Rotationally invariant theory of stimulated Raman scattering

R. Holmes and A. Flusberg*
Avco Research Laboratory, Incorporated, Everett, Massachusetts 02149
(Received 21 August 1987)

We present a detailed derivation of a new, rotationally invariant formalism for the theory of stimulated Raman scattering. The formalism is applied to Raman transitions of well-defined rotational symmetry, e.g., rotational Raman (*S*) transitions, yielding the explicit dependence of gain on light polarization, phase mismatch, and frequency offset. For a linearly polarized laser, the Stokes fields parallel and perpendicular to the laser field are decoupled. Far from Stokes–anti-Stokes phase matching, their gain coefficients are in the ratio of 4:3; at phase matching, both are suppressed and have zero exponential gain. For a circularly polarized laser, the circularly polarized Stokes fields circulating in the same and opposite senses as the laser field are decoupled. The gain of the opposite-sense field, which is decoupled from the anti-Stokes field, is independent of phase mismatch. Far from phase matching, the gain of the same-sense Stokes field is $\frac{1}{6}$ that of the opposite-sense field; at phase matching, the same-sense field has zero exponential gain. An unpolarized laser is shown to have the lowest Raman gain; far from phase matching, its Stokes fields decouple into two incoherent, oppositely circularly polarized fields, but at phase matching all Stokes polarizations have zero exponential gain. The gradual transition away from phase matching is also treated explicitly, and it is shown that when the gain is suppressed the maximum Stokes growth occurs when the Stokes-laser beat is a fraction of a Raman linewidth off resonance.

I. INTRODUCTION

The theory of stimulated Raman scattering (SRS) has undergone a great deal of development in the last two decades, ranging from the initial steady-state, monochromatic, ray-optics calculations for a dispersionless gas[1] to more extensive theories, including the transient response of the phonon,[2] the broadband laser,[3] wave-optics effects,[4] dispersion,[5] and quantum-electrodynamic effects.[6] An additional process which has been discussed in the literature is the effect of Stokes–anti-Stokes (SA) coupling and its dependence on the phase mismatch and frequency offset from the Raman resonance in the associated four-wave-mixing process in which two laser photons are simultaneously converted to a nearly copropagating Stokes and anti-Stokes photon.[7–9] The theory of higher-order Stokes and anti-Stokes radiation at phase matching has also been thoroughly discussed.[10] The polarization of laser light scattered spontaneously by gases has also been presented.[11] Polarization of the light generated by four-wave mixing has been discussed.[12] Also there are treatments of stimulated Raman scattering using the Hamiltonian approach with only pump and Stokes,[13] and including the anti-Stokes field.[14] Resonant and nonresonant effects have been treated.[15] However, no general, rotationally invariant theory of the polarization dependence of the Raman gain has been formulated. Such a theory is particularly crucial for the understanding of rotational SRS, in which two units of angular momentum are transferred to the molecules of the medium. The transfer of angular momentum implies, for example, that the four-wave mixing responsible for parametric gain suppression[7,8] disappears when the (pump) laser and Stokes light copropagate and are circularly polarized in opposite senses.[16]

This paper describes in detail a new, rotationally invariant formalism for the Raman equations, which was only briefly summarized by us previously.[17] The starting equations are the standard Hamiltonian given by Wang[13] and the Lagrangian density given by Shen and Bloembergen,[7] except that we take into account from the start that the interaction Hamiltonian must be invariant under rotation if the medium is isotropic.

We derive the polarization dependence of the Raman gain for plane waves. The eigenvalue spectrum is obtained by considering the growth of the phonon amplitude along the axis of propagation of the pump light. The corresponding eigenmodes of the phonon and the eigenpolarizations of the field are then obtained. In general, each eigenvalue is determined by the ratio of the laser intensity to the phase mismatch, the offset from the Raman resonance, and the polarization state or coherency matrix of the pump laser. We calculate the eigenvalues of the gain for linearly, circularly, elliptically, and partially polarized lasers. Although there have been notable treatments of polarization in stimulated Raman scattering in the past,[18] to our knowledge this is the first SRS formalism which is general enough to treat pump light of *arbitrary polarization,* including partial polarization.

In general, there are three terms of three different symmetries under rotation which contribute to the Raman effect: the "trace" (or scalar, $J=0$) term, invariant under rotation; the "magnetic-dipole" ($J=1$) term, which transforms like a magnetic dipole (vector); and the "electric-quadrupole" ($J=2$) term which transforms like a second-rank tensor. Each term may make a contribution $g_0^{(J)}$ to the gain.[19] With no loss of generality, we will treat the case in which only a single coefficient $g_0^{(J)}$ is nonzero. This will be exactly true for S (rotational) transitions ($J=2$) and approximately true for many vibrational transitions ($J=0$) as well.

II. CONVENTIONS

The following discussions involve the electromagnetic field vectors and the phonon amplitudes. The following conventions are utilized.

Let z be the direction of propagation of the light. The complex optical electric field $\mathbf{E}_C^T$ may then be decomposed into circular components as

$$\mathbf{E}_C^T = E_+\hat{\boldsymbol{\varepsilon}}_+ + E_-\hat{\boldsymbol{\varepsilon}}_- ,$$

where E_+ (E_-), the complex amplitude of right-handed (left-handed) polarization, may be expanded in terms of spectral components as

$$E_\pm = \sum_j E_{j\pm} = \sum_j E_{j\pm}^0 \exp[i(k_j z - \omega_j t)] .$$

Here the sum is over all the spectral components of both the laser and Stokes fields. The polarization unit vectors are given by

$$\hat{\boldsymbol{\varepsilon}}_\pm = (\hat{\mathbf{x}} \pm i\hat{\mathbf{y}})/\sqrt{2} ,$$

where $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ are unit vectors along x and y, respectively. Hence,

$$|E_C^T|^2 = |E_+|^2 + |E_-|^2 .$$

To obtain the corresponding real electric fields $\mathbf{E}_r^T$, $\mathbf{E}_{+,r}$, and $\mathbf{E}_{-,r}$ the complex conjugates must be added:

$$\mathbf{E}_{+,r} = (1/\sqrt{2})(E_+\hat{\boldsymbol{\varepsilon}}_+ + E_+^*\hat{\boldsymbol{\varepsilon}}_-) ,$$

$$\mathbf{E}_{-,r} = (1/\sqrt{2})(E_-\hat{\boldsymbol{\varepsilon}}_- + E_-^*\hat{\boldsymbol{\varepsilon}}_+) .$$

