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Stimulated Raman scattering: Unified treatment of spontaneous initiation and spatial propagation

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A theory of stimulated Raman scattering (SRS) is presented which treats in a unified manner the buildup of SRS from spontaneous Raman scattering and the spatial propagation which leads to gain. Maxwell-Bloch equations describe the coupling between the Stokes field operator and the collective atomic operators, which are driven by a classical laser field under low-signal-gain conditions, where the atomic ground states and laser field remain undepleted. An analytic expression is derived for the forward-propagating Stokes field operator in a one-dimensional spatial approximation when the Fresnel number of the excited medium is unity. Steady-state and transient Stokes intensities are evaluated under low-gain (spontaneous) and high-gain (stimulated) conditions. The Stokes intensity is found to be exactly independent of a laser bandwidth arising solely from phase fluctuations. The power spectrum of steady-state SRS is evaluated, giving a gain-narrowed spectral profile when the laser is narrow band, and a spectrum identical to the laser spectrum when the laser is broad band. A strong analogy between SRS and two-level superfluorescence is found; in both cases the macroscopic, collective dipole moment is initiated by quantum fluctuations.

I. INTRODUCTION

The generation of intense, frequency-shifted radiation by the nonlinear process of stimulated Raman scattering (SRS) is a commonly used technique in a number of fields, including tunable laser development[1,2] and high-energy laser pulse compression.[3] In spite of extensive theoretical and experimental studies into many important aspects of SRS, the details of the origin of the process have not yet been fully understood. The nature of spontaneous Stokes Raman scattering, which arises from atoms behaving independently, is well understood, as is the propagation and resulting amplification of an externally applied, classical electromagnetic wave, involving the collective behavior of many atoms. These two problems have been treated separately by suitably postulated semiclassical theories.[4,5] However, no theory to date has given an explicit quantum mechanical treatment of the situation in which spontaneously scattered Stokes photons are amplified via propagation through the Raman medium, thus serving as the source for the initiation of the SRS. Previous methods of treating this situation, which will be referred to here as the Raman "generator," include photon rate equations,[6,7] single-mode models,[8] and semiclassical wave equations with phenomenological source terms,[9] and have yielded reasonable results for relatively simple quantities, such as the steady-state SRS output intensity. These methods are nevertheless inadequate for treating more complicated quantities such as the buildup of transient intensities, the dependence on laser bandwith, the spectrum of the SRS output, and higher order statistical properties, including SRS photon counting distributions.

The purpose of this paper is to present a full discussion of a previously reported[10] quantum-mechanical theory of stimulated Raman scattering which treats both the buildup of SRS from spontaneous Raman scattering and the effects of spatial propagation in a unified way. The problem of SRS buildup is found to be mathematically equivalent to the problem of superfluorescence in an extended medium composed of inverted two-level atoms. In each of these cases the macroscopic collective dipole moment, which is initially zero, grows from quantum fluctuations, and the angular, temporal, and spectral properties of the radiated light depend on the geometry of the excited medium in a complicated, and only partially understood, way. To treat the SRS buildup problem, we make use of techniques which have been recently developed for treating two-level superfluorescence, and which include the effects of all of the frequency modes of the radiation field, and one-dimensional spatial propagation.[11,12]

The one-dimensional behavior of the propagation results from the simplifying assumption that the medium is excited in a pencil-shaped region, with Fresnel number of order unity. Such an excited region will radiate predominantly out of the two end faces of the pencil with diffraction-limited angular patterns. This motivates the averaging of the field in the two directions transverse to the pencil's axis.[11,12] This leaves only the longitudinal spatial coordinate as a propagation variable. In this way operator Maxwell-Bloch equations are obtained which account for the collective behavior of the radiators and the propagation of the radiated field. Homogenous (collisional) broadening of the Raman line is accounted for by the inclusion in the Bloch equation of a damping constant and a fluctuating Langevin operator. Inhomogeneous broadening, atomic and laser saturation, and the effects of group-velocity dispersion are ignored. The operator equations admit exact analytic solutions, from which intensities, spectra, and higher order statistical properties can be calculated.

A nonzero bandwidth of the driving laser may be accounted for in an approximate way by use of the phase-diffusion model,[13] in which the laser's amplitude is taken to be constant while its phase is assumed to fluctuate randomly. We find the result, perhaps surprising, that within this model the buildup of the SRS intensity is *exactly* independent of laser bandwidth. This result has been conjectured previously from qualitative, semiclassical arguments,[5,9,13-15] and has been partially confirmed experimentally[14,15] although other measurements[1] seem to indicate a slight dependence of Raman gain on laser bandwidth. Further, we show that when the laser bandwidth is much larger than the Raman linewidth the SRS is generated having the same spectral width as the laser.

