Theory of stimulated Raman scattering with broad-band lasers

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The authors have extended the theory of stimulated Raman scattering to include the effects of laser bandwidths, in both the transient and steady-state regimes. The case of two interacting laser beams, a pump laser and a probe (Stokes) laser, is treated. Using the phase-diffusion model for laser bandwidth, the authors demonstrate that in the absence of dispersion, the forward Raman gain is essentially independent of the laser bandwidths in the high-gain limit, while in the low-gain limit the gain coefficient is inversely proportional to the sum of the bandwidths. It is further shown that when the pump-laser bandwidth is much larger than the linewidth of the Raman medium, the stimulated Stokes output assumes the same spectrum as the pump laser in the high-gain limit. A possible interpretation of these results is discussed assuming a "phase-locking" of the Stokes phase to the fluctuations in the pump-laser phase, due to the nonlinear interaction of the two beams through the Raman medium.

I. INTRODUCTION

The effects of finite laser bandwidth are being recognized as important in the study of nonlinear optical processes. Resonance fluorescence,[1-6] two-photon absorption,[7,8] second harmonic generation,[9] multiphoton ionization,[10-13] and stimulated Raman scattering are all areas in which key elements of understanding depend on the ability to model the laser, not as a monochromatic wave of definite phase and amplitude, but as a multimode broad-band wave with fluctuating phase and amplitude. The problem of stimulated Raman scattering (SRS) is especially timely in light of ongoing efforts to use it as method for developing new coherent light sources[14] as well as compressing high-energy laser pulses to achieve higher peak powers for use in laser fusion.[15] In applications of these types a detailed understanding of all the factors influencing the efficiencies of the processes is obviously desirable. However, one important factor, laser bandwidth, has not yet been fully explored.

Recently, two groups[15,16] have observed a large forward-backward asymmetry of the Raman gain, which they attribute to the broad-band nature of the pump laser used. These were in the absence of other effects, such as self-focusing or extraneous feedback, which are known to produce anomalous gains.[17] These asymmetries are consistent with several theoretical predictions[18,19] that in the backward direction (counterpropagating pump and Stokes waves) the gain coefficient is proportional to $(\Gamma + \Gamma_L)^{-1}$, where Γ and Γ_L are the spectral widths of the Raman medium and the pump laser, respectively; while, in the forward direction, in the absence of dispersion of the Stokes wave relative to the pump wave, the gain coefficient is

proportional to Γ^{-1} alone. Carman et al.[18] refer to this as "the rather startling conclusion... that the Stokes gain is independent of the frequency spectrum of the (pump) laser... even if this spectrum is much broader than Γ." Thus when Γ_L is much larger than Γ the forward gain is much larger than the backward gain. These results go against intuition based on the idea that gain should depend on the number of photons per unit frequency in the pump beam. Apparently, the concept of photons as independent incoherent bundles is inadequate to describe the subtleties in the SRS problem.

The purpose of this paper is to further develop the theory of SRS, including the effects of finite laser bandwidths, in a way that allows explicit calculation of the gain and spectrum of the Stokes wave. We consider, as in Fig. 1, a medium of three-level atoms interacting with two classical
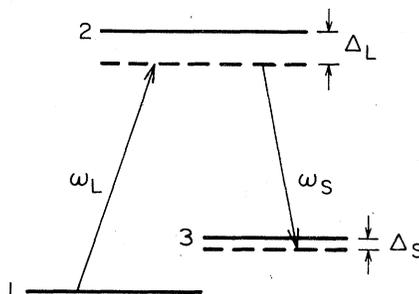

FIG. 1. Three-level atom interacting with a pump laser with frequency ω_L and a probe (Stokes) laser with frequency ω_S. The cumulative detunings are Δ_L and Δ_S.

electromagnetic waves; one a pump (laser) wave and the other a Stokes wave, differing in frequency by the Raman shift of the medium. Both waves have constant amplitudes, but phases which fluctuate randomly, giving rise to bandwidths Γ_L and Γ_S. This is the "phase-diffusion model," which has been used recently as a means to include bandwidth effects into the calculation of light-scattering spectra,[2-6] as well as multiphoton ionization.[10,11] The calculations are performed as statistical averages over the random phase variables of the two waves. Previous treatments[18,19] of SRS attempted to accommodate both fluctuating phases and amplitudes. We will discuss later why the inclusion of amplitude fluctuations in this problem is such a difficult task. Within the stated model, we verify the independence of the forward gain from the pump width Γ_L and the Stokes width Γ_S, in the high-gain limit. Further we will show that in the case that the width of the pump laser Γ_L is broader than the Raman linewidth Γ, the amplified Stokes signal assumes the spectral width of the pump laser, regardless of its initial width.

Akhmanov, D'yakov and Pavlov[19] have separated the problem into four regimes of interest: (i) $\Gamma_L \ll \Gamma$, with no dispersion; (ii) $\Gamma_L \ll \Gamma$, with dispersion; (iii) $\Gamma_L \gg \Gamma$, with no dispersion; (iv) $\Gamma_L \gg \Gamma$, with dispersion. Case (i) was considered by Bloembergen and Shen,[20] who predicted an enhancement of the forward gain for a multimode laser. In this paper we treat mainly case (iii), where the laser linewidth is broader than the atomic linewidth. Here there is no enhancement, but neither is there a significant suppression of the gain, compared to that calculated in case (i) in the limit $\Gamma_L = 0$. Carman et al.[18] have also treated case (iii), and they reached essentially the same conclusions by calculating numerical solutions to the problem. Cases (ii) and (iv) treat the effects of dispersion. There is a consensus[18,19,21] that broadening of the laser in the presence of dispersion does result in a lowering of the gain, due to the inability of the Stokes wave to stay correlated with the pump fluctuations as they propagate. However, Akhmanov et al.[19] have shown, further, that there is a critical pump intensity, above which the effects of dispersion are overcome and the gain coefficient increases again to nearly the narrow-band value.

Dzhotyan et al.[22] have treated the problem by assuming the pump and Stokes waves to be composed of many monochromatic modes, with uniform frequency spacings large compared to the Raman linewidth Γ. This results in significant interaction only between certain resonant pairs of modes (one pump and one Stokes). This approach can be

thought of as complementary to the present approach, in which the energy in the waves is taken to be spread continuously over a small frequency interval. The multimode approach of Dzhotyan et al. is a generalization of an idea developed by Giordmaine and Kaiser[22] (and discussed by Byer and Herbst[22]), in which the pump and Stokes waves each consist of two modes. This treatment illustrates the relationship of SRS with four-wave parametric interactions. Another related discussion is that of Harris,[22] in which the threshold for parametric oscillation with multimode lasers is shown to depend only on the total power in the pump laser.

In Sec. II we derive the equations of motion for the Raman problem in a novel way by using the "two-photon vector model" of Takatsuji[24] and Grischkowsky et al.[25] In Sec. III we review the general solutions of the equations, following Carman et al.[18] and evaluate the gain with monochromatic pumping for both the transient and steady-state limits. Then we apply the phase-diffusion model to evaluate the gain under arbitrary broad-band pumping conditions, again in both the transient and steady-state limits. In Sec. IV we calculate the spectrum of the amplified Stokes wave by considering its autocorrelation function in the steady-state limit. In Sec. V we discuss a possible interpretation of the results obtained, and in Sec. VI we summarize the main results of the paper.

II. EQUATIONS OF MOTION

Here we give a novel derivation of the usual equations of motion for the Raman problem and a discussion of the physical model leading to them. For simplicity, we treat the case of near-resonance Raman Stokes scattering, in which only three atomic levels need to be considered. Thus we consider a vapor of atoms with energy levels shown in Fig. 1. A pump laser is tuned near (but not on) the 1-2 transition and a probe laser is tuned near (but not on) the 2-3 transition. It is sufficient to treat the pump laser as a prescribed field as long as it is not depleted. The probe laser will experience gain in a manner dependent on both the amplitude and phase structure of the pump laser.