Hence,

$$\mathbf{E}_{+,r}\cdot\mathbf{E}_{+,r} = |E_+|^2 ,$$

$$\mathbf{E}_{-,r}\cdot\mathbf{E}_{-,r} = |E_-|^2 ,$$

$$\mathbf{E}_r^T = \mathbf{E}_{+,r} + \mathbf{E}_{-,r} ,$$

$$\mathbf{E}_r^T\cdot\mathbf{E}_r^T = |E_+|^2 + |E_-|^2 + \tfrac{1}{2}(E_+E_- + \text{c.c.}) .$$

Since $(E_+E_- + \text{c.c.})$ oscillates at twice the optical frequency, it does not contribute to the time-averaged intensity $\langle (E_r^T)^2 \rangle$,

$$\langle \mathbf{E}_r^T\cdot\mathbf{E}_r^T \rangle = \langle |E_+|^2 \rangle + \langle |E_-|^2 \rangle .$$

Now, the basis vectors $\hat{\boldsymbol{\varepsilon}}_+$ and $\hat{\boldsymbol{\varepsilon}}_-$ are $(\hat{\mathbf{x}},\hat{\mathbf{y}})$ representations of photons of ± 1 unit of angular momentum along the axis of propagation. Hence, an alternative expression which explicitly exhibits the transformation properties of the fields under a rotation of axes is obtained by rewriting $\hat{\boldsymbol{\varepsilon}}_\pm$ as a vector ket, $\hat{\boldsymbol{\varepsilon}}_\alpha = |j\alpha\rangle$ with $j=1$, so that

$$\mathbf{E}_{\alpha,r} = (1/\sqrt{2})(E_\alpha |1\alpha\rangle + E_\alpha^* |1-\alpha\rangle) , \tag{1}$$

$$\mathbf{E}_r^T = \sum_{\alpha=\pm 1} \mathbf{E}_{\alpha,r} . \tag{2}$$

These fields will be used henceforth; thus, the subscript r will be dropped. A similar convention will be used below to write the real phonon amplitude Q_m^T in a form that explicitly exhibits that the phonon transforms under rotation like a rank-J spherical tensor,

$$\tilde{Q}_m^T = (1/\sqrt{2})(Q_m \langle Jm| + Q_m^* \langle J-m|) . \tag{3}$$

III. ROTATIONALLY INVARIANT EQUATIONS

The procedure to derive the equations of motion utilizes the Lagrangian density and an interaction Hamiltonian following Wang.[13] The energy of the interaction is given by

$$H_{\text{int}} = \sum_i \mathbf{P}_i\cdot\mathbf{E}_i , \tag{4}$$

where $\mathbf{P}_i$ is the induced polarization at the ith molecule and $\mathbf{E}_i$ is the electric field of electromagnetic wave at the ith molecule.

The form of the above Hamiltonian for a Raman interaction, without the linear material response, is[13]

$$H_{\text{int}} = \sum_i (\partial\alpha/\partial q)_{m\alpha\beta}^{ijk} q_{im} E_{j\alpha} E_{k\beta} , \tag{5}$$

where $\partial\alpha/\partial q$ is the differential polarizability at optical frequency ω_k and polarization β caused by excitation of a transition m at molecule i, and by light at frequency ω_j and polarization α; q_{im} is the excitation amplitude of transition m at the ith molecule; and $E_{j\alpha}$ is the amplitude of the light field at frequency ω_j and polarization α.

We assume that the differential polarizability is approximately independent of the exciting modes j and k. This assumption is equivalent to neglecting the dispersion in the susceptibility. We also assume that the medium is isotropic and consists of a single species of molecules of density N. Thus, one may write[13]

$$Q_m = N^{1/2} \langle q_{im} \rangle , \tag{6}$$

where $\langle q_{im} \rangle$ is the expectation value of q_{im}.

A further assumption is that the susceptibility itself, for a given molecule and rotational state, is independent of rotational orientation and light polarization, Hence, the interaction Hamiltonian may be written

$$H_{\text{int}} = N(\partial\alpha/\partial Q) \sum_m \tilde{Q}_m^T : \mathbf{E}^T\mathbf{E}^T , \tag{7}$$

where $\tilde{Q}_m^T$ and $\mathbf{E}^T$ are given by Eqs. (1)–(3). Note that our starting assumption, that the interaction Hamiltonian is invariant under rotation, taken together with the implication of Eq. (3) that the phonon amplitude transforms like a spherical tensor of rank J, implies immediately that only sums of bilinear combinations of the field which transform like a rank-J tensor can appear in H_{int}. This will be seen explicitly below.

Upon combining the bras of Q and the kets of E, one has the following expanded expression for the interaction term H_{int}:

$$H_{\rm int}=(N/2^{3/2})(\partial\alpha/\partial Q)\sum_{m,\alpha,\beta}Q_mE_\alpha E_\beta\langle J\,m\mid 1\,1\,\alpha\beta\rangle+2Q_mE_\alpha E^*_\beta\langle J\,m\mid 1\,1\alpha\,-\beta\rangle+Q_mE^*_\alpha E^*_\beta\langle J\,m\mid 1\,1\,-\alpha\,-\beta\rangle$$
$$+Q^*_mE_\alpha E_\beta\langle J\,-m\mid 1\,1\,\alpha\beta\rangle+2Q^*_mE_\alpha E^*_\beta\langle J\,-m\mid 1\,1\,\alpha\,-\beta\rangle+Q^*_mE^*_\alpha E^*_\beta\langle J\,-m\mid 1\,1\,-\alpha\,-\beta\rangle\ . \quad (8)$$

Had we not assumed the form of Eq. (3) for the phonon, the most general form, which would have included a superposition of tensors of ranks 0, 1, and 2, would have yielded instead a sum of terms with different values of J. This more general case is beyond the scope of this paper.