II. EQUATIONS OF MOTION

Here we will briefly discuss the physical model and theoretical methods which lead to the equations of motion for the atomic system and the Stokes field, including the effects of collisional damping and fluctuations. The resulting form of of the equations of motion is known from previous treatments which used either an equivalent two-level representation,[16] or a formal operator averaging technique with a multilevel atom.[17,18]

A. Atomic dynamics

We consider a collection of identical atoms or molecules initially in their ground states $|1\rangle$, contained in a pencil-shaped volume with length L and cross-sectional area A. The atomic positions are random, but fixed, and the average number density (atoms cm^{-3}) is N. A laser with (classical) electric field

$$\mathcal{E}_L(z,t)=E_L(z,t)e^{i(\omega_L t-k_L z)}+E_L^\dagger(z,t)e^{-i(\omega_L t-k_L z)} \quad (1)$$

propagates through the volume in the z direction, which is parallel to the pencil axis. As shown in Fig. 1, an atom may absorb a laser photon at frequency ω_L and scatter a photon at the Stokes frequency $\omega_S=\omega_L-\omega_{31}$, ending up in the final state $|3\rangle$. Two time orderings contribute to the process, as shown. We will treat the laser field mode(s) as a classical electromagnetic wave and assume that it does not undergo depletion or any other back reaction from the medium. On the other hand, the remaining modes of the radiation field will be treated quantum mechanically, to allow for the spontaneous initiation of the Raman scattering.

The behavior of a single atom is described in terms of the atomic operators $\hat{\sigma}_{ij}(t)$, which at the initial time are given by

$$\hat{\sigma}_{ij}(0)=|i\rangle\langle j|, \quad (2)$$

and at later times evolve according to the Heisenberg operator equation of motion (in the dipole approximation)

$$\frac{d}{dt}\hat{\sigma}_{ij}=i\omega_{ij}\hat{\sigma}_{ij}+i\sum_k(d_{jk}\hat{E}\hat{\sigma}_{ik}-d_{ki}\hat{E}\hat{\sigma}_{kj}), \quad (3)$$

where $d_{kl}=\hbar^{-1}\langle k|\hat{d}|l\rangle$ is the atomic dipole matrix element (divided by $\hbar$) and $\hat{E}(\vec{r},t)$ is the total electric field operator

$$\hat{E}(\vec{r},t)=\mathcal{E}_L(z,t)+\hat{\mathcal{E}}_S(\vec{r},t), \quad (4)$$

$\mathcal{E}_L(z,t)$ is the classical laser field given in Eq. (1), and $\hat{\mathcal{E}}_S(\vec{r},t)$ is the operator for the remaining field modes, of special interest being those near the Stokes frequency ω_S.

In the case that the laser frequency is far from any intermediate resonances, the levels $|m\rangle$ may

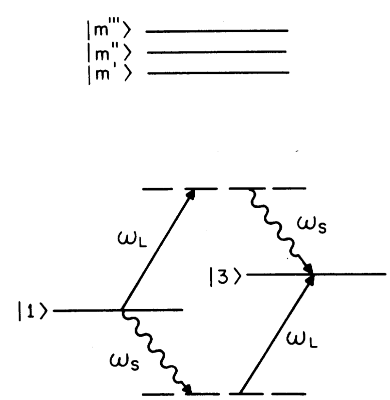

FIG. 1. An atom initially in its ground state $|1\rangle$ is driven by a laser field with frequency ω_L, which is not necessarily in near resonance with any intermediate state $|m\rangle$. Raman scattering at frequency $\omega_S=\omega_L-\omega_{31}$ may occur with either of two time orderings, as shown, leaving the atom in the final state $|3\rangle$.

be adiabatically eliminated from the equation of motion for $\hat{\sigma}_{31}(t)$, which is the atomic operator that couples to the Stokes field. Furthermore, when the fields are weak such that the atom remains essentially in its ground state, the equations may be linearized to yield the usual equation[16,17] for the slowly varying atomic operator $\hat{Q}(t)$ (see Appendix A)

$$\frac{d}{dt}\hat{Q}(t) = -i\kappa_1^* E_L(z,t)\hat{E}_S^{(+)}(\vec{r},t), \tag{5}$$

where $\hat{Q}(t)$ is defined by

$$\hat{Q}(t) = \hat{\sigma}_{31}(t)e^{-i(\omega_L-\omega_S)t+i(k_L-k_S)z}, \tag{6}$$

and the coupling constant is

$$\kappa_1 = \sum_m d_{3m}d_{m1}\left(\frac{1}{\omega_{m1}-\omega_L}+\frac{1}{\omega_{m1}+\omega_S}\right). \tag{7}$$

$E_S^{(+)}(\vec{r},t)$ is the positive frequency component of the field, defined by

$$\hat{\mathcal{E}}_S(\vec{r},t) = \hat{E}_S^{(-)}(\vec{r},t)e^{i(\omega_S t-k_S z)} + \hat{E}_S^{(+)}(\vec{r},t)e^{-i(\omega_S t-k_S z)}, \tag{8}$$

where $k_S = \omega_S/c$.