Consider the fields $\vec{E}_L$ (pump laser) and $\vec{E}_S$ (Stokes, or probe laser) acting on the three-level atom of Fig. 1. Let

$$\vec{E}_L = \mathscr{E}_L \hat{x}_L \cos(\omega_L t - k_L z + \varphi_L) = \mathscr{E}_L \hat{x}_L \cos\phi_L, \quad (1a)$$

$$\vec{E}_S = \mathscr{E}_S \hat{x}_S \cos(\omega_S t \mp k_S z + \varphi_S) = \mathscr{E}_S \hat{x}_S \cos\phi_S, \quad (1b)$$

where $\mathcal{E}_L$ and $\mathcal{E}_S$ are the (real) amplitudes of the waves, with linear polarization vectors $\hat{x}_L$ and $\hat{x}_S$, carrier frequencies ω_L and ω_S, propagation vectors $\vec{k}_L = k_L \hat{z}$, $\vec{k}_S = k_S \hat{z}$, and slowly varying phases φ_L and φ_S. The state of the atom can be written

$$\psi = a_1 e^{-i\omega_1 t}\psi_1 + a_2 e^{-i(\omega_1 t + \phi_L)}\psi_2$$
$$+ a_3 e^{-i(\omega_1 t + \phi_L - \phi_S)}\psi_3 , \tag{2}$$

where a_1, a_2, and a_3 are the slowly varying coefficients in a "doubly rotating frame,"[23] and ψ_1, ψ_2, and ψ_3 are the stationary eigenstates of the atomic Hamiltonian, with energies $\hbar\omega_1$, $\hbar\omega_2$, and $\hbar\omega_3$. At $t = 0$ the atom is in the ground state ($a_1 = 1$, $a_2 = a_3 = 0$) and afterwards the coefficients evolve according to Schrödinger's equation

$$i\dot{a}_1 = -\tfrac{1}{2}\Omega_L a_2 , \tag{3a}$$

$$i\dot{a}_2 = (\Delta_L - \dot{\varphi}_L)a_2 - \tfrac{1}{2}\Omega_L a_1 - \tfrac{1}{2}\Omega_S a_3 , \tag{3b}$$

$$i\dot{a}_3 = (\Delta_S - \dot{\varphi}_L + \dot{\varphi}_S)a_3 - \tfrac{1}{2}\Omega_S a_2 , \tag{3c}$$

where the detunings are $\Delta_L = \omega_{21} - \omega_L$ and $\Delta_S = \omega_{31} + \omega_S - \omega_L$. The Rabi frequencies for the two transitions are given by $\Omega_L = d_{12}\mathcal{E}_L/\hbar$ and $\Omega_S = d_{23}\mathcal{E}_S/\hbar$, where $d_{12} = \vec{d}_{12} \cdot \hat{x}_L$ and $d_{23} = \vec{d}_{23} \cdot \hat{x}_S$ are dipole matrix elements. The rotating-wave approximation[23] (RWA) has been invoked in writing Eq. (3). This is valid when the detunings are small enough ($\Delta_L \ll \omega_{21}$, $\Delta_S \ll \omega_{23}$).

Several authors[24-26] have discussed the case in which level 2 may be eliminated from the Eqs. (3a)–(3c). When Δ_L is much larger than Δ_S and the fields have no appreciable Fourier components at the atomic frequencies, we may set $\dot{a}_2 \approx 0$ in Eq. (3b) and get (neglecting $\dot{\varphi}_L$)

$$a_2 \approx (\tfrac{1}{2}\Omega_L a_1 + \tfrac{1}{2}\Omega_S a_3)/\Delta_L . \tag{4}$$

This approximation is the basis of the "two-photon vector model" of Takatsuji[24] and Grischkowsky et al.,[25] and is discussed more fully in Appendix A. Using this approximation in the Schrödinger equation [Eq. (3)], one may obtain two equations for a_1 and a_3, which are identical in form to those of a one-photon transition with effective Rabi frequency $\Omega_e = \tfrac{1}{2}\Omega_L\Omega_S/\Delta_L$ and effective detuning $\Delta_e = \Delta_S + \tfrac{1}{4}(\Omega_L^2 - \Omega_S^2)/\Delta_L$, which shows the effect of ac Stark shifting. We write the resulting equations in the convenient Bloch form,[23] using $U + iV = 2a_1 a_3^*$, and $W = a_3 a_3^* - a_1 a_1^*$,

$$\dot{U} = -(\Delta_e - \dot{\varphi}_L + \dot{\varphi}_S)V - \Gamma U , \tag{5a}$$

$$\dot{V} = (\Delta_e - \dot{\varphi}_L + \dot{\varphi}_S)U + \Omega_e W - \Gamma V , \tag{5b}$$

$$\dot{W} = -\Omega_e V . \tag{5c}$$

Here we have included the phenomenological col-

lisional dephasing rate Γ, which is the halfwidth at half maximum (HWHM) in rad sec^{-1} for the (Raman) transition between levels 1 and 3.

To describe Raman amplification, one must solve the wave equation for the Stokes wave

$$\frac{\partial^2 \vec{E}_S}{\partial z^2} - \frac{1}{v^2}\frac{\partial^2 \vec{E}_S}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 \vec{P}}{\partial t^2} , \tag{6}$$

where v is the velocity in the medium, and $\vec{P}$ is the polarization of the medium. Considering only Fourier components close to the carrier frequency ω_S, and only the linear polarization $\hat{x}_S$, leads to a polarization

$$P = N \langle \psi | \vec{d} \cdot \hat{x}_S | \psi \rangle = 2Nd_{23}\,\mathrm{Re}(a_2 a_3^*)\cos\phi_S$$
$$+ 2Nd_{23}\,\mathrm{Im}(a_2 a_3^*)\sin\phi_S , \tag{7}$$

where N is the atomic number density. Making use of Eq. (7) for the polarization, Eq. (4) to again eliminate a_2, and the slowly varying envelope approximation,[23] one can write Eq. (6) in the form

$$\pm\frac{\partial \mathcal{E}_S}{\partial z} + \frac{1}{v}\frac{\partial \mathcal{E}_S}{\partial t} = -\kappa_2 \mathcal{E}_L V , \tag{8a}$$

$$\mathcal{E}_S\left(\pm\frac{\partial \varphi_S}{\partial z} + \frac{1}{v}\frac{\partial \varphi_S}{\partial t}\right) = -\kappa_2 \mathcal{E}_L U - \frac{d_{23}}{d_{12}}\kappa_2 \mathcal{E}_S (W + 1) , \tag{8b}$$

where $\kappa_2 = \pi N \omega_S v d_{12} d_{23}/c^2\hbar\Delta_L$ and the plus and minus signs are for copropagating and counterpropagating pump and probe beams, respectively. Equations (5) and (8), along with a similar one for the pump laser $\mathcal{E}_L$, completely describe the propagation and material response for the Raman problem, including the effects of phase modulation ($\dot{\varphi}_L$, $\dot{\varphi}_S$). They have been derived by Takatsuji[24] and by Courtens[26] in essentially the same form and used for considerations of optical transient phenomena.

In contrast, we are interested in the special case that the atoms are weakly excited and the pump laser is a prescribed, external field. Thus we take for the inversion $W = -1$, $\dot{W} = 0$ in Eq. (5). We also assume exact resonance ($\Delta_e = 0$). The remaining four equations, (5a), (5b), (8a), and (8b), can be combined into two complex equations. Defining the quantities

$$E_L = \mathcal{E}_L e^{i\varphi_L}; \quad E_S = \mathcal{E}_S e^{i\varphi_S};$$
$$Q = (U + iV)e^{i(\varphi_L - \varphi_S)} , \tag{9}$$

we obtain directly

$$\pm\frac{\partial E_S}{\partial z} + \frac{1}{v}\frac{\partial E_S}{\partial t} = -i\kappa_2 Q^* E_L , \tag{10a}$$

$$\frac{\partial Q^*}{\partial t} = -\Gamma Q^* + i\kappa_1 E_L^* E_S , \tag{10b}$$

where $\kappa_1 = d_{12}d_{23}/2\hbar^2\Delta_L$. These two coupled equations are the starting point for many theories of

stimulated Raman scattering. They have usually been derived in the coupled wave approach of nonlinear optics[27] for the case of molecular Raman scattering, where Q is the normal-mode coordinate of a molecular vibration and is often called an optical-phonon wave. In those treatments, perturbation theory was used from the beginning and the coupling constants κ_1 and κ_2 were given in terms of molecular polarizabilities. Here we have provided a connection between the "two-photon vector model" and the standard theories of Raman propagation. We have given the explicit relations, Eq. (9), between the variables used in the earlier nonlinear-optics theories and the more modern optical resonance or Bloch vector picture, which has been used here, and continues to give insight into many laser-related problems.

III. EVALUATION OF RAMAN GAIN

General solutions of Eq. (10) have been obtained in the case of copropagating waves by Carman et al.[18] In this case the prescribed undepleted pump-laser field E_L depends only on the local time variable $\tau = t - z/v$. It is assumed that the waves travel with equal velocity $(k_L = k_S)$, i.e., there is no dispersion. Denoting by $E_S(0, \tau)$ the Stokes field at the input of the cell $(z = 0)$, the solution for the Stokes output field is[18]

$$E_S(z, \tau) = E_S(0, \tau) + (\kappa_1 \kappa_2 z)^{1/2} \int_0^\tau \frac{e^{-\Gamma(\tau - \tau')}}{[p(\tau) - p(\tau')]^{1/2}}$$

$$\times I_1(\{4\kappa_1 \kappa_2 z[p(\tau) - p(\tau')]\}^{1/2})$$

$$\times E_L(\tau) E_L^*(\tau') E_S(0, \tau') d\tau', \qquad (11)$$

where $I_1(x)$ is the Bessel function of imaginary argument,[28] and

$$p(\tau) = \int_0^\tau |E_L(\tau'')|^2 d\tau''$$

is the integrated power in the pump laser up to time τ.