With the above Hamiltonian, one then has the interaction term for the Lagrangian density. The Lagrangian density L may be written

$$L=L_{\rm em}+L_{\rm mat}+L_{\rm int}\ ,$$

where

$$L_{\rm em}=[(E^T)^2-(B^T)^2]/8\pi\ ,$$

B^T is the magnetic field strength,

$$L_{\rm mat}=\tfrac{1}{2}\sum_m[(\dot Q^T_m)^2-\omega_0^2(Q^T_m)^2]\ ,$$

ω_0 is the Raman resonance frequency, and $L_{\rm int}=H_{\rm int}$.

Now take the variation of the Lagrangian with respect to E^T_γ and Q^T_n. One obtains the following set of equations:

$$\Box^2E_\gamma=(4\pi/c^2)\partial^2/\partial t^2(\partial L_{\rm int}/\partial E^*_\gamma)\ , \quad (9)$$

$$(\partial^2/\partial t^2+\omega_0^2+2\Gamma\partial/\partial t)Q_n=\partial L_{\rm int}/\partial Q^*_n\ . \quad (10)$$

In Eq. (9) the D'Alembertian notation is used. In Eq. (10) the phenomenological damping constant Γ has been introduced in the usual way. To complete these expressions, one must evaluate the right-hand sides. To do this one must take the derivatives with respect to the (complex) quantities $Q^*_m=q_1-iq_2$ and $E^*_\gamma=e_1-ie_2$. These derivatives may be expressed as follows:

$$\frac{\partial}{\partial Q^*_n}=\frac{\partial}{\partial q_1}\frac{\partial q_1}{\partial Q^*_n}+\frac{\partial}{\partial q_2}\frac{\partial q_2}{\partial Q^*_n}=\frac{1}{2}\left[\frac{\partial}{\partial q_1}+i\frac{\partial}{\partial q_2}\right]\ ,$$

$$\frac{\partial}{\partial E^*_\gamma}=\frac{1}{2}\left[\frac{\partial}{\partial e_1}+i\frac{\partial}{\partial e_2}\right]\ .$$

Upon performing the algebra, one has

$$\partial L_{\rm int}/\partial E^*_\gamma=N/\sqrt{2}(\partial\alpha/\partial Q)\left[\sum_{m,\alpha}Q_mE_\alpha\langle J\,m\mid 1\,1\,\alpha-\gamma\rangle+Q^*_mE_\alpha\langle J\,-m\mid 1\,1\alpha\,-\gamma\rangle\right.$$
$$\left.+Q_mE^*_\alpha\langle J\,m\mid 1\,1\,-\alpha\,-\gamma\rangle+Q^*_mE^*_\alpha\langle J\,-m\mid 1\,1\,-\alpha\,-\gamma\rangle\right]\ , \quad (11)$$

$$\partial L_{\rm int}/\partial Q^*_n=(N/\sqrt{2^3})(\partial\alpha/\partial Q)\left[\sum_{\alpha,\beta}2E_\alpha E^*_\beta\langle J\,-n\mid 1\,1\alpha\,-\beta\rangle+E_\alpha E_\beta\langle J\,-n\mid 1\,1\alpha\beta\rangle\right.$$
$$\left.+E^*_\alpha E^*_\beta\langle J\,-n\mid 1\,1\,-\alpha\,-\beta\rangle\right]\ . \quad (12)$$

Now the temporal frequencies of the right-hand sides of Eqs. (9) and (10) must equal the temporal frequencies of the respective left-hand sides. The temporal frequency of a specified component j of E_α is ω_j and the temporal components of Q are $\omega_r=\omega_0+\Delta\omega$, the Raman frequencies. Note that the optical frequency ω_j of a component of E_α is always greater than zero for the forward-propagating case considered here and that $\omega_j>\omega_r$ for Raman scattering. Thus the last two terms of the sums in both Eqs. (11) and (12) may be eliminated by invoking the frequency-matching condition. The first terms also simplify considerably, and one arrives at the following equations:

$$\Box^2E_{j\gamma}=-(4\pi\omega_j^2/c^2)(N/\sqrt{2})(\partial\alpha/\partial Q)$$
$$\times\left[\sum_{\Delta\omega}\sum_{m,\alpha}\langle J\,m\mid 1\,1\,\alpha\,-\gamma\rangle Q_mE_{jr-,\alpha}\right.$$
$$\left.+\langle J\,-m\mid 1\,1\alpha\,-\gamma\rangle Q^*_mE_{jr+,\alpha}\right]\ , \quad (13)$$

$$(\partial^2/\partial t^2+\omega_0^2+2\Gamma\partial/\partial t)Q_n$$
$$=(N/\sqrt{2})(\partial\alpha/\partial Q)\sum_{\Delta\omega}\sum_{\alpha,\beta,k}\langle J\,-n\mid 1\,1\alpha\,-\beta\rangle$$
$$\times E_{kr+,\alpha}E^*_{k\beta}\ , \quad (14)$$

where $E_{j\alpha}$ is the jth mode (frequency ω_j) of polarization α, $jr+$ is a mode index corresponding to the "upshifted" frequency $\omega_j+\omega_0+\Delta\omega$, and $jr-$ is a mode index corresponding to the "downshifted" frequency $\omega_j-\omega_0-\Delta\omega$. Equations (13) and (14) may be expressed in a somewhat more familiar form using the slowly varying envelope approximation. The usual expression relating the envelopes to their fields is as follows:

