & c_{42} & c_{43} & 0\end{bmatrix}\begin{bmatrix}1\\ u\\ v\\ w\end{bmatrix}=\lambda\begin{bmatrix}1\\ u\\ v\\ w\end{bmatrix}, \quad (101)$$

which implies for the nonzero roots that

$$u^{(\pm)}=\frac{\lambda^{(\pm)}}{c_{12}}=\pm\frac{1}{2i\epsilon D}\left(\frac{1+\epsilon^2}{1-\epsilon^2}\right)(1-D)^{1/2}\times[(1+\epsilon^2)^2-4\epsilon^2D]^{1/2},$$

$$v^{(\pm)}=0, \quad (102)$$

$$w^{(\pm)}=\frac{\lambda^{(\pm)2}}{c_{12}c_{24}}=\frac{c_{42}}{c_{12}}=-\frac{(1+\epsilon^2)^2-4\epsilon^2D}{2i\epsilon D(1-\epsilon^2)},$$

or, expressing the eigenvectors in terms of the second basis vector,

$$\begin{bmatrix}\mathcal{E}_{1x}^{(-)}(0)\\ \mathcal{E}_{1y}^{(-)}(0)\\ \mathcal{E}_{2x}^{(-)*}(0)\\ \mathcal{E}_{2y}^{(-)*}(0)\end{bmatrix}=\begin{bmatrix}i\epsilon\{1-B+C\}\\ 1\{1+B\epsilon^2+C\}\\ i\epsilon\{1+B-C\}\\ -1\{1-B\epsilon^2-C\}\end{bmatrix}=\begin{bmatrix}\mathcal{E}_{2x}^{(+)*}(0)\\ -\mathcal{E}_{2y}^{(+)*}(0)\\ \mathcal{E}_{1x}^{(+)}(0)\\ -\mathcal{E}_{1y}^{(+)}(0)\end{bmatrix}, \quad (103)$$

where

$$B=2\left(\frac{1-\epsilon^2}{1+\epsilon^2}\right)D(1-D)^{-1/2}\times[(1+\epsilon^2)^2-4\epsilon^2D]^{-1/2}=\frac{1}{i\epsilon u^{(-)}}, \quad (104)$$

$$C=\left(\frac{1-4\epsilon^2D/(1+\epsilon^2)^2}{1-D}\right)^{1/2}=\frac{w^{(-)}}{u^{(-)}}.$$

Another way of expressing these results [cf. (83)] is

$$\frac{\mathcal{E}_{10}^{(-)}}{\mathcal{E}_{20}^{(-)*}}=\frac{1-B+C}{1+B-C}=\frac{\mathcal{E}_{20}^{(+)*}}{\mathcal{E}_{10}^{(+)}}, \quad (105a)$$

$$\delta_1^{(-)}=-\frac{1+B\epsilon^2+C}{1-B+C}\frac{1}{\epsilon}=\delta_2^{(+)*},$$

$$\delta_2^{(-)*}=-\frac{1-B\epsilon^2-C}{1+B-C}\frac{1}{\epsilon}=\delta_1^{(+)}. \quad (105b)$$

It should be remarked that the exchange symmetry that shows up in the eigenvectors (103) is a general property of any matrix of the form (85). For, if $(1,u,v,w)^T$ is an eigenvector of (85) associated with an eigenvalue $+\lambda$, then it is easily verified that

$$\begin{bmatrix}a & c & d & f\\ -c & b & f & e\\ -d & f & -a & c\\ f & -e & -c & -b\end{bmatrix}\begin{bmatrix}v\\ -w\\ 1\\ -u\end{bmatrix}=-\lambda\begin{bmatrix}v\\ -w\\ 1\\ -u\end{bmatrix}. \quad (106)$$