A. Stokes gain for monochromatic pump and input waves

It is instructive to review the properties of the solution (11) for the case that the pump wave and the input Stokes wave are constant and monochromatic. In this case we have $E_L(\tau) = E_L^*(\tau') = \mathcal{E}_L$ and $E_S(0, \tau') = \mathcal{E}_{S0}$. This leads to

$$E_S(z, \tau) = \mathcal{E}_{S0} + \mathcal{E}_{S0} \frac{(\alpha z)^{1/2}}{2}$$

$$\times \int_0^\tau \frac{e^{-\Gamma x}}{\sqrt{x}} I_1[(\alpha z x)^{1/2}] dx, \qquad (12)$$

where we have used $x = \tau - \tau'$, $p(\tau) = \mathcal{E}_L^2 \tau$ and $\alpha = 4\kappa_1 \kappa_2 \mathcal{E}_L^2$. Following Wang[27] we present analytic

approximations and numerical evaluations of the Stokes output, given by Eq. (12), for two different limits.

1. Transient limit

The transient limit occurs for times much shorter than the reciprocal of the Raman linewidth $(\Gamma \tau \ll 1)$. After replacing $e^{-\Gamma x}$ by 1 in Eq. (12), the integral can be done to give $E_S(z, \tau) = \mathcal{E}_{S0} I_0[(\alpha z \tau)^{1/2}]$, which for large enough $\alpha z \tau$ leads to the asymptotic form for the output Stokes intensity

$$E_S^2(z, \tau) \simeq \frac{\mathcal{E}_{S0}^2}{2\pi} \frac{e^{2(\alpha z \tau)^{1/2}}}{(\alpha z \tau)^{1/2}}$$

$$\text{(high gain, } \Gamma\tau \ll 1). \qquad (13)$$

We have used the property $I_i(x) \to e^x/(2\pi x)^{1/2}$, for $x \to \infty$, for any i.[28] Equation (13) is the usual result for the transient Raman effect.[27] It is interesting to note that, in the transient limit, the Raman gain given by Eq. (13) does not depend on the Raman linewidth Γ.

2. Steady-state limit

The steady-state limit occurs for times much larger than the reciprocal of the Raman linewidth $(\Gamma\tau \gg 1)$. Extending the upper limit to infinity, the integral in Eq. (12) can be done exactly[29] to give for the Stokes intensity

$$E_S^2(z, \tau) = \mathcal{E}_{S0}^2 e^{gz} \quad \text{(arbitrary gain, } \Gamma\tau \gg 1), \quad (14a)$$

where

$$g = \frac{\alpha}{2\Gamma} = \frac{\pi N \omega_S}{c^2} \frac{v d_{12}^2 d_{23}^2}{\hbar^3 \Delta_L^2} \frac{\mathcal{E}_L^2}{\Gamma}. \qquad (14b)$$

The steady-state gain coefficient g is the usual one derived for stimulated electronic Raman scattering.[30] It does depend on the Raman linewidth Γ, in contrast to the transient case.

Equations (13) and (14a) for the output Stokes intensity, along with numerical evaluation of Eq. (12), are plotted in Fig. 2, as a function of gz, or equivalently, pump laser intensity, for both a transient case $(\Gamma\tau = 10^{-2})$ and a steady-state case $(\Gamma\tau = 10^2)$. Here we interpret τ as the pulse duration of the pump laser. Equation (13) for the transient gain $(\Gamma\tau = 10^{-2})$ is seen to agree well with the exact numerical results when $\log_{10}(E_S^2/\mathcal{E}_{S0}^2)$ > 1, while Eq. (14a) for the steady-state gain $(\Gamma\tau = 10^2)$ agrees everywhere. Note that the values below $\log_{10}(E_S^2/\mathcal{E}_{S0}^2) = 0$ are unphysical. The other four curves in Fig. 2 show the effects of laser bandwidth on the gain, as described in Sec. III B.

B. Raman gain for broad-band pump and/or input waves

We now evaluate the Raman gain in the case that the spectral width of either the pump laser or the

Stokes input wave (or both) is greater than the Raman linewidth $(\Gamma_L + \Gamma_S \gg \Gamma)$. This can be accomplished by performing an average of the general solution, Eq. (11), over a statistical ensemble chosen to model the bandwidths. An especially useful model is that of phase diffusion,[2-6] in which the field amplitude $\mathscr{E}_L$ is constant, but the phase suffers abrupt changes at an average rate $2\Gamma_L$ (see Appendix B). The field autocorrelation functions are then

$$\langle\langle E_L(\tau)E_L^*(\tau')\rangle\rangle = \mathscr{E}_L^2 e^{-\Gamma_L|\tau-\tau'|} \qquad (15a)$$

and

$$\langle\langle E_S(0,\tau)E_S^*(0,\tau')\rangle\rangle = \mathscr{E}_{S0}^2 e^{-\Gamma_S|\tau-\tau'|}, \qquad (15b)$$

which lead directly to Lorentzian line shapes with halfwidths (HWHM) equal to Γ_L for the pump laser and Γ_S for the input Stokes wave. The brackets $\langle\langle\ \rangle\rangle$ indicate an ensemble average over the statistical fluctuations of the field.[31] This model describes a stabilized laser operating far above threshold, but it also proves to be very convenient mathematically.

The intensity of the output Stokes wave is given by $\langle\langle|E_S(z,\tau)|^2\rangle\rangle$. To evaluate the intensity of the Stokes wave we first introduce some notation:

$$f(x) = (e^{-\Gamma x}/\sqrt{x})I_1[(\alpha z x)^{1/2}], \qquad (16a)$$

$$F(\tau) = \int_0^\tau f(\tau - \tau')\frac{E_L(\tau)E_L^*(\tau')}{\mathscr{E}_L^2}\frac{E_S(0,\tau')}{\mathscr{E}_{S0}}d\tau'. \qquad (16b)$$

Then from Eq. (11), using $p(\tau) = \mathscr{E}_L^2\tau$ in accordance with the phase diffusion model for the pump laser, we have

$$E_S(z,\tau) = E_S(0,\tau) + \mathscr{E}_{S0}\{\tfrac{1}{2}[(\alpha z)^{1/2}]\}F(\tau), \qquad (17a)$$

$$\langle\langle|E_S(z,\tau)|^2\rangle\rangle = \mathscr{E}_{S0}^2 + \mathscr{E}_{S0}(\alpha z)^{1/2}$$
$$\times\langle\langle E_S^*(0,\tau)F(\tau)\rangle\rangle$$
$$+ \mathscr{E}_{S0}^2[\tfrac{1}{4}(\alpha z)]\langle\langle|F(\tau)|^2\rangle\rangle. \qquad (17b)$$

The second term in Eq. (17b) can be easily evaluated using Eq. (15),

$$\langle\langle E_S^*(0,\tau)F(\tau)\rangle\rangle = \mathscr{E}_{S0}\int_0^\tau f(\tau-\tau')$$
$$\times e^{-\Gamma_L(\tau-\tau')}e^{-\Gamma_S(\tau-\tau')}d\tau', \qquad (18)$$

where we have assumed statistical independent of the waves at $z=0$; that is

$$\langle\langle E_L(\tau)E_L^*(\tau')E_S(0,\tau')E_S^*(0,\tau)\rangle\rangle$$
$$= \langle\langle E_L(\tau)E_L^*(\tau')\rangle\rangle\langle\langle E_S(0,\tau')E_S^*(0,\tau)\rangle\rangle$$

(see Ref. 32). This integral is identical to the integral in Eq. (12), but with Γ replaced by $\Gamma + \Gamma_L + \Gamma_S$. Thus when $\Gamma_L + \Gamma_S \gg \Gamma$, this term grows with a very small gain coefficient. This is in
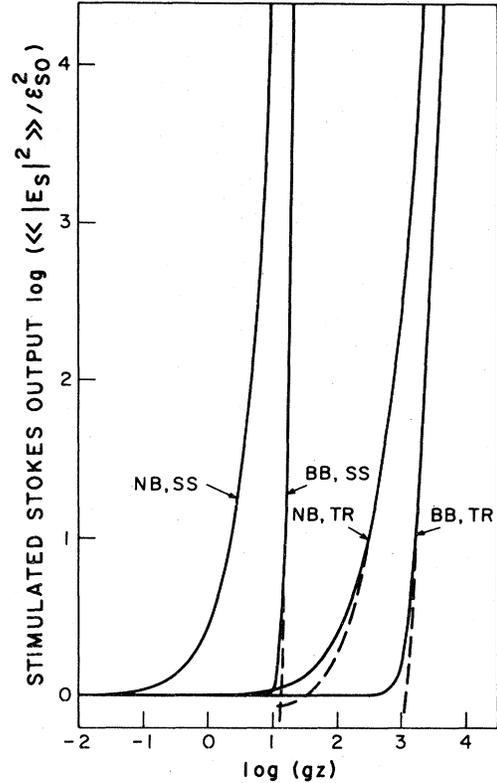