$$Q_n=q_n\exp(i\omega_rt)\ ,$$

$$E_{j\alpha}=E^0_{j\alpha}\exp[i(k_jz-\omega_jt)]\ .$$

Then one may drop the second derivatives with respect to time and propagation path in this approximation. Also one may apply the standard assumption that the Raman frequency ω_r is approximately the Raman resonance frequency ω_0, i.e., the Raman linewidth Γ is much less than the Raman resonance frequency ω_0. A further assumption is that the axial modes of the electromagnetic fields are far apart compared to the Raman linewidth so that only modes separated by precisely ω_r combine to contribute to the phonon amplitude. With these assumptions one may derive the following result:

$$\left[\frac{\partial}{\partial z}+\frac{1}{v_j}\frac{\partial}{\partial t}+(2ik_j)^{-1}\nabla_\perp^2\right]E^0_{j\gamma}=\frac{2\pi i k_j}{n_j^2}(N/\sqrt{2})(\partial\alpha/\partial Q)\left[\sum_{m,\alpha}\langle J\,m\mid 1\,1\,\alpha\,-\gamma\rangle q_m E^0_{jr-,\alpha}\exp(i\Delta k_{j-}z)\right.$$
$$\left.+\langle J\,-m\mid 1\,1\,\alpha\,-\gamma\rangle q_m^* E^0_{jr+,\alpha}\exp(i\Delta k_{j+}z)\right], \quad (13')$$

$$\left[\frac{\partial}{\partial t}+(\Gamma-i\Delta\omega)\right]q_n=\frac{1}{2i\omega_r}(N/\sqrt{2})(\partial\alpha/\partial Q)\left[\sum_{\alpha,\beta,k}\langle J\,-n\mid 1\,1\,\alpha\,-\beta\rangle E^0_{kr+,\alpha}E^{0*}_{k\beta}\exp(i\Delta k_{k+}z)\right], \quad (14')$$

where $v_j=\omega_j/k_j$, $\nabla_\perp^2$ is the transverse Laplacian operator, n_j is the refractive index at optical frequency ω_j, $\Delta k_{j+}=k_{jr+}-k_j$ is the wave-number difference of light differing in frequency by the Raman frequency ω_r, $\Delta k_{j-}=k_{jr-}-k_j$, and $\Delta\omega=\omega_r-\omega_0$ is the difference between the Raman frequency and the Raman resonance frequency.

Equations (13′) and (14′) are the frequency-matched expressions for the slowly varying envelopes of the electric fields and the optical phonons. They are the rotationally invariant generalization of the multiwave Raman equations presented by Armstrong *et al.*[20] and discussed by Ackerhalt.[21] These equations include the effect of energy-level degeneracy presented by Zabolotskii *et al.*[22]

IV. RESULTS

One may observe how the above expressions reduce to those previously given in the literature for pump and Stokes radiation only, in the steady-state, for various symmetries. Under these assumptions the equation of the Stokes envelope $E_{s\alpha}$ reduces to

$$\left[\frac{\partial}{\partial z}+\frac{1}{v_s}\frac{\partial}{\partial t}+(2ik_s)^{-1}\nabla_\perp^2\right]E_{s\gamma}=\frac{\pi k_s(N\partial\alpha/\partial Q)^2}{2n_s^2\omega_r(1-i\Delta\omega/\Gamma)}\sum_{m,\alpha,\beta,\mu}\langle J\,-m\mid 1\,1\,\alpha\,-\gamma\rangle\langle J\,-m\mid 1\,1\,\beta\,-\mu\rangle E^*_{p\alpha}E_{p\beta}E_{s\mu}, \quad (15)$$

where all subscripts s pertain to the Stokes frequency and $E_{p\alpha}$ is the pump envelope of frequency component ω_p equal to $\omega_s+\omega_r$.

With the above expression, one may identify the Stokes amplitude gain,

$$g_s(\omega_r)=\frac{\pi k_s[N(\partial\alpha/\partial Q)]^2}{2n_s^2\omega_r\Gamma[1+(\Delta\omega/\Gamma)^2]}. \quad (16)$$

When one notes that by definition over photon amplitude Q is a factor of $\sqrt{2}$ smaller than that of Wang, this expression becomes identical to his.[13] However, we now find that the Stokes gain also depends on the polarization of the pump and Stokes radiation. To pursue this polarization dependence further, it will be useful to consider Stokes and pump radiations which are plane waves, and to perform the calculation in the retarded frame, with the assumption that the group velocities v_p and v_s of the pump and Stokes radiation are equal. It is convenient to study the polarization dependence using the phonon amplitudes and their growth in the direction of propagation of the pump radiation. This will then determine the growth of the Stokes radiation, as may be seen from Eq. (14) or (14′). To account properly for the parametric gain suppression as a result of the SA coupling,[7] the anti-Stokes field A will be included with the pump field P and the Stokes field S. Furthermore, spectrally broad pump light is of interest so an arbitrary number of longitudinal modes will be included.

In order to simplify the notation used in the equations, one may define a reduced phonon $R_m^{(J)}$, proportional to the slowly varying phonon amplitudes q_{-m},

$$R_m^{(J)}=q_{-m}\exp[i(k_p-k_s)z]\times[2^{3/2}\omega_r(\Gamma-i\Delta\omega)/N(\partial\alpha/\partial Q)]. \quad (17a)$$

