Polarization Dependence of Gain in Stimulated Raman Scattering

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We present 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 (Stokes) transitions, yielding the explicit dependence of gain on light polarization, phase mismatch, and frequency offset.

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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 phonon response,[2] the spectrally wide laser,[3] wave-optics (diffractive) effects,[4] strong dispersion,[5] and quantum-electrodynamic effects.[6] An additional process which has been discussed in the literature is the effect of Stokes–anti-Stokes 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 an anti-Stokes photon.[7,8] However, to our knowledge, no general theory of the polarization dependence of the gain has been formulated. Such a theory is 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[8] disappears when the (pump) laser and Stokes photons copropagate and are circularly polarized in opposite senses.[9] We sketch such a general theory in this Letter. We show that rotational Raman conversion may be described in terms of six complex eigenvalues, three of which correspond to growth and three to attenuation. In general, the value of 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 these eigenvalues for a linearly, circularly, elliptically, and partially polarized laser. To our knowledge, this is the first SRS formalism which is general enough to treat pump light of *arbitrary polarization*, including partial polarization. It is particularly applicable to very recent experiments on and modeling of stimulated rotational Raman scattering.[10]

Consider a pump, Stokes, and anti-Stokes waves, with fields $\mathbf{E}_P$, $\mathbf{E}_S$, and $\mathbf{E}_A$, respectively. We model each as consisting of an arbitrary number of axial modes of equal intermode spacing $\Delta\omega$. Factoring out the central optical frequencies, we write the fields in terms of slowly varying amplitudes as follows:

$$\mathbf{E}_F=[\tfrac{1}{2}\textstyle\sum_n \mathbf{F}_n e^{in\Delta\omega(z/c-t)}]e^{i(\mathbf{k}_F\cdot\mathbf{z}-\omega_F t)}+\text{c.c.}, \tag{1}$$

where $F=P$, S, or A; ω_F is the circular frequency and $\mathbf{k}_F$ is the wave vector of wave F. For simplicity we restrict our analysis to the steady-state response of the Raman phonons. We assume that $\omega_P-\omega_S=\omega_A-\omega_P$, but that the central beat frequency $\omega_P-\omega_S$ does not necessarily coincide with the Raman shift Ω. We also assume that $\Delta\omega\gg\Gamma$, where 2Γ is the FWHM Raman linewidth, but that the total spectral width of each wave is small enough that the difference in group velocities between the waves (due to dispersion in the medium) has no appreciable effect. As has been shown by many authors,[11] this assumption makes the multimode problem similar in many respects to that of a single axial mode. The slowly varying phonon amplitude $\mathbf{R}$ may then be expanded in spherical tensors of rank J and various components m $(-J\le m\le +J)$ as follows:

$$R_m^{(J)}=\sum_{\alpha,\beta,n}\langle Jm\,|\,1\,1\,\alpha\,-\beta\rangle[P_{\alpha,n}S_{\beta,n}^*+A_{\alpha,n}P_{\beta,n}^*e^{-i\Delta\mathbf{k}\cdot\mathbf{z}}], \tag{2}$$

where $\langle Jm\,|\,1\,1\,\alpha\,-\beta\rangle$ is a Clebsch-Gordan coefficient, $F_{\alpha,n}$ is spherical component α of the amplitude of axial mode n of field F $(=\text{A},P,\text{S})$, and $\Delta\mathbf{k}$ is the phase mismatch, given by $2\mathbf{k}_P-\mathbf{k}_A-\mathbf{k}_S$. The form of Eq. (2) is intuitively obvious: The field amplitudes are rank-1 spherical tensors which must be combined as indicated in Eq. (2) to form spherical tensors of rank J so that both sides of Eq. (2) 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 along the z direction. The equations for the spatial evolution of the waves is then

$$\begin{aligned}\partial_z S_{\beta,n}&=\textstyle\sum_J g^{(J)}\sum_{\mu M}\langle 1\,\beta\,|\,1\,J\,\mu\,-M\rangle P_{\mu,n}R_M^{(J)*},\\ \partial_z A_{\alpha,n}&=-\textstyle\sum_J g^{(J)*}(k_A/k_S)\sum_{\mu M}\langle 1\,\alpha\,|\,1\,J\,\mu\,M\rangle P_{\mu,n}R_M^{(J)}e^{i\Delta k z},\end{aligned} \tag{3}$$

where $g^{(J)}$ is given by

$$g^{(J)}=N^{(J)}g_0^{(J)}/(1+i\lambda). \tag{4}$$

Here $N^{(J)}$ is a normalization constant, $g_0^{(J)}$ is a (real) Stokes-amplitude gain coefficient, and λ is the normalized frequency offset, $(\omega_P-\omega_S-\Omega)/\Gamma$. For convenience we have normalized the sums of the squares of the pump field amplitudes to unity, so that the real coefficients $g_0^{(J)}$ are proportional to the pump intensity.