This means that if one exchanges the Stokes and anti-Stokes eigenpolarizations with a change of the sign of the y (or x) components [i.e., reflecting the weak-wave vibrational ellipses across the x (or y) axis or the major (or minor) axis of the incident vibrational ellipse] one obtains the eigenvector associated with the negative eigenvalue. A second way of exchanging Stokes and anti-Stokes waves is simply to let $\Omega\rightarrow-\Omega$ or $D\rightarrow D^*$. If, at the same time, we let $\epsilon\rightarrow-\epsilon$ (i.e., reverse the sense of the laser polarization), then all the matrix elements of (85) are complex conjugated, so that the eigenvalues and eigenvectors are also complex conjugated. But by inspection of (103), one sees that the operation $\epsilon\rightarrow-\epsilon$ changes the sign of the x (or y) components of the Stokes and anti-Stokes waves [i.e., reversing the sense of the incident vibrational ellipse not only reverses the sense of scattered vibrational ellipses, but also reflects them across the y (or x) axis], so that the two ways of exchange yield the same physical result as is expected. In particular, in either case the gain $g=-2\ \mathrm{Im}\gamma$ reverses sign.

Since $\gamma^{(+)}$ turns outsto be the "lossy" root, we shall henceforth eliminate it from discussion. The zero root has already been discussed. The "gainy" root $\gamma^{(-)}$ has a normalized gain $G^{(-)}$ (53b) which is given for various values of ϵ and $\Omega\tau$ in Table II. For a given eccentricity ϵ

TABLE II. Forward gain versus ϵ and $\Omega\tau$. The gain is normalized with respect to incident intensity and is that of the gainy root expressed in units of $\frac{3}{4}g_0$ [see (93).]

$\epsilon\downarrow$ $\Omega\tau\rightarrow$	0.55	0.6	0.7	0.8	0.9	1.0	1.04
0.0	1.41329	1.41362	1.39887	1.36953	1.33102	1.28719	1.26880
0.1	1.41199	1.41506	1.40548	1.38071	1.34604	1.30534	1.28801
0.2	1.41068	1.42166	1.42689	1.41503	1.39110	1.35899	1.34454
0.3	1.41625	1.43940	1.46690	1.47387	1.46517	1.44491	1.43434
0.4	1.43737	1.47511	1.52837	1.55619	1.56361	1.55534	1.54857
0.5	1.47935	1.53134	1.00861	1.65475	1.67558	1.67669	1.67274
0.6	1.53821	1.60130	1.69642	1.75543	1.78512	1.79195	1.78958
0.7	1.60011	1.67013	1.77602	1.84242	1.87688	1.88646	1.88474
0.8	1.65002	1.72364	1.83506	1.90512	1.94176	1.95241	1.95086
0.9	1.67984	1.75506	1.86890	1.94051	1.97803	1.98901	1.98748
1.0	1.68906	1.76471	1.87919	1.95122	1.98895	2.00000	1.99846

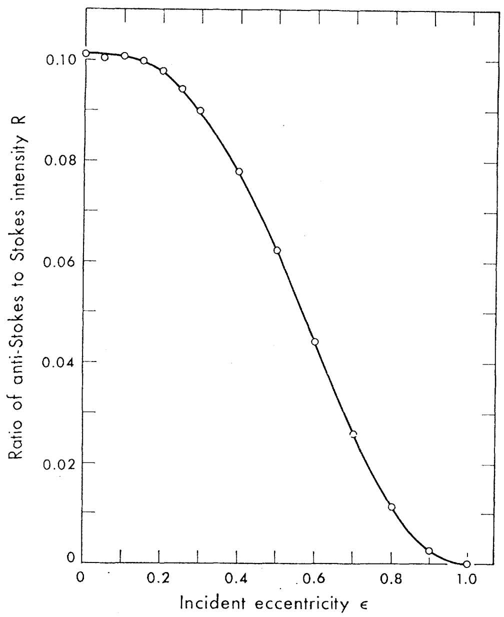

FIG. 6. Ratio of anti-Stokes to Stokes intensities. This ratio for linear polarization ($\epsilon=0$) is $(5+6\sqrt{\frac{2}{3}})^{-1}$ and for circular polarization ($\epsilon=1$) is zero, indicating the absence of the anti-Stokes channel.