R may then be expanded in spherical tensors of rank J and various components m $(-J\le m\le +J)$ as follows:[17]

$$R_m^{(J)}=\sum_{\alpha,\beta,n}\langle J\,m\mid 1\,1\,\alpha\,-\beta\rangle(P_{\alpha,n}S^*_{\beta,n}+A_{\alpha,n}P^*_{\beta,n}e^{-i\Delta\mathbf{k}\cdot\mathbf{z}}), \quad (17b)$$

where $\langle J\,m\mid 1\,1\,\alpha\beta\rangle$ is a Clebsch-Gordan coefficient, $F_{\alpha,n}$ is a spherical component α of the amplitude of axial mode n of field F $(=A,P,S)$, and $\Delta\mathbf{k}$ is the phase mismatch given by $2\mathbf{k}_p - \mathbf{k}_A - \mathbf{k}_S$. The form of Eq. (17) is intuitively obvious: The field amplitudes are rank-one spherical tensors which must be combined as indicated in Eq. (17b) to form spherical tensors of rank J so that both sides of Eq. (17b) transform in the same way under an arbitrary rotation of coordinate axes. Let the (plane) waves propagate along slightly different directions centered on z so that $\Delta\mathbf{k}$ lies primarily along the z direction. The equations for the spatial evolution of the waves are then

$$\partial_z S_{\beta,n} = g_s^{(J)} \sum_{\mu,M} \langle 1\beta \mid 1 J \mu -M \rangle P_{\mu,n} R_M^{(J)*} ,$$
$$\partial_z A_{\alpha,n} = -g_s^{(J)*}(k_A/k_S) \sum_{\mu,M} \langle 1\alpha \mid 1 J \mu M \rangle P_{\mu,n} R_M^{(J)} e^{i\Delta k z}. \tag{18}$$

A sum over J on the right-hand sides of Eqs. (18) gives the general case where $g_s^{(J)}$ is a Stokes amplitude gain similar to Eq. (16):

$$g_s^{(J)} = \frac{\pi k_s [N(\partial\alpha/\partial Q)^{(J)}]^2 N^{(J)}}{2n_s^2 \omega_r \Gamma(1+i\lambda)} . \tag{19}$$

Here λ is the normalized frequency offset $(\omega_r - \omega_0)/\Gamma$ and $N^{(J)}$ is a normalization which arises from the Clebsch-Gordan coefficients; they may be calculated by inspection:

$$N^{(0)}=3, \quad N^{(1)}=2, \quad N^{(2)}=\sqrt{5/3} .$$

To find the eigenvalues of Eqs. (17) and (18) in the small-signal limit of no significant pump-light depletion, we differentiate Eqs. (17) with respect to z and substitute Eqs. (18) into the resulting equation. We obtain

$$\partial_z R_m^{(J)} = g^{(J)*} \sum_M (D_{mM}^{(J)} + B_{mM}^{(J)}) R_M^{(J)} - i\Delta k \sum_{\alpha,\beta,n} h_{m\alpha\beta}^{(J)} A_{\alpha,n} P_{\beta,n}^* e^{-i\Delta k z} , \tag{20}$$

where

$$D_{mM}^{(J)} = \sum_{\alpha,\mu} d_{m\alpha\mu M}^{(J)} C_{\alpha\mu} ,$$
$$B_{mM}^{(J)} = -(k_A/k_S) \sum_{\alpha,\mu} b_{m\alpha\mu M}^{(J)} C_{\alpha\mu} , \tag{21}$$

$$d_{m\alpha\mu M}^{(J)} = \sum_{\beta} \langle J m \mid 1 1 \alpha -\beta \rangle \langle 1\beta \mid 1 J \mu -M \rangle ,$$
$$b_{m\alpha\mu M}^{(J)} = \sum_{\beta} \langle J m \mid 1 1 \beta -\alpha \rangle \langle 1\beta \mid 1 J \mu M \rangle , \tag{22}$$

$$h_{m\alpha\beta}^{(J)} = \langle J m \mid 1 1 \alpha -\beta \rangle , \tag{23}$$

and $C_{\alpha\mu}$ is the coherency matrix of the pump laser:

$$C_{\alpha\mu} = \sum_n P_{\alpha,n} P_{\mu,n}^* . \tag{24}$$

Differentiating Eq. (20) with respect to z and substituting Eq. (18) into the resulting equation yields[17]

$$\partial_z^2 R = g^*(D+B)\partial_z R - i\Delta k[\partial_z R - g^*(D+B)R] - i\Delta k g^* B R , \tag{25}$$

where R is now understood to be a column vector with $2J+1$ elements given by Eqs. (17), while D and B are matrices with matrix elements given by Eqs. (21); we have suppressed the superscripts and subscripts to simplify the notation. Equation (25), which determines the phonon growth for arbitrary phase mismatch, is the primary result of this paper.

Following Ref. 17, one obtains the eigenvalues and eigenvectors of Eq. (25) by substituting

$$R = R_0 e^{u g_0 I_0 z} \tag{26}$$

so that u represents the complex eigenvalue, normalized to $g_0 I_0$ (I_0 is the laser intensity); the real part of u is proportional to the gain. Since Eq. (25) is homogeneous in R, the roots u must satisfy

$$\det[u(u+i\kappa)I - u(D+B)\eta - i\eta\kappa D] = 0 , \tag{27}$$

where I is the identity matrix, $\eta = g^*/g_0 I_0$, and $\kappa = \Delta k / g_0 I_0$ is the phase mismatch factor.

Before discussing the roots u in the general case, we investigate the limits of both small and large phase mismatch. According to Eq. (27), the eigenvalues u in the phase-matched limit ($|\kappa| \ll 1$) are obtained by diagonalizing the matrix $D+B$. On the other hand, far from phase matching ($|\kappa| \gg 1$) we find instead that the eigenvalues are obtained by diagonalizing D; this may also be seen directly from Eqs. (17) and (18) when the terms in those equations proportional to the anti-Stokes field are neglected.