FIG. 2. Normalized Stokes output intensity as a function of gain coefficient gz (or equivalently, pump laser intensity) under various physical conditions and differing levels of approximation. The curve labeled "NB, SS" is the narrow-band steady-state result obtained from Eq. (14a), or Eq. (12) with $\Gamma\tau = 10^2$, where Γ is the Raman linewidth and τ is the laser pulse length. The "NB, TR" curves are the narrow-band transient results, obtained exactly from Eq. (12) with $\Gamma\tau = 10^{-2}$ (solid curve), or approximately from Eq. (13) (dashed curve). The curves labeled "BB, SS" are the broad-band steady-state results obtained exactly from Eqs. (17b), (18), and (20b) with $\Gamma\tau = 10^2$ (solid curve), or approximately from Eq. (25) (dashed curve). The "BB, TR" curves are the broad-band transient results obtained exactly from Eqs. (17b), (18), and (20b) with $\Gamma\tau = 10^{-2}$ (solid curve), or approximately from Eq. (22) (dashed curve). The broad-band curves are for a bandwidth ratio $(\Gamma_L + \Gamma_S)/\Gamma = 10^5$, where Γ_L and Γ_S are the bandwidths of the pump and probe (Stokes) lasers.

contrast to the third term in Eq. (17b), which, as we will see, grows with essentially the narrowband gain given in Eq. (14a). Thus we expect the third term of Eq. (17b) to be dominant in the high-gain limit. Using the correlation functions given by Eq. (15), one can evaluate this third term as

$$\langle\langle|F(\tau)|^2\rangle\rangle = \int_0^\tau \int_0^\tau f(\tau-\tau')f(\tau-\tau'')\frac{\langle\langle E_L^*(\tau')E_L(\tau'')\rangle\rangle\langle\langle E_S(0,\tau')E_S^*(0,\tau'')\rangle\rangle}{\mathcal{E}_L^2 \qquad \mathcal{E}_{S0}^2}d\tau'\,d\tau'' \qquad (19a)$$

$$= \int_0^\tau \int_0^\tau f(\tau-\tau')f(\tau-\tau'')e^{-(\Gamma_L+\Gamma_S)|\tau'-\tau''|}\,d\tau'\,d\tau'', \qquad (19b)$$

where we used the fact that $E_L(\tau)E_L^*(\tau) = \mathcal{E}_L^2$ is independent of the statistical averaging. To evaluate the double integral in Eq. (19b), we note that the exponential factor is much different from zero only near the line $\tau'=\tau''$. In the limit that $f(x)$ changes very slowly in a time $(\Gamma_L+\Gamma_S)^{-1}$ (i.e., Γ, $\alpha z \ll \Gamma_L+\Gamma_S$), we can replace the exponential factor by the properly normalized δ function $2(\Gamma_L+\Gamma_S)^{-1}\delta(\tau'-\tau'')$. We then get

$$\langle\langle|F(\tau)|^2\rangle\rangle \cong \frac{2}{\Gamma_L+\Gamma_S}\int_0^\tau f^2(\tau-\tau')\,d\tau' \qquad (20a)$$

$$= \frac{2}{\Gamma_L+\Gamma_S}\int_0^\tau \frac{e^{-2\Gamma x}}{x}I_1^2[(\alpha z x)^{1/2}]\,dx. \qquad (20b)$$

In similar fashion to the integral in Eq. (12), this integral can be evaluated analytically as well as numerically, in the two limits:

1. Transient limit

As before, for $\Gamma\tau \ll 1$, the exponential can be replaced by 1 and the integral done (in this case asymptotically, using the asymptotic form for I_1) to give

$$\langle\langle|F(\tau)|^2\rangle\rangle = \frac{1}{\pi(\Gamma_L+\Gamma_S)}\frac{e^{2(\alpha z\tau)^{1/2}}}{\alpha z\tau}. \qquad (21)$$

Thus, in the high-gain limit where Eq. (17b) is dominated by the last term, we find that the Stokes output intensity in the transient limit is

$$\langle\langle|E_S(z,\tau)|^2\rangle\rangle = \frac{\mathcal{E}_{S0}^2}{2\pi}\frac{e^{2(\alpha z\tau)^{1/2}}}{2(\Gamma_L+\Gamma_S)}$$

$$\text{(high gain, } \Gamma\tau \ll 1). \qquad (22)$$

Because of the form of the exponential, this result for the broad-band transient will be nearly indistinguishable from the result, Eq. (13), for the narrowband transient.

2. Steady-state limit

To evaluate the steady-state limit of Eq. (20b), we extend the upper limit of the integration to infinity, and do the integral to give[33]

$$\langle\langle|F(\infty)|^2\rangle\rangle = \frac{\alpha z}{4\Gamma(\Gamma_L+\Gamma_S)}{}_2F_2\left(\tfrac{3}{2},1;2,3,\frac{\alpha z}{2\Gamma}\right), \qquad (23)$$

where $_2F_2$ is the generalized hypergeometric function, which can be evaluated asymptotically for large argument as[34]

$$_2F_2(\tfrac{3}{2},1;2,3,x) \to (4/\sqrt{\pi})\,e^x/x^{5/2}. \qquad (24)$$

This leads to an asymptotic form for the Stokes intensity under broad-band steady-state high-gain conditions:

$$\langle\langle|E_S(z,\infty)|^2\rangle\rangle = \mathcal{E}_{S0}^2\frac{\Gamma}{\Gamma_L+\Gamma_S}\frac{e^{gz}}{(\pi gz)^{1/2}}$$

$$\text{(high gain, } \Gamma\tau \gg 1). \qquad (25)$$

It can be seen by comparing the Stokes intensities given by Eqs. (25) and (14a) that under the conditions assumed (copropagating waves, no dispersion), the growth of a Stokes wave, in the steady-state high-gain limit, is virtually unaffected by either its input bandwidth or that of the pump laser. We can demonstrate this result by writing the output Stokes intensity as

$$\langle\langle|E_S(z,\infty)|^{22}\rangle\rangle = \mathcal{E}_{S0}^2 e^G, \qquad (26)$$

$$G = G_{NB} - \ln\{[(\Gamma_L+\Gamma_S)/\Gamma](\pi G_{NB})^{1/2}\}, \qquad (27)$$

where $G_{NB} = gz$ is the narrow-band gain coefficient from Eq. (14b). Thus for large gain the difference between G and G_{NB} becomes relatively insignificant. We will present a possible interpretation for this result in Sec. V. Equation (27) is similar to the result, conjectured by Carman et al.,[18] that $G = G_{NB} - \ln[(\Gamma_L/\Gamma)G_{NB}]$. The difference between our result and theirs (when $\Gamma_S = 0$) may be due to the fact that they allowed also for amplitude fluctuations of the pump laser, whereas we have restricted ourselves to phase modulation alone, in order to make an explicit calculation tractible. The calculation becomes intractible when amplitude fluctuations are present because, then, $p(\tau)$ in Eq. (11) is a random variable which makes the statistical average difficult to perform.

As in the narrow-band case, we plot the broad-band Stokes intensities, Eqs. (22) and (25), in Fig. 2, along with numerical evaluation of the Stokes intensity from $\langle\langle|E_S(z,\tau)|^2\rangle\rangle$, defined by Eqs. (17b), (18), and (20b). Since Eq. (20b) is valid only for $\Gamma_L+\Gamma_S \gg \Gamma$, αz, we have plotted the extreme case that $(\Gamma_L+\Gamma_S)/\Gamma = 10^5$, in order to demonstrate the validity of the asymptotic forms Eqs. (22) and (25). Again we have plotted the transient and steady-state cases: $\Gamma\tau = 10^{-2}$ and 10^2. Again we see agreement of the asymptotic forms with the exact numerical results when $\log\langle\langle|E_S(z,\tau)|^2\rangle\rangle/\mathcal{E}_{S0}^2 > 1$.

We may now compare the narrow-band and broad-band results. For this extreme steady-state case, $(\Gamma_L+\Gamma_S)/\Gamma = 10^5$, we see a significant sup-

pression of the gain in the turn-on region for both transient and steady-state limits. However, according to Eq. (27), at very high gains the difference between the narrow- and broad-band stimulated outputs becomes less and less, relative to their absolute magnitudes. It is interesting that the broad-band output exhibits a threshold-type behavior, in contrast to the exponential behavior of the narrow-band output.