of the incident vibrational ellipse, there is a frequency shift $\Omega_{\text{opt}}^{(-)}$ of the scattered light which will maximize the gain. This optimum frequency shift is found by computation from (93) and is plotted against ϵ in Fig. 2 as the solid curve. The maximized gain $G(\Omega_{\text{opt}}^{(-)})$ is plotted against ϵ in Fig. 3 as the solid curve. Expressing $\delta_1^{(-)}$ and $\delta_2^{(-)}$ of (105b) in polar form and using (54), we can find the angles $\phi_1^{(-)}$ and $\phi_2^{(-)}$ that the major axes of the vibrational ellipse of the Stokes and anti-Stokes waves, respectively, make with respect to that of the incident light. These angles evaluated at $\Omega_{\text{opt}}^{(-)}$ turn out to be equal and are plotted against ϵ in Fig. 4 as the single solid curve. The eccentricities $\epsilon_1^{(-)}$ and $\epsilon_2^{(-)}$ of these vibrational ellipses evaluated at $\Omega_{\text{opt}}^{(-)}$ are plotted against ϵ in Fig. 5 as the solid lines. The ratio of anti-Stokes to Stokes intensities $R=|\mathcal{E}_{20}^{(-)*}/\mathcal{E}_{10}^{(-)}|^2 \times(1+|\delta_2^{(-)}|^2)/(1+|\delta_1^{(-)}|^2)$ is plotted against ϵ in

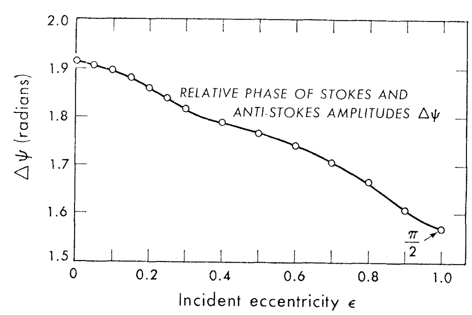

FIG. 7. Relative phase of Stokes to anti-Stokes amplitudes $\mathcal{E}_{10}$ and $\mathcal{E}_{20}^*$. The limit for linear polarization ($\epsilon=0$) is $\Delta\psi=-\arctan(2\sqrt{2})$ and for circular polarization ($\epsilon=1$) is $\Delta\psi=\frac{1}{2}\pi$.

Fig. 6, and the relative phase $\Delta\psi$ between the $\mathcal{E}_{10}^{(-)}$ and $\mathcal{E}_{20}^{(-)*}$ is plotted against ϵ in Fig. 7, these quantities being evaluated at $\Omega_{\text{opt}}^{(-)}$.

To obtain a physical insight into these solutions, let us consider the special cases when $\epsilon=0$ and $\epsilon=1$ (linearly and circularly polarized laser, respectively). When $\epsilon=0$, the net propagation matrix $\Lambda_{\alpha\beta}$ (85) becomes

$$\begin{bmatrix} 2D & 0 & 2D & 0 \\ 0 & -3+\frac{3}{2}D & 0 & \frac{3}{2}D \\ -2D & 0 & -2D & 0 \\ 0 & \frac{3}{2}D & 0 & 3-\frac{3}{2}D \end{bmatrix} = [\Lambda_{\alpha\beta}(\epsilon=0)], \qquad (107)$$

the nonzero eigenvalues and eigenvectors of which are [cf. (93) and (103)]

$$\gamma^{(\pm)}=\pm\tfrac{3}{2}g_0(1-D)^{1/2}, \qquad (108)$$

$$(\mathcal{E}_\alpha^{(\pm)}(0))=\begin{bmatrix} 0 \\ 1\mp(1-D)^{-1/2} \\ 0 \\ -1\mp(1-D)^{-1/2} \end{bmatrix}. \qquad (109)$$