Let us now consider the possible values of J. The $J=0$, $J=1$, and $J=2$ phonons correspond to scattering with isotropic, magnetic-dipole, and electric-quadrupole rotational symmetry, respectively, as described by Placzek.[19] All three types of scattering may contribute to the electronic Raman effect. For a $Q(0)$ vibrational transition the selection rules allow only $J=0$, i.e., the phonons carry no angular momentum. In this case, as we will show below, the eigenvalues of the (scalar) phonon are just those calculated by Shen and Bloembergen[7] and the Stokes polarization is identical to that of the pump. Far from an electronic resonance, the $J=1$ scattering is negligible for both the vibrational and rotational Raman effects.[19] Although $Q(j)$ vibrational transitions ($j \neq 0$) may in general contain elements of both $J=0$ and $J=2$, $J=0$ often dominates (e.g., for diatomic molecules). For all pure rotational (S) transitions the selection rules allow only $J=2$. After a brief discussion of the $J=0$ case, we will focus our attention on the case of electric-quadrupole scattering ($J=2$).

For $J=0$, the phonon and fields may be written, using Eqs. (17) and (18) in the small-signal limit, as follows:

$$R_0=\langle 00\,|\,11\,1-1\rangle[P_+S_+^* +P_-S_-^* +\exp(-i\Delta kz)(A_+P_+^*+A_-P_-^*)]\ , \tag{28a}$$

$$\frac{\partial}{\partial z}S_\pm = g_s^{(0)}\langle 1\pm1\,|\,10\pm10\rangle P_\pm R_0^*\ , \tag{28b}$$

$$\frac{\partial}{\partial z}A_\pm = -g_s^{(0)*}(k_A/k_S)\langle 1\pm1\,|\,10\pm10\rangle P_\pm R_0\exp(i\Delta kz)\ , \tag{28c}$$

$$\frac{\partial}{\partial z}P_\pm = 0\ . \tag{28d}$$

It is evident from Eqs. (28b) and (28c) that both S and A have the same polarization as the pump light. With this in mind, the scalar equation for the components of S and A parallel to the pump light are sufficient to describe their evolution. Hence, evaluating the Clebsch-Gordan coefficients and simplifying, one has

$$\frac{\partial}{\partial z}S=\frac{g_s^{(0)}}{3}[\,|P|^2S+\exp(i\Delta kz)P^2A^*]\ , \tag{29a}$$

$$\frac{\partial}{\partial z}A=-\frac{g_s^{(0)*}}{3}(k_A/k_S)[\,|P|^2A+\exp(i\Delta kz)P^2S^*]\ . \tag{29b}$$

Accounting for the normalization chosen in the definition of $g_s^{(0)}$ in Eq. (19), these are the well-known equations for scalar-field SA coupling.[7,8]

We now return to the more complicated case of electric-quadrupole scattering. At the phase-matching angle we find that for quantization along z the matrix $D+B$ is already diagonal; Eq. (20) reduces to

$$\partial_z R_m^{(2)}=(mg_0^{(2)}/2)(1+i\lambda)^{-1}(C_{++}-C_{--})R_m^{(2)}\ , \tag{30}$$

where $m=0,\pm2$. Thus for pump light which is right circularly polarized $(+)$, the full gain occurs at the phase-matching angle with the Stokes radiation left circularly polarized $(-)$. (Note that $g_0^{(2)}$ is the resonance Stokes-amplitude gain for the $+\rightarrow-$ transition.) The $+\rightarrow+$ transition $(m=0)$ has zero gain, however, because of parametric gain suppression associated with the allowed SA coupling. For light with a coherency matrix which corresponds to equal amounts of left- and right-circular polarization, e.g., linearly polarized or unpolarized light, the gain is zero for *all* the phonons, i.e., we have complete parametric gain suppression at phase matching. It is apparent that the gain varies continuously between its high value and 0 as the polarization is varied between these two limits, e.g., for either elliptically or partially polarized light.

Far from phase matching we must diagonalize D. Consider the case $\lambda=0$, which leads to the highest gain. We find that for circularly polarized $(+)$ pump light the three eigenvalues are $u=1$, $\frac{1}{6}$, and 0; the first two eigenvalues correspond to circularly polarized $(-)$ and $(+)$ Stokes light, respectively. For linearly polarized light, say along x, we find instead $u=\frac{2}{3}$, $\frac{1}{2}$, and 0; now the first two eigenvalues correspond to Stokes light linearly polarized along x and y, respectively. As expected, these are precisely the relative gains which would be predicted from the ratios of the spontaneous rotational Raman cross sections.[19] For unpolarized light we obtain $u=\frac{1}{2}$, $\frac{1}{2}$, and $\frac{1}{6}$. Evidently, far from phase matching, unpolarized light may be though of as being "decomposed" by the rotational Raman effect into two mutually incoherent *circularly* polarized components, each with half the total intensity; this yields the larger eigenvalues of $\frac{1}{2}$. Off resonance (i.e., $\lambda\neq0$) all the above results apply except that each eigenvalue is multiplied by a factor of $(1+i\lambda)^{-1}$.

We return to the general solution of Eq. (27) for arbitrary values of κ. In Fig. 1 we have plotted the normalized gain $\mathrm{Re}(u)$ for a linearly polarized, circularly polarized, and unpolarized pump laser, all on Raman resonance $(\lambda=0)$ and with $\omega_r \ll \omega_p$. The positive (negative) branches are predominantly Stokes (anti-Stokes) roots when $\kappa\neq0$, and correspond to amplification (attenuation). The plots show that the transition between the phase-matched an unmatched regimes occurs around $\kappa=1$, as expected. Note that no root crossings occur except at $\kappa=0$; thus, for a linearly (x) polarized pump laser the dominant root corresponds to parallel (x) Stokes polarization for all values of $\kappa\neq0$. The plot for an unpolarized pump shows the maximum gain to be smaller than for a linearly polarized pump throughout; indeed, the larger unpolarized-pump eigenvalue is identical throughout with that for $x\rightarrow y$, while the smaller is identical with $+\rightarrow+$. We remark that in view of Eqs. (13′) and (14′) an unpolarized multimode laser may be viewed as a type of single-mode laser whose polarization is modulated on a short time scale compared to the phonon lifetime Γ^{-1}; our result should apply to any such arbitrary modulation.