Homogeneous broadening of the Raman transition, which must be accounted for in a steady-state theory, will be described by adding a term in Eq. (5) which leads to damping of $\hat{Q}(t)$ at a (collisional dephasing) rate Γ, and also a term which leads to random fluctuations[19,20]

$$\frac{d}{dt}\hat{Q}(t) = -\Gamma\hat{Q}(t) - i\kappa_1^* E_L(z,t)\hat{E}_S^{(+)}(\vec{r},t) + \hat{F}(t). \tag{9}$$

$\hat{F}(t)$ is the quantum statistical Langevin operator describing the collision-induced fluctuations. The Langevin force is taken to be delta correlated, and obeys

$$\langle \hat{F}^\dagger(t')\hat{F}(t'')\rangle = 2\Gamma\delta(t'-t''), \tag{10a}$$

$$\langle \hat{F}(t')\hat{F}^\dagger(t'')\rangle = 0, \tag{10b}$$

which guarantees consistency of the operator properties of $\hat{Q}(t)$.[19]

Equation (9) describes the response of a single atom at position $\vec{r}$ to the electric fields and to collisional fluctuations. To treat spatial propagation it is convenient to formulate the atomic response in terms of collective atomic operators, defined as[11,12]

$$\hat{Q}(z,t) = \frac{1}{n}\sum_{\{\alpha\}_z}\hat{Q}^\alpha(t), \tag{11}$$

where $\hat{Q}^\alpha(t)$ is the atomic operator for the atom located at a position $\vec{r}^\alpha$, and obeys an equation of the form of Eq. (9). The sum in Eq. (11) is over all atoms lying within a thin transverse slice of the pencil-shaped medium, with thickness Δz, centered at the longitudinal position z. The average number of atoms in such a slice is $n = NA\Delta z$. The thickness of a slice is assumed to be much smaller than a Stokes wavelength ($\Delta z \ll \lambda_S$), while the volume of a slice is much greater than a cubic wavelength ($A\Delta z \gg \lambda_S^3$). This justifies the neglect of atomic dipole-dipole interactions ($N\lambda_S^3 \ll 1$), while allowing a continuum description of the atomic medium ($NA\Delta z \gg 1$).[11,12] In the continuum limit the collective operator has the property

$$\langle \hat{Q}^\dagger(z,0)\hat{Q}(z',0)\rangle = (1/\rho)\delta(z-z'), \tag{12}$$

where $\rho = NA$ is the linear density of atoms (atoms/cm) along the pencil-shaped region. Similarly the collective Langevin operator $\hat{F}(z,t)$, defined analogously with Eq. (11), obeys

$$\langle \hat{F}^\dagger(z,t)\hat{F}(z',t')\rangle = (2\Gamma/\rho)\delta(z-z')\delta(t-t'). \tag{13}$$

B. Maxwell-Bloch equations

The Stokes field operator $\hat{\mathcal{E}}_S(\vec{r},t)$ obeys the wave equation

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\hat{\mathcal{E}}_S(\vec{r},t) = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\hat{P}_S(\vec{r},t), \tag{14}$$

where $\hat{P}_S(\vec{r},t)$ is the macroscopic polarization operator.

The approximately one-dimensional character of the pencil-shaped medium may now be invoked.[11,12] Assume that $L \gg A^{1/2} \gg \lambda_S$ and that the Fresnel number $\mathcal{F} \equiv A/\lambda_S L$ is of the order of unity ($\mathcal{F} \simeq 1$). This means that the diffraction-limited angular divergence ($\sim\lambda_S/A^{1/2}$) is of the order of the geometrical angle subtended by the pencil ($\sim A^{1/2}/L$), and, therefore, only a single transverse spatial mode contributes strongly to emission along the pencil axis. This motivates the averaging of the wave equation (14) over the transverse slices defined in Eq. (11) for the collective atomic operators. This procedure has been used successfully in treating superflourescence.[11,12] The slice-averaged polarization operator becomes (see Appendix A)

$$\hat{P}_S(z,t) = \frac{1}{\Delta V}\sum_{\{\alpha\}_z}\sum_m \hbar[d_{1m}\hat{\sigma}_{1m}^\alpha(t) + d_{m3}\hat{\sigma}_{m3}^\alpha(t)] + \text{H.a.}$$
$$= N\hbar\kappa_1^* E_L(z,t)\hat{Q}^\dagger(z,t)e^{i(\omega_S t-k_S z)} + \text{H.a.}, \tag{15}$$

where $\Delta V = A\Delta z = n/N$ is the volume of the transverse slice centered at z. The wave equation becomes one-dimensional under this approximation, and, along with the equation for the collective atomic operator $\hat{Q}(z,t)$, can be written in the Maxwell-Bloch form by using the slowly varying-envelope approximation.[21] This gives