The gain of the gainy eigenroot is

$$g^{(-)}=-2\,\mathrm{Im}\gamma^{(-)}$$

$$=g_0\tfrac{3}{2}\sqrt{2}\left[\frac{\Omega\tau}{(1+\Omega^2\tau^2)^{\frac{1}{2}}}\left(1-\frac{\Omega\tau}{(1+\Omega^2\tau^2)^{\frac{1}{2}}}\right)\right]^{1/2}, \qquad (110)$$

which has a maximum at

$$\Omega_{\text{opt}}^{(-)}\tau=3^{-1/2} \qquad (111)$$

and a maximized value of

$$g^{(-)}(1/\sqrt{3})=g_0\tfrac{3}{4}\sqrt{2}. \qquad (112)$$

Note that this gain is even larger than the largest gain in the backward direction for $\epsilon=0$ [i.e., that of the major eigenmode $g^{(+)}=g_0$ in (57)]. However, the eigenpolarizations of these two cases are orthogonal; the forward-going polarization by inspection of (109) is perpendicular to that of the laser for both Stokes and anti-Stokes, whereas, the major backward-going polarization (56) is parallel. Comparison with the minor backward eigenmode, which yields perpendicular-linear scattering, shows the surprising result that the participation of the anti-Stokes channel *enhances* the gain in the forward direction for perpendicular-linear scattering [comparing (112) with $g^{(-)}=\frac{3}{4}g_0$ in (57)], whereas, the anti-Stokes channel completely suppresses the gain in the forward direction for the parallel-linear scattering[2] [comparing $g^{(+)}=g_0$ in (57) with $g^{(0)}=0$ with the eigenvector $(1,0,-1,0)^T$]. But by inspection of (109), one sees that there is a component of Stokes and anti-Stokes which is in phase with respect to each other, so that there is a constructive reinforcement of the cross coupling upon the self-coupling; the opposite effect occurs for the parallel-linear scattering.

The ratio of Stokes to anti-Stokes intensities for the gainy eigenmode of (109) at the optimum frequency shift is

$$R^{-1}=\frac{|\mathcal{E}_{1y}^{(-)}|^2}{|\mathcal{E}_{2y}^{*(-)}|^2}=5+6\sqrt{(\tfrac{2}{3})}\cong 9.89898\,,\qquad (\epsilon=0,\Omega_{\text{opt}}^{(-)}\tau=3^{-1/2}), \quad (113)$$

and the relative phase $\Delta\psi$ between $\mathcal{E}_{10}$ and $\mathcal{E}_{20}^*$ is[17]

$$\Delta\psi=\arctan(-2\sqrt{2})=1.91065 \text{ rad}. \quad (114)$$

In the case of circular polarization of the incident light ($\epsilon=1$), the net propagation matrix becomes

$$\begin{bmatrix} -3+\frac{7}{2}D & i(-3+\frac{5}{2}D) & \frac{1}{2}D & \frac{1}{2}iD \\ -i(-3+\frac{5}{2}D) & -3+\frac{7}{2}D & \frac{1}{2}iD & -\frac{1}{2}D \\ -\frac{1}{2}D & \frac{1}{2}iD & 3-\frac{7}{2}D & i(-3+\frac{5}{2}D) \\ \frac{1}{2}D & \frac{1}{2}D & -i(-3+\frac{5}{2}D) & 3-\frac{7}{2}D \end{bmatrix}=[\Lambda_{\alpha\beta}(\epsilon=1)], \quad (115)$$

the nonzero eigenvalues and eigenvectors of which are, by inspection [also cf. (93) and (103)],

$$\gamma^{(\pm)}=\pm 3g_0(1-D)\,, \quad (116)$$

$$(\mathcal{E}_\alpha^{(-)}(0))=\begin{bmatrix} i \\ 1 \\ 0 \\ 0 \end{bmatrix},\quad (\mathcal{E}_\alpha^{(+)}(0))=\begin{bmatrix} 0 \\ 0 \\ i \\ -1 \end{bmatrix}. \quad (117)$$