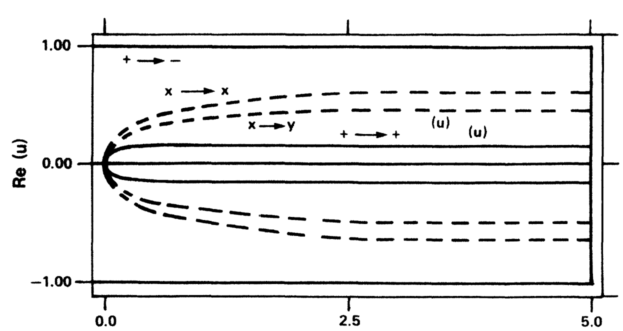

FIG. 1. Normalized gain $\mathrm{Re}(u)$ vs phase-mismatch factor κ calculated from Eq. (12) for $\lambda=0$. Solid (dashed) curves; circularly (linearly) polarized pump light, with sample polarizations given for each. Unpolarized-pump solutions (u) coincide with those for $x\rightarrow y$ and $+\rightarrow+$, as shown.

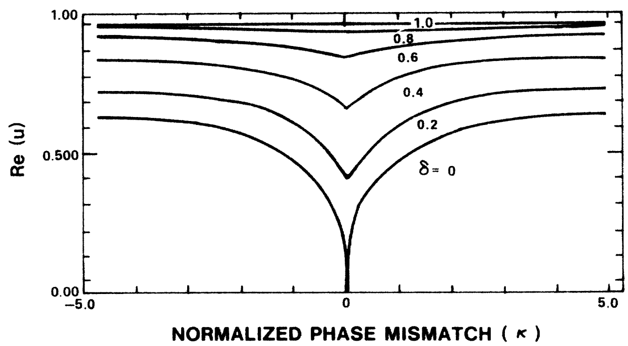

FIG. 2. Maximum normalized resonant gain vs κ for various pump polarization ellipticities. The quantity δ indicates the ratio of the minor to the major axis of the polarization ellipse.

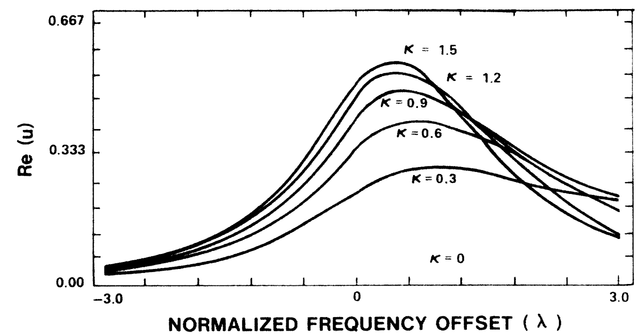

FIG. 5. Maximum real part of the six normalized roots for linearly polarized light. Non-negative momentum mismatches κ are shown. Anti-Stokes–Stokes ratio equals 1.22.

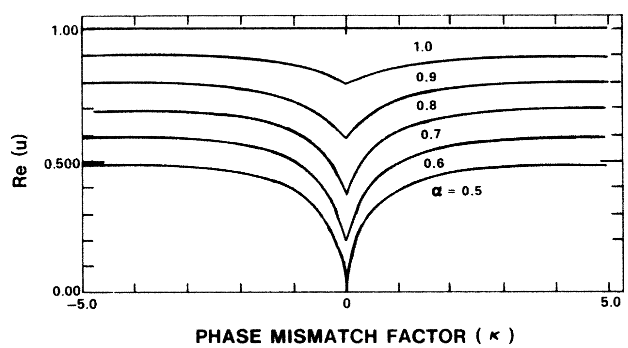

FIG. 3. Maximum normalized resonant gain vs κ for various degrees of circular polarization. The parameter α is given by $\alpha = C_{++}/(C_{++} + C_{--})$.

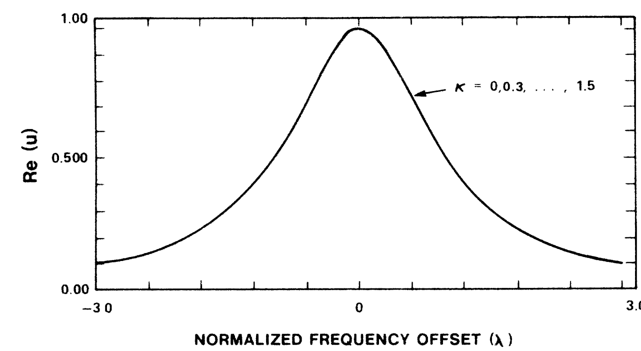

FIG. 6. Maximum real part of the six normalized roots vs normalized frequency offset. The normalized phase mismatch is varied. The laser is circularly polarized. The Anti-Stokes–Stokes wave number ratio is 1.22. The curve shape is Lorentzian.

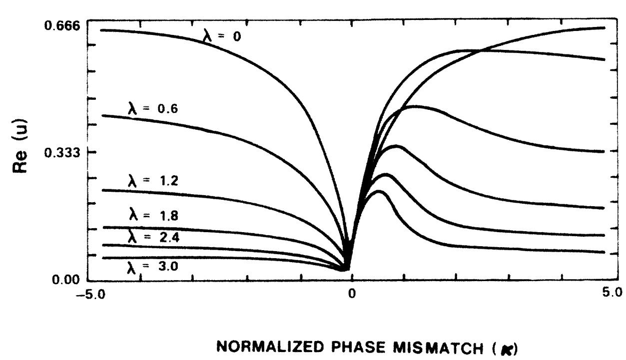

FIG. 4. Maximum real part of the six normalized roots vs normalized phase mismatch. The normalized frequency offset is varied. The laser is linearly polarized. The anti-Stokes–Stokes wave number ratio is 1.22.