$$\left(\pm\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\hat{E}_S^{(-)}(z,t)=-i\kappa_2\hat{Q}^\dagger(z,t)E_L(z,t)\,, \tag{16a}$$

$$\frac{\partial}{\partial t}\hat{Q}^\dagger(z,t)=-\Gamma\hat{Q}^\dagger(z,t)+i\kappa_1E_L^*(z,t)\hat{E}_S^{(-)}(z,t)+\hat{F}^\dagger(z,t)\,, \tag{16b}$$

where $\kappa_2=2\pi N\hbar\,\omega_S\kappa_1^*/c$. The plus and minus signs in Eq. (16a) refer to forward and backward Stokes emission [obtained by replacing $k_S\to -k_S$ in Eq. (8)], respectively. In the perturbative and slowly varying-envelope approximations these oppositely propagating waves are decoupled from one another.[11,12,22]

Equation (16) describes the coupling between atomic medium and fields, and from its solutions the spatial and temporal buildup of the Stokes field will be obtained. Its form is identical to that found in the semiclassical treatment[4,5,13] of Raman amplification, with the exceptions of the operator nature of the Stokes field $\hat{E}_S^{(-)}(z,t)$ and the Raman variable $\hat{Q}(z,t)$, and the presence of the collisional Langevin operator $\hat{F}^\dagger(z,t)$.[23]

C. Solution of Maxwell-Bloch equations

The Maxwell-Bloch equations (16) can be solved exactly in analytic form for arbitrary time dependence of the laser field $E_L(z,t)$ in the case of forward Stokes emission in a dispersionless medium. The solution, familiar from semiclassical theory[4,5] is applicable here in the quantum case because the Maxwell-Bloch equations (16) are linear with respect to the operators.

The initial conditions for the solution are obtained by noting that in a dispersionless medium the laser field $E_L(z,t)$ depends only on the local time variable $\tau=t-z/c$, and thus a laser pulse whose leading edge is at $t=z/c$ leaves the atomic operator $\hat{Q}(z,\tau)$ unperturbed for $\tau<0$. Thus Eq. (12) should be extended to read

$$\langle\hat{Q}^\dagger(z,\tau=0)\hat{Q}(z',\tau=0)\rangle=(1/\rho)\delta(z-z')\,. \tag{17}$$

Also, stationarity of the Langevin force implies, from Eq. (13), that

$$\langle\hat{F}^\dagger(z,\tau)\hat{F}(z',\tau')\rangle=(2\Gamma/\rho)\delta(z-z')\delta(\tau-\tau')\,. \tag{18}$$

The initial value for the Stokes field $\hat{E}_S^{(-)}(0,\tau)$ is given at the input face of the medium, $z=0$, for all time t. This means that backward Stokes emission is explicitly ignored. Depending on the initial state of the radiation field, this condition describes either an externally incident Stokes wave which can experience Raman amplification, or the vacuum field from which Stokes emission can build up.

With these initial conditions the Maxwell-Bloch equations are solved, by a method discussed in Appendix B, to give for the forward-propagating Stokes field operator

$$\begin{aligned}\hat{E}_S^{(-)}(z,\tau)={}&\hat{E}_S^{(-)}(0,\tau)-i\kappa_2E_L(\tau)e^{-\Gamma\tau}\int_0^z dz'\hat{Q}^\dagger(z',0)I_0([4\kappa_1\kappa_2(z-z')p(\tau)]^{1/2})\\&+(\kappa_1\kappa_2 z)^{1/2}E_L(\tau)\int_0^\tau d\tau' e^{-\Gamma(\tau-\tau')}E_L^*(\tau')\hat{E}_S^{(-)}(0,\tau')\frac{I_1(\{4\kappa_1\kappa_2 z[p(\tau)-p(\tau')]\}^{1/2})}{[p(\tau)-p(\tau')]^{1/2}}\\&-i\kappa_2E_L(\tau)\int_0^\tau d\tau'\int_0^z dz' e^{-\Gamma(\tau-\tau')}\hat{F}^\dagger(z',\tau')I_0(\{4\kappa_1\kappa_2(z-z')[p(\tau)-p(\tau')]\}^{1/2})\,,\end{aligned} \tag{19}$$

where the $I_n(x)$ are modified Bessel functions[24] and

$$p(\tau)=\int_0^\tau |E_L(\tau'')|^2 d\tau'' \tag{20}$$

is the power of the laser field integrated up to time τ.

It is interesting to note that if an expectation value is taken of the solution in Eq. (19), then the usual semiclassical result[4,5] is obtained for the average Stokes field $\langle\hat{E}_S^{(-)}(z,\tau)\rangle$ by noting that $\langle\hat{F}^\dagger(z',\tau')\rangle=0$, $\langle\hat{Q}^\dagger(z,0)\rangle=0$, and $\langle\hat{E}_S^{(-)}(0,\tau')\rangle$ is the classical field amplitude of an input Stokes wave.