Note the absence of the anti-Stokes and Stokes components for the gainy and lossy roots, respectively. The gain of the gainy root is the same as for the major backward eigenmode (59a):

$$g^{(-)}=-2\,\text{Im}\gamma^{(-)}=6g_0[\Omega\tau/(1+\Omega^2\tau^2)], \quad (118)$$

with maximum occurring at $\Omega_{\text{opt}}^{(-)}\tau=1$ and with the value $g^{(-)}=3g_0$. Also the eigenpolarization is identical to that of the major backward case $[(1,-i)^T]$. However the minor backward eigenmode $[(1,i)^T]$ gets suppressed in the forward direction—$g^{(0)}=0$ for the eigenvector $(1,i,-1,i)^T$; this is to be contrasted with the linear-polarization case discussed above, where the *major* backward eigenmode gets suppressed. These results can be understood in terms of the absence and presence of the anti-Stokes channels for the counter-rotating and co-rotating eigenpolarizations, respectively, as has already been discussed in the Introduction. Note however, that $\text{Re}\gamma$ differs for the forward and backward counter-rotating eigenpolarization [cf. (116) and (59)]. But when $\epsilon=1$ (and $\delta_1=-1$), the over-all precession of (83b) and (49b) obviously becomes indistinguishable from retardation and combines with $\text{Re}\gamma$ to give the same total propagation constant for the forward and backward counter-rotating eigenpolarization

$$\kappa+\text{Re}\gamma^{(-)}{}_{\text{forward}}+\Gamma=3g_0[(1+\Omega^2\tau^2)^{-1}+\tfrac{1}{6}]$$
$$=\kappa+\text{Re}\gamma^{(+)}{}_{\text{backward}}-\Gamma. \quad (119)$$

Hence, not only does this eigenpolarization have the same gain, but also the same speed in the forward and backward directions due to the absence of the anti-Stokes channel.

Let us next consider the case of $\Omega=0$, where the Stokes and anti-Stokes merge with the incident wave (also called "degenerate four-photon scattering"):[13]

Whereas in backward scattering the case of $\Omega=0$ leads to birefringence when $\epsilon=0$, to optical activity when $\epsilon=1$, and to a combination of the two effects for the in-between values of ϵ; in forward scattering with $\Omega=0$, only optical activity occurs for all ϵ without any birefringent effects. When $\Omega=0$, $D=1$ [hence $\gamma^{(\pm)}=0$ in (93)] and $\Lambda_{\alpha\beta}$ becomes

$$\begin{bmatrix} 2-\frac{3}{2}\epsilon^2 & -\frac{1}{2}i\epsilon & 2-\frac{3}{2}\epsilon^2 & \frac{1}{2}i\epsilon \\ \frac{1}{2}i\epsilon & 2\epsilon^2-\frac{3}{2} & \frac{1}{2}i\epsilon & -2\epsilon^2+\frac{3}{2} \\ -2+\frac{3}{2}\epsilon^2 & \frac{1}{2}i\epsilon & -2+\frac{3}{2}\epsilon^2 & -\frac{1}{2}i\epsilon \\ \frac{1}{2}i\epsilon & 2\epsilon^2-\frac{3}{2} & \frac{1}{2}i\epsilon & -2\epsilon^2+\frac{3}{2} \end{bmatrix}$$
$$=[\Lambda_{\alpha\beta}(\Omega=0)], \quad (120)$$

which has two degenerate zero roots ($\Lambda_{\alpha\beta}$ is non-Hermitian) associated with two eigenpolarizations, which by inspection are

$$\gamma=0,\quad \gamma'=0, \quad (121)$$

$$(\mathcal{E}_\alpha(0))=\begin{bmatrix} z \\ 0 \\ -z \\ 0 \end{bmatrix}\rightarrow\begin{bmatrix} \text{Im}\,z \\ 0 \end{bmatrix},$$
$$(\mathcal{E}_\alpha'(0))=\begin{bmatrix} 0 \\ z' \\ 0 \\ z' \end{bmatrix}\rightarrow\begin{bmatrix} 0 \\ \text{Re}\,z' \end{bmatrix}, \quad (122)$$

for any z or z′. We must add $(\mathcal{E}_1+\mathcal{E}_2)$ to form 2-vectors from the 4-vectors, since the Stokes and anti-Stokes waves are indistinguishable. Clearly, any arbitrarily polarized weak wave with $\Omega=0$ can be expanded in terms of these eigenpolarizations. The complete solution (83b) then produces optical activity with a rotatory power $\frac{3}{2}\Gamma=3\epsilon g_0$ (sign determined by right-hand rule). (In the backward case, the rotatory power[18] is $+5g_0/4$

[17] $\Delta\psi$ represents the relative phase of the x components of the Stokes and anti-Stokes waves. Since, for $\epsilon=0$, these components are zero for the gainy root, $\Delta\psi$ is the limiting value for small ϵ. A more useful phase difference is that between the y components $\mathcal{E}_{1y}^{(-)}(0)$ and $\mathcal{E}_{2y}^{(-)*}(0)$, which is $\Delta\psi_y=\arctan\sqrt{2}=0.95527$ rad.