With no loss of generality, we will now treat the case in which only a single coefficient $g_0^{(J)}$ is nonzero. As will be discussed below, this will be exactly true for S rotational transitions, and approximately true for many vibrational transitions, as well.

To find the eigenvalues of Eqs. (2)-(3) in the small-signal limit of no significant pump depletion, we differentiate Eq. (2) with respect to z and substitute Eq. (3) into the resulting equation. We obtain

$$\partial_z R_m^{(J)}=g^{(J)*}\sum_\mu (D_{mM}^{(J)}+B_{mM}^{(J)})R_M^{(J)}-i\Delta k\sum_{\alpha,\beta,n} h_{m\alpha\beta}^{(J)}A_{\alpha,n}P_{\beta,n}^{*}e^{-i\Delta k z}, \tag{5}$$

where

$$D_{mM}^{(J)}=\sum_{\alpha,\mu} d_{m\alpha\mu M}^{(J)}C_{\alpha\mu},\quad B_{mM}^{(J)}=-\frac{k_A}{k_S}\sum_{\alpha,\mu} b_{m\alpha\mu M}^{(J)}C_{\alpha\mu}, \tag{6}$$

$$d_{m\alpha\mu M}^{(J)}=\sum_\beta \langle J\,m\,|\,1\,1\,\alpha-\beta\rangle\langle 1\,\beta\,|\,1\,J\,\mu-M\rangle,\quad b_{m\alpha\mu M}^{(J)}=\sum_\beta \langle J\,m\,|\,1\,1\,\beta-\alpha\rangle\langle 1\,\beta\,|\,1\,J\,\mu M\rangle, \tag{7}$$

$$h_{m\alpha\beta}^{(J)}=\langle J\,m\,|\,1\,1\,\alpha-\beta\rangle, \tag{8}$$

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

$$C_{\alpha\mu}=\sum\nolimits_n P_{\alpha,n}P_{\mu,n}^{*}. \tag{9}$$

Differentiating Eq. (5) with respect to z and substituting Eq. (3) into the resulting equation yields

$$\partial_z^2 R=g^{*}(D+B)\partial_z R-i\Delta k[\partial_z R-g^{*}(D+B)R]-i\Delta k\, g^{*}BR \tag{10}$$

where R is now understood to be a column vector with $2J+1$ elements given by Eq. (2), while D and B are matrices with matrix elements given by Eq. (6); we have suppressed the superscripts and subscripts to simplify the notation. Equation (10), which determines the phonon growth for arbitrary phase mismatch, is the central result of this Letter.

To obtain the eigenvalues and eigenvectors of Eq. (10), we substitute

$$R=R_0e^{ug_0I_0z} \tag{11}$$

so that u represents the complex eigenvalue, normalized to g_0I_0 (I_0 is the laser intensity); the real part of u is proportional to the gain. Since Eq. (10) is homogeneous in R, the roots u must satisfy

$$\det[u(u+iK)I-u\eta(D+B)-iK\eta D]=0\ , \tag{12}$$

where I is the identity matrix, $\eta=g^*/g_0I_0$, and $K=\Delta k/g_0I_0$ is the phase-mismatch factor.

Before discussing the roots u in the most general case, we investigate the limits of both small and large phase mismatch. According to Eq. (12), the eigenvalues u in the phase-matched limit ($|K|\ll 1$) are obtained by diagonalizing the matrix $D+B$. On the other hand, far from phase matching ($|K|\gg 1$) we find instead that the eigenvalues are obtained by diagonalizing D; this may also be seen directly from Eqs. (2) and (3) 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 respectively to scattering with isotropic, magnetic-dipole, and electric-quadrupole rotational symmetry, as described by Placzek.[12] 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. This case is straightforward and will not be discussed in detail; 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 effect.[12] 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 $J=2$. In the remainder of this Letter we will focus our attention on this case.

At the phase-matching angle we find that for quantization along z the matrix $D+B$ is already diagonal; Eq. (5) reduces to

$$\partial_z R_m^{(2)} =(mg_0^{(2)}/2)(1+i\lambda)^{-1}(C_{++}-C_{--})R_m^{(2)}, \tag{13}$$

where $m=0,\pm 2$. Thus for pump light which is right

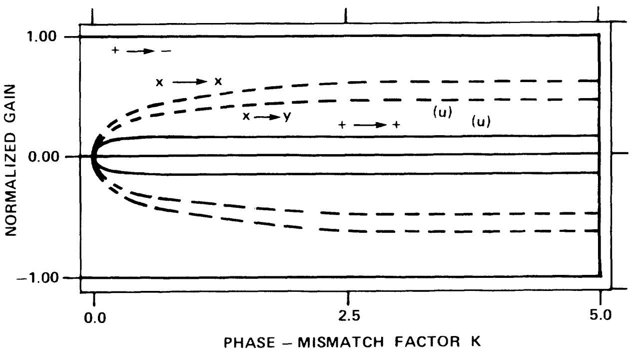

FIG. 1. Normalized gain Re(u) vs phase-mismatch factor K calculated from Eq. (12) for $\lambda=0$. Solid (dashed) curves: circularly (linearly) polarized pump, with sample polarizations given for each. Unpolarized-pump solutions (u) coincide with those for $x\rightarrow y$ and $+\rightarrow +$, as shown.