III. INTENSITY OF RAMAN SCATTERING

The buildup of the intensity of forward stimulated Raman scattering from spontaneous Raman scattering may be obtained from Eq. (19) by calculating the normally ordered expectation value

$$I_S(z,\tau)=\frac{Ac}{2\pi\hbar\,\omega_S}\langle\hat{E}_S^{(-)}(z,\tau)\hat{E}_S^{(+)}(z,\tau)\rangle\,\cdot \tag{21}$$

$I_S(z,\tau)$ is the number of Stokes photons emitted per second through the end face of the pencil-shaped excited region, into the solid angle A/L^2 defined by the geometry of the region. We will consider only the case that no Stokes wave is externally incident on the medium, and so we have for the initial field

$$\langle \hat{E}_S^{(-)}(0,\tau')\hat{E}_S^{(+)}(0,\tau'')\rangle = 0\,, \qquad (22)$$

that is, vacuum fluctuations are not detected with a photodetector. Using Eq. (22), and the fact that $\hat{E}_S^{(-)}(0,\tau)$, $\hat{Q}^\dagger(z,0)$, and $\hat{F}^\dagger(z,\tau)$ are statistically independent and obey Eqs. (17) and (18), we find

$$I_S(z,\tau) = (Ac/2\pi\hbar\,\omega_S\rho)\,|\kappa_2 E_L(\tau)|^2\Big(e^{-2\Gamma\tau}\int_0^z dz'\,I_0^2([4\kappa_1\kappa_2(z-z')p(\tau)]^{1/2}) + 2\Gamma\int_0^\tau d\tau'\int_0^z dz'\,e^{-2\Gamma(\tau-\tau')}I_0^2(\{4\kappa_1\kappa_2[z-z'][p(\tau)-p(\tau')]\}^{1/2})\Big), \qquad (23)$$

with the first and second terms here arising, respectively, from the second and fourth terms in Eq. (19). This result can be simplified further by performing the z' integrations,[25] yielding

$$I_S(z,\tau) = (Ac/2\pi\hbar\,\omega_S\rho)\,|\kappa_2 E_L(\tau)|^2 z\Big(e^{-2\Gamma\tau}\{I_0^2([4\kappa_1\kappa_2 z p(\tau)]^{1/2}) - I_1^2([4\kappa_1\kappa_2 z p(\tau)]^{1/2})\} + 2\Gamma\int_0^\tau d\tau'\,e^{-2\Gamma(\tau-\tau')}[I_0^2(\{4\kappa_1\kappa_2 z[p(\tau)-p(\tau')]\}^{1/2}) - I_1^2(\{4\kappa_1\kappa_2 z[p(\tau)-p(\tau')]\}^{1/2})]\Big). \qquad (24)$$

Equation (24) along with Eq. (20), gives a very general expression for the Stokes intensity for arbitrary time, Raman gain, and laser pulse shape $E_L(\tau)$. Its predictions will be studied in some detail below under various limiting conditions.

A. Spontaneous Raman scattering

An important and illustrative limit of Eq. (24) occurs when the laser intensity is sufficiently low that Raman gain is negligible, and the atoms scatter light independently and spontaneously. In this limit, which occurs if

$$4\kappa_1\kappa_2 z p(\tau) \ll \Gamma\tau, \qquad (25)$$

for all τ, the factor $\exp[-2\Gamma(\tau-\tau')]$ in the integrand in Eq. (24) is sharply peaked compared to the Bessel functions, allowing the latter to be pulled out of the integral at $\tau'=\tau$, leading to

$$I_S(z,\tau) = (Ac/2\pi\hbar\,\omega_S\rho)\,|\kappa_2 E_L(\tau)|^2 z \times [1 + e^{-2\Gamma\tau}\{I_0^2([4\kappa_1\kappa_2 z p(\tau)]^{1/2}) - I_1^2([4\kappa_1\kappa_2 z p(\tau)]^{1/2}) - 1\}], \qquad (26)$$

where $I_0(x)\simeq 1$ and $I_1(x)\simeq x/2$, for $x\ll 1$, have been used. It can further be shown that under the condition (25) the term proportional to $\exp(-2\Gamma\tau)$ is small compared to unity for all times τ, and thus the intensity is

$$I_S(z,\tau) = (Ac/2\pi\hbar\,\omega_S\rho)\,|\kappa_2 E_L(\tau)|^2 z\,. \qquad (27)$$

This result indicates that the intensity of spontaneous Raman scattering follows the laser intensity adiabatically, a result which is known from conventional treatments of Raman scattering from single atoms.[26]