Although the principal interest here is in the high-gain limit, some comments can also be made about the low-gain steady-state limit, important to such experimental techniques as CARS (coherent anti-Stokes Raman scattering). Thus we will find the terms in Eq. (17b) which are of lowest order (linear) in αz. It can be shown[34] that the third term, containing $\langle\langle |F(\tau)|^2 \rangle\rangle$, is quadratic in αz as $\alpha z \to 0$ and can thus be neglected. The first and second terms in Eq. (17b) are evaluated, using Eq. (18) with the upper limit τ taken to infinity, to give[29]

$$\langle\langle |E_S(z,\infty)|^2 \rangle\rangle = \mathcal{E}_{S0}^2 (2e^{g_{BB}\varepsilon/2} - 1), \qquad (28)$$

$$g_{BB} = \alpha/2(\Gamma + \Gamma_L + \Gamma_S). \qquad (29)$$

In the limit $\alpha z \to 0$, this reduces to

$$\langle\langle |E_S(z,\infty)|^2 \rangle\rangle = \mathcal{E}_{S0}^2 (1 + g_{BB}z)$$

$$\text{(low gain, } \Gamma\tau \gg 1). \qquad (30)$$

We see that in the low-gain steady-state limit, the SRS grows linearly with the ("broad-band") gain coefficient g_{BB}. This is the result that would naively be predicted on the basis of photons per mode, as discussed in Sec. I.

C. Raman gain for arbitrary bandwidths

Here we analyze, numerically, the properties of the stimulated output when the condition $\Gamma_L + \Gamma_S \gg \Gamma$, αz is *not* necessarily upheld, as was assumed in Sec. III B. First note that if we take $(\Gamma_L + \Gamma_S)/\Gamma = 10^2$, rather than 10^5 as used in Fig. 2, the analysis of Sec. III B is valid only for $gz < 10^2$, making prediction of the broad-band transient above $gz = 10^2$ impossible by those methods. However, also note that the solution in Eq. (19b), before approximation to obtain Eq. (20), contains the information we are seeking in the general case. Thus we evaluated Eq. (19b), by a numerical method discussed in Appendix C, and obtained the output Stokes intensity $\langle\langle |E_S(z,\tau)|^2 \rangle\rangle$ defined by Eqs. (17b), (18), and (19b). These results are shown in Fig. 3, where we have covered a large region of the interesting parameters: $\Gamma\tau$ and $(\Gamma_L + \Gamma_S)/\Gamma$ both vary between 10^{-2} and 10^2.

Beginning with Fig. 3(a), we see that the laser bandwidth has little effect on the gain in the transient limit ($\Gamma\tau = 10^{-2}$). This is not surprising,

as a short laser pulse of duration $\tau = 10^{-2}/\Gamma$ has a spectral width of $10^2\Gamma$ without phase diffusion ($\Gamma_L = 0$). Thus we see no effect of additional broadening by phase diffusion until $\Gamma_L + \Gamma_S \gtrsim 10^2\Gamma$, at which point the gain becomes slightly depressed. Progressing in Figs. 3(b) and 3(c) to longer pulse duration τ, we see the general result that no effect of phase diffusion broadening is apparent until $\Gamma_L + \Gamma_S \gtrsim 1/\tau$. The steady-state limit occurs in Fig. 3(d), where no difference is seen between $\Gamma\tau = 10$ and $\Gamma\tau = 10^2$.

IV. SPECTRUM OF THE STOKES OUTPUT IN STEADY STATE

In Secs. I–III the input Stokes wave has been taken to have a width Γ_S. But because all of the power in the broad-band pump laser is effective for amplifying the Stokes wave, it is interesting to ask what becomes of the spectral distribution of the Stokes wave after it has been amplified. In this section we calculate the spectrum of the Stokes wave in the steady-state high-gain limit, in two different cases: $\Gamma_L = 0$ and $\Gamma_L \gg \Gamma$.
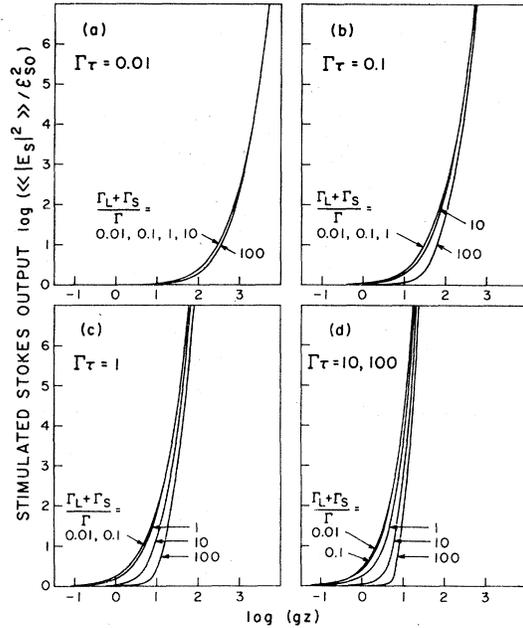

FIG. 3. Normalized Stokes output intensity as a function of gz (or pump-laser intensity), evaluated numerically using Eqs. (17b), (18), and (19b), for various values of the bandwidth ratio $(\Gamma_L + \Gamma_S)/\Gamma$, where Γ is the Raman linewidth, and Γ_L and Γ_S are the bandwidths, due to phase diffusion broadening, of the pump and probe (Stokes) lasers. Four different pulse lengths τ are shown: (a) $\Gamma\tau = 10^{-2}$, transient limit; (b) $\Gamma\tau = 10^{-1}$; (c) $\Gamma\tau = 1$; (d) $\Gamma\tau = 10$ and 10^2, steady-state limit. In all cases the effect of the laser bandwidths becomes relatively unimportant at high gains.

For a stationary wave $E(\tau)$, the definition of the power spectrum $P(\omega)$ is

$$P(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\omega s} K(s)\, ds \,, \tag{31}$$

where ω is the frequency as measured from the frequency of the carrier wave and $P(\omega)$ is the Fourier transform of the electric field autocorrelation function $K(s)$,

$$K(s) = \langle\langle E(\tau) E^*(\tau + s)\rangle\rangle \,. \tag{32}$$

It is easy to show from Eq. (31) that $P(\omega)$ is normalized as follows:

$$\int_{-\infty}^{\infty} P(\omega)\, d\omega = K(0) = \langle\langle |E(\tau)|^2\rangle\rangle \,. \tag{33}$$

As an example, when Eq. (31) is used to calculate the laser spectrum $P_L(\omega)$, from the correlation function Eq. (15), one finds

$$P_L(\omega) = \frac{\Gamma_L/\pi}{\omega^2 + \Gamma_L^2}\, \mathcal{E}_L^2 \,, \tag{34}$$

a Lorentzian, as stated in Sec. III. We can use the general solution, Eq. (11), to determine $K(s)$ for the Stokes wave

$$K(s) \simeq \mathcal{E}_{S0}^2 [\tfrac{1}{4}(\alpha z)] \langle\langle F(\tau) F^*(\tau + s)\rangle\rangle \,, \tag{35}$$

where we have again kept only the term which dominates in the high-gain limit. In steady state $(\tau \to \infty)$ we expect that $K(s)$ will depend only on s and not τ. $K(s)$ can be evaluated as

$$K(s) = \mathcal{E}_{S0}^2 \frac{\alpha z}{4} \int_0^\tau d\tau' \int_0^{\tau+s} d\tau'' f(\tau - \tau') f(\tau + s - \tau'') \frac{\langle\langle E_L(\tau) E_L^*(\tau') E_L^*(\tau+s) E_L(\tau'')\rangle\rangle}{\mathcal{E}_L^4} \frac{\langle\langle E_S(0,\tau') E_S^*(0,\tau'')\rangle\rangle}{\mathcal{E}_{S0}^2} \tag{36a}$$

$$= \mathcal{E}_{S0}^2 \frac{\alpha z}{4} \int_0^\tau dx \int_0^{\tau+s} dy\, f(x) f(y) G(x,y,s) e^{-\Gamma_S |s-y+x|} \,, \tag{36b}$$

where

$$G(x,y,s) = \exp[\Gamma_L(|s+x| + |s-y| - |s-y+x| - |x| - |y| - |s|)] \,. \tag{36c}$$

Here $G(x,y,s)$ is the four-time correlation function of the pump-laser field, assuming the phase diffusion model, and is evaluated in Appendix B. In deriving Eq. (36b) we have used $x = \tau - \tau'$ and $y = \tau + s - \tau''$. In steady state the upper integration limits are extended to infinity and $K(s)$ becomes independent of τ. In order to simplify the absolute values, the integral is transformed to the triangular region above the $y = x$ line by use of the property $G(y,x,s) = G(x,y,-s)$. Then for the Stokes spectrum we have

$$P_S(\omega) = \frac{1}{2\pi} \mathcal{E}_{S0}^2 \frac{\alpha z}{4}$$

$$\times \int_0^\infty dy \int_0^y dx\, f(x) f(y) \mathcal{L}(x,y,\omega) \,, \tag{37a}$$

where

$$\mathcal{L}(x,y,\omega) = 2\,\mathrm{Re}\, e^{-\Gamma_L(y+x)} \int_{-\infty}^{\infty} ds\, e^{-i\omega s}$$

$$\times \exp[\Gamma_L(|s+x| + |s-y| - |s-y+x| - |s|) - \Gamma_S |s-y+x|] \,. \tag{37b}$$