[18] Note that this effect is more similar to Faraday rotation than to natural optical activity in that reflecting a linearly polarized weak wave backward upon itself almost doubles the rotation, but differs from the Faraday effect in that the rotation is not exactly double.

for $\epsilon=1$.) In the particular limit $\epsilon=0$, no birefringence results. This fact may appear strange at first, since when the strong wave is linearly polarized, the molecules clearly must align themselves parallel to the field, and should produce birefringence upon a weak wave. This indeed is what happened for a backward-going weak wave, as has been discussed earlier, and for $\Omega\tau\gg1$, as we shall see presently. But in the forward direction with $\Omega=0$, the weak wave becomes indistinguishable from the strong wave and the vector sum of a linearly polarized strong and weak wave produces a slightly changed alignment direction for the molecules; the sum field propagates with a negligibly modified speed, whether the weak wave is polarized parallel or perpendicular to the strong wave. Indeed, for any ϵ of the strong wave, any arbitrarily polarized degenerate weak wave becomes part of the strong wave, so that the nonlinear solution (31) still applies to the vector addition of the strong and weak waves. Since $\mathcal{E}_1+\mathcal{E}_2\ll\mathcal{E}_0$, the alterations of Γ and κ are negligible, and the weak wave merely undergoes the strong wave's precession and retardation, which is in agreement with (121). Angular momentum conservation also forbids birefringence of the weak wave. Birefringence would imply angular momentum exchange with the strong wave; however, since the weak wave is indistinguishable from the strong wave, and the latter cannot exchange angular momentum with an isotropic medium ($\Omega=0$), no birefringence can occur.

Let us next consider the special case $\Omega=\infty$, or more precisely, $\Omega\tau\gg1$, in which case the molecular alignment fails to respond to the weak waves and $\chi_{ik}{}^{11,22,12,12}=0$ (i.e., there is neither self-coupling nor cross coupling). Then $D=0$, and

$$\begin{bmatrix} -3\epsilon^2 & -3i\epsilon & 0 & 0 \\ 3i\epsilon & -3 & 0 & 0 \\ 0 & 0 & 3\epsilon^2 & -3i\epsilon \\ 0 & 0 & 3i\epsilon & 3 \end{bmatrix} = [\Lambda_{\alpha\beta}(\Omega=\infty)], \quad (123)$$

the eigenvalues and eigenvectors of which, by inspection, are

$$\gamma^{(\pm)}=\pm\tfrac{3}{2}g_0(1+\epsilon^2), \quad \gamma^{(0)}=0,0; \quad (124)$$

$$(\mathcal{E}_\alpha{}^{(\pm)}(0))=\begin{bmatrix}0\\0\\i\epsilon\\-1\end{bmatrix}, \begin{bmatrix}i\epsilon\\1\\0\\0\end{bmatrix},$$

$$(\mathcal{E}_\alpha{}^{(0)}(0))=\begin{bmatrix}0\\0\\-1\\i\epsilon\end{bmatrix}, \begin{bmatrix}1\\i\epsilon\\0\\0\end{bmatrix}. \quad (125)$$