As a check on the reliability on the present method, it is interesting to compare the magnitude of the spontaneous scattering, given in Eq. (27), with that predicted from the spontaneous Raman scattering differential cross section obtained from the conventional Kramers-Heisenberg treatment[7]

$$\frac{d\sigma}{d\Omega} = \frac{\omega_L\omega_S^3\hbar^2}{c^4}\left|\sum_m d_{3m}d_{m1}\left(\frac{1}{\omega_{m1}-\omega_L}+\frac{1}{\omega_{m1}+\omega_S}\right)\right|^2. \qquad (28)$$

Using this formula, and defining the laser intensity $I_L=(c/2\pi\hbar\,\omega_L)|E_L|^2$, in units of photons $\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$, we can rewrite the present result [Eq. (27)] in more familiar form as

$$I_S(z,\tau) = \mathfrak{F}^{-2} I_L\frac{d\sigma}{d\Omega}\,d\Omega\,NV, \qquad (29)$$

where $\mathfrak{F}=A/\lambda_S L$ is the Fresnel number and $d\Omega=A/L^2$ is the geometrical solid angle of the excited region with volume $V=Az$. Thus when $\mathfrak{F}=1$, as we have assumed from the start, the present treatment Eq. (29) exactly reproduces the known result for spontaneous scattering. While this exact agreement is encouraging, it should be considered at least partly fortuitous, since the transverse aspects of the spatial propagation have been treated fairly crudely.

B. Effect of laser bandwidth

A very important general consequence of the full solution given in Eqs. (24) and (20) is the fact that the Stokes intensity is found to be *exactly* independent of the phase of the laser field, as it depends only on the square modulus $|E_L(\tau)|^2$ of the field. A similar result is found in the semiclassical theory of the Raman amplifier, but *only* in the case that the laser bandwidth (arising from phase fluctuations) is much smaller than the Raman linewidth Γ, or in the limit of very high Raman gain.[27] In the present treatment of the Raman generator the independence from laser phase fluctuations is upheld for *arbitrary* laser bandwidths and Raman gains. This result has been conjectured previously on phenomenological grounds,[5,13,14] and will be discussed further in Sec. V.

Let us therefore make a simplifying assumption: In the remainder of the paper the laser intensity $|E_L(\tau)|^2$ will be assumed to be constant, after being switched on at $\tau=0$, while the phase of the field $E_L(\tau)$ will be taken to be a fluctuating random variable, with classical Gaussian statistics, driven by delta-correlated fluctuations. This is known as the phase-diffusion model for laser bandwidth,[28] and leads to the following field autocorrelation function:

$$\langle\langle E_L(\tau)E_L^*(\tau')\rangle\rangle = |E_L|^2 e^{-\Gamma_L|\tau-\tau'|}, \tag{30}$$

where Γ_L is the bandwidth (half-width at half maximum HWHM) of the laser, and the double brackets $\langle\langle\ \rangle\rangle$ indicate a classical average over the ensemble of phases. Using this model we find from Eq. (20) that $p(\tau)=|E_L|^2\tau$, allowing Eq. (24) to be written more simply as

$$I_S(z,\tau)=\tfrac{1}{2}\Gamma gz\Big(e^{-2\Gamma\tau}\,[I_0^2((2gz\Gamma\tau)^{1/2})-I_1^2((2gz\Gamma\tau)^{1/2})] +2\Gamma\int_0^\tau d\tau' e^{-2\Gamma\tau'}[I_0^2((2gz\Gamma\tau')^{1/2}) - I_1^2((2gz\Gamma\tau')^{1/2})]\Big), \tag{31}$$

where g is defined as

$$g=2\kappa_1\kappa_2|E_L|^2/\Gamma, \tag{32}$$

and will be seen below to be the steady-state Raman gain coefficient which is familiar from semiclassical theory.[4,6,13]

In terms of this gain coefficient, the spontaneous scattering intensity Eq. (27) may be written more compactly as