The transform $\mathcal{L}$ can be calculated under the condition $x \leqslant y$. We write $\mathcal{L}$ as the sum of two parts $\mathcal{L} = \mathcal{L}_1 + \mathcal{L}_2$. Defining

$$A_{\pm} = \frac{i\omega + (2\Gamma_L + \Gamma_S)}{\omega^2 + (2\Gamma_L + \Gamma_S)^2} \pm \frac{i\omega + \Gamma_S}{\omega^2 + \Gamma_S^2} \,, \tag{38a}$$

we can write

$$\mathcal{L}_1 = 2\,\mathrm{Re}\, A_- e^{-(\Gamma_L + \Gamma_S)(y-x)}$$

$$+ 2\,\mathrm{Re}\, A_+ e^{-\Gamma_L(y-x)} \cos\omega(y-x)$$

$$- 2\,\mathrm{Im}\, A_- e^{-\Gamma_L(y-x)} \sin\omega(y-x) \,, \tag{38b}$$

$$\mathcal{L}_2 = 2(-\mathrm{Re}\, A_- \cos\omega x + \mathrm{Im}\, A_- \sin\omega x)$$

$$\times e^{-\Gamma_L(y+x) - \Gamma_S y}$$

$$- 2(\mathrm{Re}\, A_- \cos\omega y + \mathrm{Im}\, A_- \sin\omega y)$$

$$\times e^{-\Gamma_L(y+x) - \Gamma_S x} \,. \tag{38c}$$

Equations (37) and (38) are now used to evaluate the Stokes output spectrum in two different cases.

A. Stokes output spectrum for a monochromatic pump laser

Here we treat the case that the pump laser is monochromatic $(\Gamma_L = 0)$ and the spectral width of the input Stokes is allowed to assume two different limits: $\Gamma_S = 0$ or $\Gamma_S \gg \Gamma$. The steady-state gain for these two limits has already been given in Eqs. (14) and (25).

The spectrum is easily obtained by setting $\Gamma_L = 0$ in Eq. (38). Then, because $A_- = 0$, we have $\mathcal{L}_2 = 0$ and

$$\mathcal{L}_1 = 4[\Gamma_S/(\omega^2 + \Gamma_S^2)] \cos\omega(y-x) \,. \tag{39}$$

Transforming back to the full x,y quadrant gives for the spectrum of the Stokes output

$$P_S(\omega) = \frac{1}{2\pi} \mathcal{E}_{S0}^2 \frac{\alpha z}{4} \frac{4\Gamma_S}{\omega^2 + \Gamma_S^2}$$

$$\times \frac{1}{2} \int_0^\infty dy \int_0^\infty dx f(x) f(y) \cos\omega(y - x) \qquad (40a)$$

$$= \mathcal{E}_{S0}^2 \frac{\alpha z}{4} \frac{\Gamma_S/\pi}{\omega^2 + \Gamma_S^2} |I|^2 , \qquad (40b)$$

where[29]

$$I = \int_0^\infty e^{i\omega x} f(x)\, dx$$

$$= \frac{2}{(\alpha z)^{1/2}} \left[\exp\!\left(\frac{gz}{2} \frac{\Gamma}{\Gamma - i\omega}\right) - 1 \right] . \qquad (40c)$$

In the high-gain limit the Stokes output spectrum is

$$P_S(\omega) = \mathcal{E}_{S0}^2 \frac{\Gamma_S/\pi}{\omega^2 + \Gamma_S^2} \exp\!\left(gz \frac{\Gamma^2}{\omega^2 + \Gamma^2}\right). \qquad (41)$$

To put this result into proper form we must assume one of the two above-mentioned limits.

The first limit is that of a monochromatic input Stokes wave ($\Gamma_S = 0$). Here we can use Eq. (14a) for the output Stokes intensity: $\langle\langle |E_S(z, \infty)|^2 \rangle\rangle$ $= \mathcal{E}_{S0}^2 e^{gz}$. Then Eq. (41) can be rewritten, in the limit $\Gamma_S \to 0$,

$$P_S(\omega) = \delta(\omega)\langle\langle |E_S(z, \infty)|^2 \rangle\rangle . \qquad (42)$$

Thus we see that the output Stokes wave is monochromatic when the input Stokes wave and the pump laser are monochromatic, as expected.

The second limit is $\Gamma_S \gg \Gamma$. Here we again consider a monochromatic pump laser ($\Gamma_L = 0$) and use Eq. (25) for the Stokes output $\langle\langle |E_S(z, \infty)|^2 \rangle\rangle$ to rewrite Eq. (41) for the Stokes output spectrum as

$$P_S(\omega) = \frac{\Gamma_S^2}{\omega^2 + \Gamma_S^2} \left(\frac{gz}{\pi\Gamma^2}\right)^{1/2} \exp\!\left(-gz \frac{\omega^2}{\omega^2 + \Gamma^2}\right)$$

$$\times \langle\langle |E_S(z, \infty)|^2 \rangle\rangle , \qquad (43a)$$

$$P_S(\omega) \cong \left(\frac{gz}{\pi\Gamma^2}\right)^{1/2} \exp\!\left(-\frac{gz}{\Gamma^2} \omega^2\right) \langle\langle |E_S(z, \infty)|^2 \rangle\rangle , \qquad (43b)$$

where the last step is valid because the halfwidth $[(\ln 2)\Gamma^2/gz]^{1/2}$ of the exponential factor is much less than Γ and Γ_S. Note that Eq. (43b) is normalized as in Eq. (33). Equation (43) describes a Lorentzian-shaped atomic line of width Γ that has been gain narrowed. The ideal that the center of the line will experience more gain than the wings is a familiar idea in laser theory. A comparison of the atomic line and the gain-narrowed Stokes line is shown in Fig. 4 for the case $gz = 15$.

B. Stokes output spectrum for a broad-band pump laser

Here we treat the case that the pump laser width is larger than the atomic width ($\Gamma_L \gg \Gamma$) and the input Stokes width Γ_S is arbitrary. The spectrum is obtained by applying several approximations to Eq. (38). First note that in the high-gain limit, only $\mathcal{L}_1$ will give a significant contribution to the spectrum because its exponentials damp as $(y - x)$, rather than $(y + x)$. Thus along the line $y = x$, $\mathcal{L}_1$ is large, while $\mathcal{L}_2$ becomes negligible. Second, note that because $\Gamma_L + \Gamma_S$ is assumed large ($\Gamma_L + \Gamma_S \gg \Gamma$, αz), we may replace the exponential factors by properly normalized delta functions, as in the argument leading to Eq. (20);

$$e^{-(\Gamma_L + \Gamma_S)(y-x)} \to (\Gamma_L + \Gamma_S)^{-1} \delta(y - x) , \qquad (44a)$$

$$e^{-\Gamma_L (y-x)} \cos\omega(y - x) \to [\Gamma_L/(\omega^2 + \Gamma_L^2)]\delta(y - x) , \qquad (44b)$$

$$e^{-\Gamma_L (y-x)} \sin\omega(y - x) \to [\omega/(\omega^2 + \Gamma_L^2)]\delta(y - x) . \qquad (44c)$$
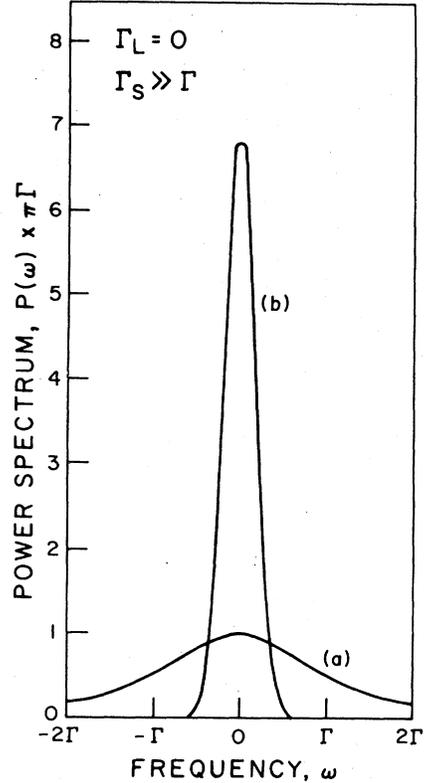
FIG. 4. Comparison of the Lorentzian Raman line shape [curve (a)] with atomic halfwidth Γ, and the gain-narrowed output Stokes spectrum [curve (b)] with halfwidth $(\ln 2\, \Gamma^2/gz)^{1/2}$ for $gz = 15$, plotted from Eq. (43b). This Stokes spectrum narrowing results when the pump laser is monochromatic and the input Stokes laser is broad band.