(Since $\Lambda_{\alpha\beta}$ is now Hermitian, four orthogonal eigenvectors exist.) These results are identical to those of the backward case (63), except for a reversal in the sense of the eigenpolarizations. From (124) we deduce birefringence when $\epsilon=0$, which is physically reasonable for light at all frequencies, except those nearer than $\Omega\lesssim\tau^{-1}$ to the incident frequency. Although (124) appears to predict optical activity when $\epsilon=1$, such is not the case because of the canceling effect of the strong-wave precession or optical activity. Equation (83b) gives for $\epsilon=1$, the same total propagation constant

$$\kappa+\gamma^{(0)}-\Gamma=\kappa+\gamma^{(-)}+\Gamma=\kappa-\gamma^{(+)}+\Gamma=\tfrac{1}{2}g_0$$

for all eigenpolarizations. (Note that these weak waves travel with the same speed as does the strong wave.) The lack of optical activity is also physically reasonable, since a circularly polarized strong wave aligns molecules randomly in the plane swept out by the rapidly rotating electric field, and these molecules cannot distinguish the sense of polarization of the weak wave. The amount of birefringence predicted for $\epsilon=0$ and the isotropic retardation predicted for $\epsilon=1$ are half the corresponding values for the low-frequency Kerr effect (i.e., $\omega_0\lesssim1/\tau$), which is understandable because the molecules, at such low frequencies, can follow the electric field and no time averaging takes place.

VI. CONCLUSIONS

The results of the calculations for the limits of linear and circular polarization of the incident light are summarized in Table III, which lists the ratios of gains and retardations for the various eigenmodes of propagation. These ratios summarize the effects of optical field-induced birefringence, optical activity, and linear and circular dichroism (or stimulated scattering) described in some detail in the previous sections of this paper. It should be emphasized that these ratios and indeed the other results obtained in these calculations apply not only to the molecular-orientation Kerr effect but also quite generally to *any* traceless tensor light-scattering process in an isotropic medium.[19]

We have not included in the present paper the effects of small-angle scattering; this extension of the present analysis involves a generalization in manner of Ref. 2 and will be published elsewhere. For parallel linear polarizations of incident and scattered light, an important feature of such near-forward scattering is the *elastic* light-by-light peak in the gain, which is twice the maximum value of the gain for the inelastic backward stimulated Rayleigh-wing scattering which occurs at $\Omega\tau=1$, but unlike the latter process, this light-by-light gain involves no frequency shift. Hence, in comparing the gains of the two eigenmodes of circular polarization,

[19] An example is stimulated rotational Raman effect (see Ref. 14). However, one must replace $D=(1+i\Omega\tau)^{-1}$ by $D=Qi/(1+2i\Omega\tau)$, where τ is the lifetime of the rotation, $\Omega=\omega_0-\omega_1-\omega_R$ is the detuning from resonance, and $Q=\omega_R\tau\gg1$. Also, there is very little self-precession and self-retardation in this case. Another example is the Kerr effect arising from clustering among spherical molecules discussed by R. W. Hellwarth, Phys. Rev. **152**, 156 (1966). However, in nonspherical cases one must consider nontraceless generalization of Eq. (7). Still another example is shear-wave Brillouin scattering; however, for the forward and backward scattering the gains are zero for kinematical reasons.

TABLE III. Ratio of gains and retardations of linear and circular eigenmodes. The gain is normalized with respect to incident intensity and expressed in units of $\frac{1}{4}g_0$. The weak-wave retardation (subtracting out the strong-wave retardation) is also normalized with respect to the incident intensity [see (53b)] and expressed in units of $\frac{1}{4}g_0$ (i.e., $\mathrm{Re}\gamma^{(\pm)} \mp \Gamma + \Gamma$ for the backward case and $\mathrm{Re}\gamma^{(0)}$, $\mathrm{Re}\gamma^{(-)} + 2\Gamma$ for the forward case). Incident linear polarization is assumed $\uparrow$ and circular polarization $\circlearrowleft$.

	$\Omega=0$ $\uparrow : \rightarrow : \circlearrowright : \circlearrowleft$	$\Omega=\Omega_{\mathrm{opt}}$ $\uparrow : \rightarrow : \circlearrowright : \circlearrowleft$	$\Omega=\infty$ $\uparrow : \rightarrow : \circlearrowright : \circlearrowleft$
Forward gain	$0:0:0:0$	$0:3\sqrt{2}^{\mathrm{a}}:6:0$	$0:0:0:0$
Backward gain	$0:0:0:0$	$4:3:6:1$	$0:0:0:0$
Forward retardation	$0:0:6:0$	$0:3\sqrt{\frac{3}{2}}^{\mathrm{a}}:3:0$	$0:-6:0:0$
Backward retardation	$4:-3:6:1$	$2:-\frac{9}{2}:3:\frac{1}{2}$	$0:-6:0:0$