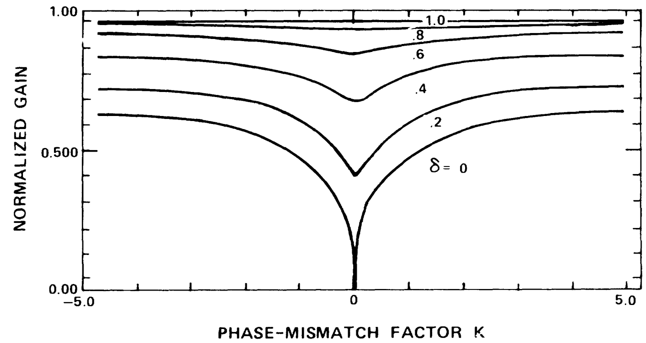

FIG. 2. Maximum normalized resonant gain vs K for various pump polarization ellipticities. The quantity δ indicates the ratio of the minor to the major axis of the polarization ellipse.

circularly polarized (+), the full gain occurs at the phase-matching angle with the Stokes light 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 Stokes–anti-Stokes 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. 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.[12,13] For unpolarized light we obtain $u=\frac{1}{2}$, $\frac{1}{2}$, and $\frac{1}{6}$. Evidently, far from phase matching unpolarized light is automatically "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 ($\lambda\neq 0$) 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. (12) for arbitrary values of K. In Fig. 1 we have plotted the normalized gain Re(u) for a linearly polarized, circularly polarized, and unpolarized pump laser, all on Raman resonance ($\lambda=0$) and with $\Omega \ll \omega_P$. The positive (negative) branches are predominantly Stokes (anti-Stokes) roots when $K\neq 0$, and correspond to amplification (attenuation). The plots show that the transition between the phase-matched and unmatched regimes occurs around $K=1$, as expected. Note that no root crossings occur except at $K=0$; thus, for a linearly (x) polarized pump the dominant root corresponds to parallel (x) Stokes polarization for all values of $K\neq 0$. 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 Eq. (1) an unpolarized multimode laser may be viewed as a type of single-mode laser whose polarization is modulated on a time scale short compared to the phonon lifetime Γ^{-1}; our result should apply to any such arbitrary modulation.

Figures 2 and 3 give plots of the maximum gain versus K for various pump polarization ellipticities and degrees

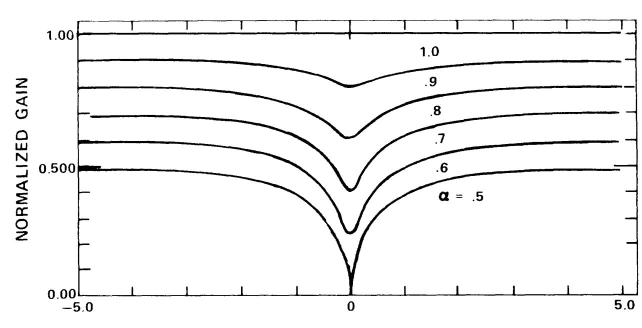

FIG. 3. Maximum normalized resonant gain vs K for various values of the parameter $\alpha=C_{++}/(C_{++}+C_{--})$ of the partially polarized pump laser.

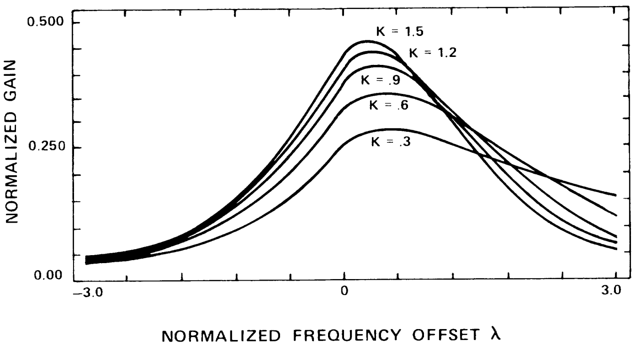

FIG. 4. Maximum normalized gain vs frequency offset λ for various values of K and an unpolarized pump.

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 frequency offset λ for various values of K, unpolarized light, and $\Omega \ll \omega_P$. For small K the maximum gain occurs near $\lambda = 1$, i.e., half a linewidth off resonance. This result is similar to that obtained by Shen and Bloembergen in their scalar theory.[7]

We note that for backward SRS the Stokes–anti-Stokes coupling plays a negligible role, since K becomes very large. Thus backward SRS is characterized by the relative gains obtained above for large K.

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