Combining these results gives

$$\mathcal{L}_1 \to [4/(\Gamma_L + \Gamma_S)][\Gamma_L/(\omega^2 + \Gamma_L^2)]\delta(y - x). \quad (45)$$

Now the spectrum of the output Stokes intensity $P_S(\omega)$ is easily evaluated from Eq. (37a):

$$P_S(\omega) = \frac{1}{2\pi}\, \mathcal{E}_{S0}^2\, \frac{\alpha z}{4}\, \frac{4}{\Gamma_L + \Gamma_S}\, \frac{\Gamma_L}{\omega^2 + \Gamma_L^2}$$

$$\times \int_0^\infty f^2(y)\,dy\,, \quad (46a)$$

$$P_S(\omega) = \frac{\Gamma_L/\pi}{\omega^2 + \Gamma_L^2}\, \langle\langle |E_S(z, \infty)|^2 \rangle\rangle\,, \quad (46b)$$

where we used Eqs. (17b) and (20a) to define the output Stokes intensity $\langle\langle |E_S(z, \infty)|^2 \rangle\rangle$. Note that $P_S(\omega)$ is normalized as required by Eq. (33). This indicates that our neglect of $\mathcal{L}_2$ is justified.

Comparing Eqs. (46b) and (34) shows that the Stokes wave assumes *exactly* the same spectrum as the pump laser during the amplification process, regardless of the spectral width of the input Stokes wave.

V. DISCUSSION

In the case just treated, that the pump laser width is greater than the atomic width ($\Gamma_L \gg \Gamma$), our interpretation is that the fluctuations in the phase of the pump laser dominate the time behavior of the amplification process. Indeed, Carman et al.[18] found numerically, in the case of a quadratic phase sweep in the pump laser, that the Stokes phase closely followed this sweep after a brief initial period. Our result for the Stokes spectrum [Eq. (46)] is *consistent* with the conjecture of Carman et al. that, when $\Gamma_L \gg \Gamma$, the Stokes phase always follows the pump phase in the high-gain limit, regardless of the phase structure of the input Stokes wave. If correct, this effect, which we will call "phase locking," also explains the fact that the gain is unaffected by the phase flucutations which lead to the bandwidth. As Carman et al. pointed out, if the phases φ_S and φ_L differ at all points by a constant $\lfloor\varphi_S(z, t) = \varphi_L(z, t) + \varphi_0\rfloor$, the phases drop out entirely from Eq. (11), leading to the narrow-band gain result, Eq. (14). Thus, the idea of "phase locking" leads to results consistent with our results for $\Gamma_L \gg \Gamma$. When both Γ_L and Γ_S are larger than Γ, we can say that the amplified Stokes wave builds up from the broadband input noise in a way which automatically satisfies $\varphi_S = \varphi_L + \varphi_0$. That is, only that part of the noise which satisfies this relation will experience large gain.

To illustrate the idea of phase locking we have compared, in Fig. 5, several steady-state gain curves. We have reproduced curves from Fig.

3(d), calculated from the exact equations (labeled "phase locked"). We have also plotted curves using Eq. (14a) (labeled "narrow band"), and also using Eq. (14a) with g replaced by g_{BB} of Eq. (29) (labeled "unlocked"). We see that at low gains the exact curve follows the "unlocked" curve, consistent with the idea that there is no correlation between the output Stokes and pump laser waves. This low-gain behavior was predicted at the end of Sec. III B. However, at high gains the exact curve approaches the "narrow-band" curves, consistent with the idea that it has become "phase locked," resulting in an enhanced gain. We thus see that phase locking appears to occur only above a certain (threshold) gain. In contrast to the behavior found in the present treatment, Dzhotyan et al.[22] found in the multimode approach (see Sec. I) that the "narrow-band" gain was appropriate even at low gains. This is a major difference between the two models.

Finally, we point out that we have treated only the case of Raman amplification, and not SRS which grows from the initial Stokes photons spontaneously emitted with frequencies near ω_S, in the absence of an external input Stokes wave at that frequency. Here we wish to make some conjectures on the outcome in the latter case. We may consider the spontaneous photons as making up the source term $E_S(0, \tau)$. Although here we certainly cannot make the decorrelation of the pump wave $E_L(\tau)$ and source term $E_S(0, \tau)$ that we made in connection with Eq. (18), we still expect that, at high gains, the major results we have obtained do apply to spontaneously generated SRS. That is, we expect the gain to be essentially in-
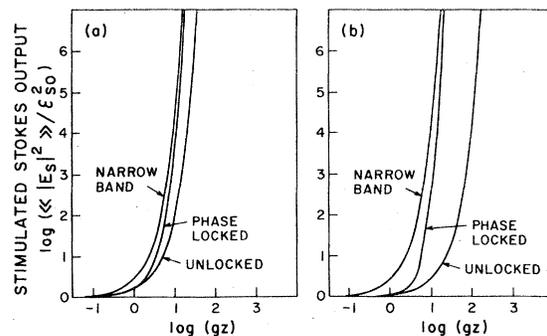

FIG. 5. Normalized Stokes output intensity, in steady-state, as a function of gz (or pump-laser intensity), for two different laser bandwidth ratios: (a) $(\Gamma_L + \Gamma_S)/\Gamma = 1$ and (b) $(\Gamma_L + \Gamma_S)/\Gamma = 10$. In both cases, the exact results (labeled "phase locked") are seen to agree, at low gains, with the results one would expect in the absence of phase locking (labeled "unlocked"), which were obtained by replacing Γ by $\Gamma + \Gamma_L + \Gamma_S$ in the expression [Eq. (14b)] for the gain coefficient g. However, at higher gains, the exact results approach the narrow-band curve [Eqs. (14a) and (14b)].

dependent of pump-laser bandwidth, and when the pump bandwidth is greater than the atomic width we expect the output SRS to assume the spectrum of the pump.

VI. SUMMARY

Using the phase diffusion model, we have extended the theory of stimulated Raman scattering, in the case of two interacting classical waves (pump and input Stokes), to allow for arbitrary bandwidth of either wave. In the forward direction if there is no dispersion, we showed that, in the high-gain limit, the gain of the Stokes wave is essentially independent of the input bandwidth of either wave. In the low-gain limit the gain coefficient was found to be inversely proportional to the sum of the bandwidths. We also calculated the spectrum of the output Stokes wave, in the high-gain limit, under various conditions. We found that when the pump bandwidth Γ_L is greater than the atomic width Γ, the Stokes wave assumes exactly the spectrum of the pump laser, regardless of the spectral width Γ_S of the input Stokes wave. When both input waves are monochromatic ($\Gamma_L, \Gamma_S = 0$), we found that the Stokes spectrum is unchanged by the amplification process. Finally, when $\Gamma_L = 0$ and $\Gamma_S \gg \Gamma$, we found that the output Stokes wave has a spectrum which is a gain-narrowed atomic profile; that is, the Stokes width becomes much narrower than the atomic width.

Note added in proof. A recent preprint by W. R. Trutna, Y. K. Park, and R. L. Byer [to appear in IEEE J. Quant. Electron. (July 1979)] has come to our attention. Broad-band SRS was treated using the coupled-wave approach (similar to that in Ref. 22) and qualitative agreement was found with our work in the high-gain limit. At low gains, however, their treatment indicates no suppression of the gain, in contrast to our results [Eq. (30)].

ACKNOWLEDGMENTS

APPENDIX A

Here we discuss more carefully the elimination of a_2 from Eq. (3). We first neglect $\dot{\varphi}_L$ in Eq. (3b), as Δ_L is assumed to be much larger than the pump laser bandwidth. The formal solution of Eq. (3b) can then be written

$$a_2(t) = \int_0^t e^{-i\Delta_L(t-t')} g(t') \, dt', \tag{A1}$$

$$g(t) = i[\tfrac{1}{2}\Omega_L a_1(t) + \tfrac{1}{2}\Omega_S a_3(t)]. \tag{A2}$$

Repeated integration by parts gives

$$a_2(t) = \frac{g(t) - e^{-i\Delta_L t}g(0)}{i\Delta_L} - \frac{\dot{g}(t) - e^{-i\Delta_L t}\dot{g}(0)}{(i\Delta_L)^2}$$

$$+ \frac{\ddot{g}(t) - e^{-i\Delta_L t}\ddot{g}(0)}{(i\Delta_L)^3} - \cdots. \tag{A3}$$

Note that since $a_1(0) = 1$ and $a_2(0) = 0$, $g(0) = i(\tfrac{1}{2})\Omega_L$. When one assumes $\Delta_L \gg \Delta_S, \dot{\varphi}_L, \dot{\varphi}_S, \Omega_L, \Omega_S$, it can be shown from Eq. (3) that $\dot{g}(t) \ll \Delta_L g(t)$. Thus when Δ_L is large, one is left with

$$a_2(t) \cong [g(t) - i\tfrac{1}{2}\Omega_L e^{-i\Delta_L t}]/i\Delta_L. \tag{A4}$$

However, because Δ_L is large, the exponential term oscillates rapidly compared to $g(t)$. Hence, in the spirit of the RWA, we neglect the rapidly oscillating part and retain only the slowly varying part:

$$a_2(t) \cong g(t)/i\Delta_L = (\tfrac{1}{2}\Omega_L a_1(t) \tfrac{1}{2}\Omega_S a_3(t))/\Delta_L. \tag{A5}$$

It is interesting that the same result is obtained by merely setting $\dot{a}_2 = 0$ in Eq. (3b).