$$I_S(z,\tau)=\tfrac{1}{2}\Gamma gz. \tag{33}$$

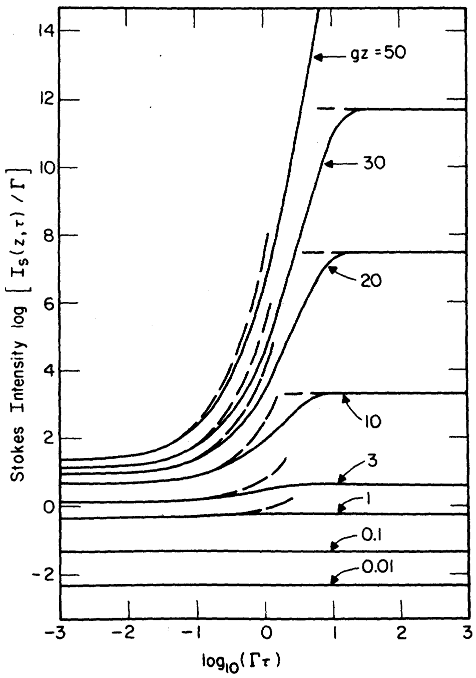

FIG. 2. Instantaneous Stokes intensity as a function of time, for a number of different values of gz (number of steady-state gain lengths). Solid curves are obtained by numerical evaluation of Eq. (31). Broken curves are analytic approximations given by Eq. (35) for small times and Eq. (40) for long times. For very small gains ($gz \le 0.1$) and/or very small times [$\log_{10}(\Gamma\tau) < -2$] the Stokes scattering is spontaneous and agrees with Eq. (33). For larger gains ($gz \ge 1$) the scattering is seen to increase rapidly at a time $\tau \simeq (1/\Gamma gz)$ and then reach a steady-state stimulated intensity at a time $\tau \simeq (gz/\Gamma)$.

C. Buildup of stimulated scattering

Here the instantaneous Stokes intensity $I_S(z,\tau)$ will be evaluated analytically and numerically, and plotted as a function of τ in order to study the transition from spontaneous Raman scattering at very small times to transient SRS at moderately small times, and finally to steady-state SRS.

In Fig. 2 we have plotted $I_S(z,\tau)$, evaluated by numerical integration of Eq. (31), for a number of different values of gz. It is seen that for very small gain ($gz=0.1, 0.01$) the Stokes intensity is a constant, given by the spontaneous scattering result Eq. (29) or (33). For larger values of gz a rapid growth of the intensity is seen at times of the order of Γ^{-1}, the molecular relaxation time. At longer times ($\Gamma\tau \gtrsim gz$) a steady-state value is eventually attained.

Another important quantity in experiments on

SRS is the Stokes pulse energy, or the total number of Stokes photons emitted per pulse into the solid angle A/L^2. Assuming a (square) laser pulse of duration τ_L, this total photon number is given by the integral of the instantaneous Stokes intensity

$$N_S(z,\tau_L)=\int_0^{\tau_L} I_S(z,\tau)d\tau, \quad (34)$$

where $I_S(z,\tau)$ is obtained from Eq. (31). This quantity is plotted in Fig. 3 as a function of laser pulse length τ_L for a number of different values of gz. As expected from Fig. 2, $N_S(z,\tau_L)$ grows linearly in τ_L for small gz, corresponding to pure spontaneous scattering, while for large gz a rapid growth is seen, leading eventually to another region of linear growth, with a slope given by the steady-state Stokes intensity shown in Fig. 2.

Also plotted in Figs. 2 and 3, as broken curves, are analytic approximations of the exact results Eqs. (31) and (34) for $I_S(z,\tau)$ and $N_S(z,\tau_L)$, obtained as follows in the transient and steady-state limits.

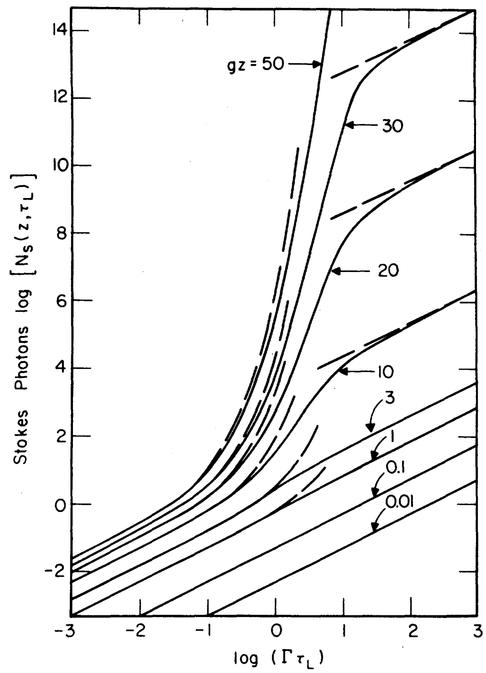

FIG. 3. Stokes photons emitted per laser pulse, of duration τ_L, for a number of different values of gz. Solid curves are obtained by numerical evaluation of Eq. (34). Broken curves are analytic approximations given by Eq. (38) for small times and Eq. (42) for long times. For high enough gains ($gz \geq 1$) the spontaneous scattering seen at very small times grows rapidly to steady-state stimulated scattering, where the total number of Stokes photons emitted is proportional to the pulse duration τ_L.