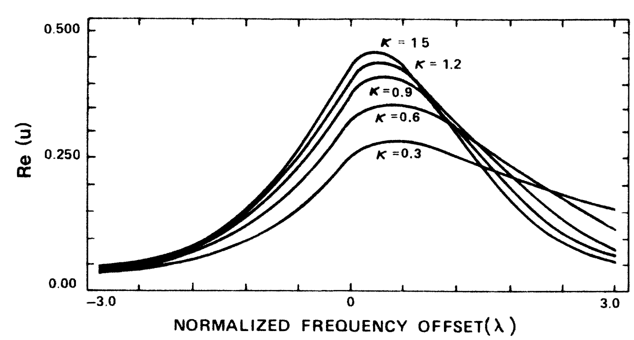

FIG. 7. Maximum real part of the six normalized roots vs normalized frequency offset. The normalized phase mismatch is varied. The laser is unpolarized. The Anti-Stokes–Stokes wave number ratio is 1.02.

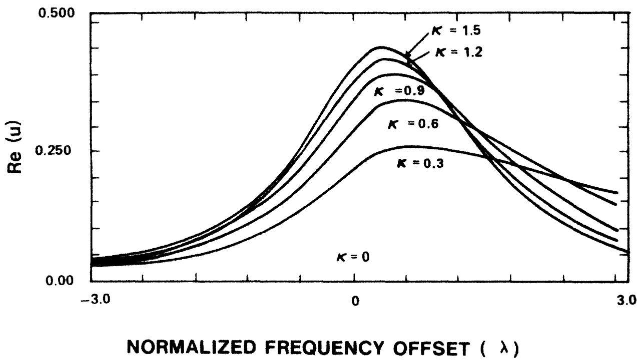

FIG. 8. Maximum real part of the six normalized roots vs normalized frequency offset. The normalized phase mismatch is varied. The laser is unpolarized. The Anti-Stokes–Stokes wave number ratio is 1.22.

Figures 2 and 3 give plots of the maximum gain versus κ for various pump-light polarization ellipticities and degrees of polarizations, respectively. The gradual transition from large helicity (circular) to small helicity (linear, unpolarized) is noteworthy.

In Fig. 4 we plot the maximum gain as a function of phase mismatch κ for various values of frequency offset λ, with linearly polarized light. For small (nonzero) κ the maximum gain occurs near $\lambda = 1$, i.e., half of linewidth off resonance. This result is similar to that obtained by Shen and Bloembergen in their scalar theory.[7]

Figures 5–8 show the homogeneous line shapes for various propagation geometries and polarizations. Figure 5 shows line profiles for linearly polarized light and various values of the phase mismatch. Figure 6 shows that the theory verifies the Lorentzian line profile regardless of phase mismatch for circularly polarized light. Figure 7 demonstrates how the SA coupling shifts and broadens the line profile near phase matching for unpolarized light; Fig. 8 shows the same effect, despite the larger Raman shift.

V. CONCLUSIONS

A new rotationally invariant formalism for Raman scattering has been derived. It has been applied to $J = 0$ (scalar) and $J = 2$ (electric quadrupole) transitions and has given results which duplicate and extend the work of other authors. Of special interest are the predictions regarding unpolarized light and the eigenvalue spectrum of six eigenvalues for a given pump polarization for S Raman transitions. For a linearly polarized pump, all eigenpolarizations of the stimulated Stokes light are suppressed for phase-matched propagation.

The method used also has a useful intuitive interpretation. The optical phonons are viewed from the outset as coherent molecular excitations which carry angular momentum, an obvious consequence of the selection rules as discussed by Placzek.[19]

Our treatment of unpolarized light has some interesting implications. Since the Raman gain coefficients depend on the polarization of the incident light, it is not obvious what happens if the light polarization changes as a function of time. The fact that the gain is essentially independent of laser spectral width makes this an especially intriguing question, since the broadening of the laser spectrum associated with the time-dependent changes in the polarization do not necessarily affect the gain of the temporally correlated component of the Stokes radiation.

A simple interpretation of our results on unpolarized light is as follows: the phonons, i.e., coherent molecular excitations, may be viewed as a "detector" of the polarization of the incident laser light, since the degree of Raman conversion, which is the "output signal" produced by the phonons, is sensitive to the incident light polarization. What happens if the polarization changes as a function of time? Viewing the phonons as a detector with a finite response time τ (given by ~ 10 times the phonon lifetime, the factor of 10 arising from the nonlinearity of SRS[2]), we would argue that the answer depends on how rapidly the polarization changes compared to τ^{-1}. If the polarization changes occur on a time scale much longer than τ then the detector should respond to the instantaneous polarization, emitting a strong (weak) signal when the light is circularly (linearly) polarized. On the other hand, if the polarization changes significantly on a time scale of τ then the detector cannot follow the instantaneous polarization. (This is the limit of unpolarized light which we have treated mathematically in Sec. III using our model of a multiaxial-mode laser with the various modes polarized differently and the mode spacing much greater than the Raman linewidth; in this model, the instantaneous polarization of the laser is obviously fluctuating on a time scale much shorter than τ.) Since the detector should average over the polarization fluctuations, we might have thought that it could, in principle, treat the light as an incoherent mixture of any two orthogonal polarizations. However, this cannot be correct since the magnitude of the signal would then depend on the choice of basis. Remembering that SRS may be thought of as an instability, we would expect that the choice of basis which maximizes the Stokes-light growth would be the one which would eventually dominate. In the simple case in which the SA coupling may be ignored, this is indeed what is found mathematically: the phonons "resolve" the unpolarized light into two incoherent, circular components of equal intensity; the gain is thus half the gain of circularly polarized light. The case of perfect phase matching, in which the SA coupling dominates the phonon response, is more complicated and cannot be understood from this simple argument; as discussed in Sec. IV, the gain is nevertheless suppressed when the incident light is unpolarized.

ACKNOWLEDGMENTS

The authors are grateful to M. Feld for helpful criticism. This work was supported by the U.S. Office of Naval Research under Contract No. N00014-84-C-0629.

*Present address: Science Research Laboratory, 15 Ward Street, Somerville, MA 02143.

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