APPENDIX B: PHASE-DIFFUSION MODEL

The phase-diffusion model for laser bandwidth describes, to good approximation, a cw laser operating well above threshold, where the intensity, $I(t) = I_0 + I'(t)$, is nearly constant, with average value I_0 and small fluctuations $I'(t)$.[35] However, well above threshold the phase $\varphi(t)$ fluctuates randomly, in a way reminiscent of a diffusing Brownian particle. Simple laser theory gives the equations for the intensity and phase as[36]

$$\dot{I}'(t) = -\lambda I'(t) + F_I(t), \tag{B1}$$

$$\dot{\varphi}(t) = F_\varphi(t), \tag{B2}$$

where $F_I(t)$ and $F_\varphi(t)$ are random Langevin forces with correlation functions

$$\langle\!\langle F_I(t_1)F_I(t_2)\rangle\!\rangle = 2D\delta(t_1 - t_2),$$

$$\langle\!\langle F_\varphi(t_1)F_\varphi(t_2)\rangle\!\rangle = 2\Gamma\delta(t_1 - t_2),$$

and

$$\langle\!\langle F_I(t_1)F_\varphi(t_2)\rangle\!\rangle = 0.$$

Here $1/\lambda$ is the correlation time of the intensity fluctuations, with mean value D/λ, and Γ is the bandwidth of the light. These δ correlations simply imply that the forces fluctuate on a time scale shorter than any other interesting time scale.

The phase-diffusion model is based on the assumption that the intensity exhibits no fluctuations, $I'(t) = 0$, and that the phase fluctuates according to Eq. (B2). The correlation function for the phase

can be derived from Eq. (B2) as

$$\langle\!\langle \varphi(t_1)\varphi(t_2)\rangle\!\rangle = \int_0^{t_1} dt \int_0^{t_2} dt' \langle\!\langle F_\varphi(t)F_\varphi(t')\rangle\!\rangle$$

$$= \Gamma(t_1 + t_2 - |t_1 - t_2|), \qquad (B3)$$

where we have taken $\varphi(0) = 0$, since the results calculated later cannot depend on $\varphi(0)$ for a stationary process. In the present context, the aim of the model is to calculate correlation functions for the field $E(t) = \mathcal{E}\, e^{i\varphi(t)}$, where we are using the notation of Eq. (9). Here we have assumed that the field amplitude $\mathcal{E}$ (and thus the intensity) is a constant. So the correlation functions can be written

$$\langle\!\langle E(t_1)\cdots E(t_m)E^*(t_{m+1})\cdots E^*(t_n)\rangle\!\rangle$$

$$= \mathcal{E}^n \langle\!\langle \exp[i\varphi(t_1)+\cdots+i\varphi(t_m)$$

$$-i\varphi(t_{m+1})-\cdots-i\varphi(t_n)]\rangle\!\rangle. \qquad (B4)$$

In order to calculate these correlations it is expedient to further assume that the phase $\varphi(t)$ is a Gaussian stochastic quantity, that is, correlation functions of any order can be expressed in terms of the two-time correlation function of the phase $\langle\!\langle\varphi(t_1)\varphi(t_2)\rangle\!\rangle$. Specifically,[37] we have

$$\langle\!\langle \varphi(t_1)\cdots\varphi(t_{2n+1})\rangle\!\rangle = 0,$$

$$\langle\!\langle \varphi(t_1)\cdots\varphi(t_{2n})\rangle\!\rangle = \sum_{\text{perm}} \langle\!\langle \varphi(t_{i_1})\varphi(t_{i_2})\rangle\!\rangle\cdots$$

$$\times \langle\!\langle \varphi(t_{i_{2n-1}})\varphi(t_{i_{2n}})\rangle\!\rangle, \qquad (B5)$$

where the summation is taken over all unique permutations of $t_1, \ldots, t_{2n}$. A useful relation can be derived from Eq. (B5), which makes it easy to calculate the correlation functions in Eq. (B4). This is

$$\left\langle\!\!\left\langle \exp\!\left(i\int_{-\infty}^{\infty} dt'\, J(t')\varphi(t')\right)\right\rangle\!\!\right\rangle$$

$$= \exp\left(-\frac{1}{2}\int_{-\infty}^{\infty} dt' \int_{-\infty}^{\infty} dt''\, J(t')J(t'')\right.$$

$$\times \langle\!\langle \varphi(t')\varphi(t'')\rangle\!\rangle\Big), \qquad (B6)$$

where $J(t')$ is an arbitrary function.[38] This relation can be proven, term by term, after expanding the exponentials and using the property Eq. (B5).

We can now calculate the desired correlation functions. By letting $J(t') = \delta(t' - t_1)$ in Eq. (B6) we get

$$\langle\!\langle E(t_1)\rangle\!\rangle = \mathcal{E}\langle\!\langle \exp[i\varphi(t_1)]\rangle\!\rangle$$

$$= \mathcal{E}\exp(-\Gamma t_1) \to 0, \qquad (B7)$$

where we have taken the stationary limit $\Gamma t_1 \to \infty$, where the initial transients have died out. Thus the average field is zero, as expected for a fluc-

tuating field. By letting $J(t') = \delta(t' - t_1) - \delta(t' - t_2)$ in Eq. (B6) we get

$$\langle\!\langle E(t_1)E^*(t_2)\rangle\!\rangle = \mathcal{E}^2\langle\!\langle \exp[i\varphi(t_1)-i\varphi(t_2)]\rangle\!\rangle$$

$$= \mathcal{E}^2\exp(-\Gamma|t_1 - t_2|). \qquad (B8)$$

The power spectrum of the field, given by the Fourier transform of the two-time correlation function in Eq. (B8), is thus a Lorentzian with halfwidth Γ. The four-time correlation function used in Sec. IV can be calculated by letting

$$J(t') = \delta(t' - t_1) + \delta(t' - t_2)$$

$$- \delta(t' - t_3) - \delta(t' - t_4),$$

which gives

$$\langle\!\langle E(t_1)E(t_2)E^*(t_3)E^*(t_4)\rangle\!\rangle$$

$$= \mathcal{E}^4\exp\{\Gamma(|t_1 - t_2| + |t_3 - t_4| - |t_1 - t_3|$$

$$- |t_1 - t_4| - |t_2 - t_3| - |t_2 - t_4|)\}. \qquad (B9)$$

This result can be used to illustrate one of the basic assumptions of the phase-diffusion model. By letting $t_3 = t_1$ and $t_4 = t_2$, and defining the intensity as $I(t) = |E(t)|^2$, we can see from Eq. (B9) that the intensity correlation function is given by

$$\langle\!\langle I(t_1)I(t_2)\rangle\!\rangle = \mathcal{E}^4 = I_0^2, \qquad (B10)$$

i.e., the intensity is always perfectly correlated with itself in the phase-diffusion model, because it does not fluctuate.

APPENDIX C

Here we describe the numerical technique used to evaluate the double integral in Eq. (19b). Let $r = (\tau - \tau')^{1/2}$, $s = (\tau - \tau'')^{1/2}$, and $a = (\alpha z)^{1/2}$. Then, we have

$$\langle\!\langle |F(\tau)|^2\rangle\!\rangle = 4\int_0^{\tau^{1/2}} dr \int_0^{\tau^{1/2}} ds\, e^{-\Gamma r^2}I_1(ar)$$

$$\times e^{-\Gamma s^2}I_1(as)\, e^{-(\Gamma_L+\Gamma_s)|s^2-r^2|} \qquad (C1)$$

$$= 8\int_0^{\tau^{1/2}} dr \int_0^{r} ds\, e^{-\Gamma r^2}I_1(ar)$$

$$\times e^{-\Gamma s^2}I_1(as)\, e^{-(\Gamma_L+\Gamma_s)(r^2-s^2)}, \qquad (C2)$$

where we used the symmetry of the integrand in Eq. (C1) with respect to interchange of r and s. Now defining

$$u(x) = \tfrac{1}{8}\langle\!\langle |F(x^2)|^2\rangle\!\rangle \qquad (C3)$$

and

$$v(x) = e^{-(\Gamma+\Gamma_L+\Gamma_s)x^2}\int_0^x ds\, e^{-(\Gamma-\Gamma_L-\Gamma_s)s^2}I_1(as),$$

$$\qquad (C4)$$

we have

$$u(x) = \int_0^x dr \, I_1(ar)v(r) \,, \qquad \text{(C5)}$$

from which we can obtain

$$u'(x) = I_1(ax)v(x) \,, \qquad \text{(C6)}$$

$$v'(x) = -2(\Gamma + \Gamma_L + \Gamma_S)xv(x) + e^{-2\Gamma x^2}I_1(ax) \,. \qquad \text{(C7)}$$

Thus we have transformed the double integral into a set of two coupled ordinary differential equations, Eqs. (C6) and (C7), which can be solved readily by standard numerical techniques.

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