[a] $\Omega_{\mathrm{opt}}\tau=1$ for all entries except these, for which $\Omega_{\mathrm{opt}}\tau=3^{-1/2}$.

one must take into account that the co-rotating–circular eigenpolarization also has such a light-by-light peak. This peak occurs in spite of the fact that in the exact forward direction this eigenmode has zero gain due to the suppression arising from the presence of the anti-Stokes channel. Indeed, this same channel gives rise to the elastic light-by-light peak, which has a maximum gain double that of the backward gain at $\Omega\tau=1$, or in terms of the units of Table III, $2(\frac{1}{4}g_0)$. However, this is still only a third of the gain at $\Omega\tau=1$ of the inelastic counter-rotating eigenmode, which we, therefore, expect to dominate in the experimental observations.

In the case of the two eigenmodes of linear polarization, on the other hand, the light-by-light peak causes the parallel-linear eigenmode (with no frequency shift) to dominate over the perpendicular-linear eigenmode (with $\Omega\tau=3^{-1/2}$), as becomes clear when we compare the maximum elastic gain of $8(\frac{1}{4}g_0)$ with the maximum inelastic gain of $3\sqrt{2}(\frac{1}{4}g_0)$. Hence, one infers that there must be an intermediate elliptical polarization at which the maximum elastic light-by-light gain crosses the maximum inelastic forward stimulated Rayleigh-wing gain. This crossover of the gains may explain the observation[20] that the self-focusing threshold for circularly polarized light is not a factor of 4 higher than that for linearly polarized light, as one expects on the basis of the ratio of Kerr coefficients (cf. entries in Table III for $\Omega=0$ and backward retardation) and also on the basis that the self-focusing threshold is inversely proportional to the peak light-by-light gain[2,21] for these two polarizations. A dominant counter-rotating scattered polarization will generate in combination with the incident light a linearly polarized component, which may affect the self-focusing threshold, and which may also explain the observation[20] that the light contained in the self-trapped filaments is linearly polarized, in spite of the fact that the incident light is circularly polarized.

Comparing the predictions of the present calculations with the observations of Foltz *et al.*[1] (cf. their Fig. 2) one has good qualitative agreement on the following points: (i) The frequency shift of the Rayleigh-wing scattering decreases upon going from circular to linear polarization (as one would expect from our Fig. 2); (ii) the intensity of the scattered line diminishes as $\epsilon \rightarrow 0$ (see our Fig. 3); (iii) the scattered line becomes increasingly polarized in the direction perpendicular to that of the laser as $\epsilon \rightarrow 0$, so that in the limit of linear polarization the scattered line shows up predominantly in the crossed-analyzer channel (see our Fig. 4); and (iv) there is never very much intensity in the anti-Stokes line—we expect at most only about 10%, which occurs with linear polarization (see our Fig. 6). However, more quantitative experimental work needs to be done using the two-cell amplifier method[22] to measure the gain as a function of the elliptical polarization of the laser, and measurements are also needed on the birefringence and optical activity predicted for an unshifted weak wave ($\Omega=0$).

Note added in manuscript. The third-order nonlinear susceptibility $\chi_{ilkm}^{(3)}$ is a fourth-rank tensor. In an isotropic medium, however, the most general form for such a tensor can be written

$$\chi_{ilkm}^{(3)}=\tfrac{1}{2}(a'+c')\delta_{il}\delta_{km} + \tfrac{1}{2}(a'-c')\delta_{im}\delta_{kl} + b'\delta_{ik}\delta_{lm}. \quad (126)$$

Therefore, the change in susceptibility for a static field E_i is

$$\Delta\chi_{ik}=a'E_iE_k+b'E_jE_j\delta_{ik}. \quad (127)$$

This is generalizable in the case of time-varying fields to the form given by Eq. (7). For tensor light scattering of the Kerr type,

$$a'=-3b'=K\tau. \quad (128)$$

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