1. *Transient Raman scattering*

For times short compared to the molecular relaxation time ($\Gamma\tau\rightarrow 0$) only the first term in Eq. (31) contributes, giving for the transient Stokes scattering at arbitrary laser intensity

$$I_S(z,\tau)=\tfrac{1}{2}\Gamma gz[I_0^2((2gz\Gamma\tau)^{1/2})-I_1^2((2gz\Gamma\tau)^{1/2})]. \quad (35)$$

This result can be approximated in the high-gain limit ($gz\Gamma\tau\gg 1$) using the asymptotic form of the modified Bessel function[29]

$$I_n(x)=\frac{e^x}{(2\pi x)^{1/2}}\left(1-\frac{4n^2-1}{8x}+\cdots\right), \quad x\gg 1 \quad (36)$$

which leads to

$$I_S(z,\tau)\simeq\frac{e^{2(2gz\Gamma\tau)^{1/2}}}{8\pi\tau}. \quad (37)$$

The dependence on the factor $\exp[2(2gz\Gamma\tau)^{1/2}]$ is reminiscent of the semiclassical result for the transient Raman amplifier.[4,13] Note that the results (35) and (37) do not depend on the molecular relaxation rate Γ, since Γ appears only in the product $g\Gamma$, and g is inversely proportional to Γ.

The total number of Stokes photons per pulse $N_S(z,\tau_L)$ can be evaluated[25] from Eqs. (34) and (35) to be, for arbitrary laser intensity and $\Gamma\tau\ll 1$,

$$N_S(z,\tau_L)=gz\Gamma\tau_L[I_0^2((2gz\Gamma\tau_L)^{1/2})-I_1^2((2gz\Gamma\tau_L)^{1/2}) -(2gz\Gamma\tau_L)^{-1/2}I_0((2gz\Gamma\tau_L)^{1/2})I_1((2gz\Gamma\tau_L)^{1/2}) \quad (38)$$

The analytic expressions Eqs. (35) and (38) are plotted in Figs. 2 and 3 and are seen to be good approximations for small times $[\Gamma\tau\lesssim(gz)^{-1}]$.

2. *Steady-state Raman scattering*

For times long compared to the molecular relaxation time ($\Gamma\tau\rightarrow\infty$) only the second term in Eq. (31) contributes, with the upper integration limit taken to infinity

$$I_S(z,\infty)=\Gamma^2 gz\int_0^\infty d\tau'\; e^{-2\Gamma\tau'}[I_0^2((2gz\Gamma\tau')^{1/2}) -I_1^2((2gz\Gamma\tau')^{1/2})], \quad (39)$$

which may be integrated[30] to give[10]

$$I_S(z,\infty)=\tfrac{1}{2}\Gamma gz[I_0(gz/2)-I_1(gz/2)]e^{gz/2}. \qquad (40)$$

In the low-gain limit ($gz \ll 1$) this result reduces to the spontaneous scattering intensity in Eq. (33), while in the high-gain limit ($gz \gg 1$) it becomes [using Eq. (36)]

$$I_S(z,\infty)\simeq\frac{\Gamma}{2(\pi gz)^{1/2}}e^{gz}. \qquad (41)$$

This result verifies that g, given by Eq. (32), is properly identified as the steady-state gain coefficient. The dependence on the factor $(\pi gz)^{-1/2}$ is reminiscent of the semiclassical result for the steady-state Raman amplifier, in the case of a broad-band laser.[13]

The total number of Stokes photons per pulse $N_S(z,\tau_L)$ is given in steady-state simply by

$$N_S(z,\tau_L)\simeq\tau_L I_S(z,\infty). \qquad (42)$$

The analytic expressions [Eqs. (40) and (42)] are plotted in Figs. 2 and 3 and are seen to be good approximations for large times ($\Gamma\tau \gtrsim gz$).

In Fig. (4) the steady-state Stokes intensity $I_S(z,\infty)$ given by Eq. (40), is plotted as a function of gz, the number of gain lengths in the medium. The transition from spontaneous (linear) growth to stimulated (expontential-like) growth is clearly demonstrated.

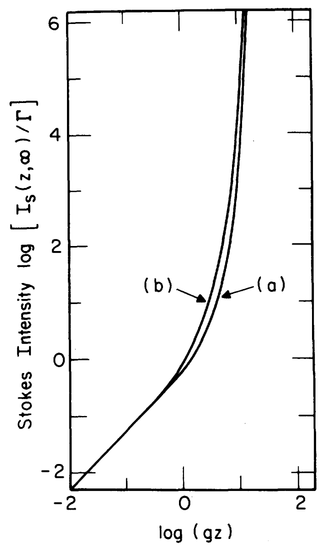

FIG. 4. Steady-state Stokes intensity as a function of gz. Curve (a) is the QED result given by Eq. (40), while curve (b) is the photon rate equation result given by Eq. (46). The curves show the transition from linear, spontaneous growth to exponential (quasiexponential), stimulated growth.

D. Phenomenological photon rate equations

It is instructive to compare the present result Eq. (40) for the steady-state SRS intensity with that resulting from the standard predictions of phenomenological photon rate equations,[6,7] in which Stokes photons produced by spontaneous Raman scattering act as a source for exponential-type stimulated buildup. Consider a pencil-shaped excitation region with area A and length L, with $A \ll L^2$, but with an arbitrary Fresnel number $\mathfrak{F}=A/\lambda_S L$. The intensity $I_S(z)$ of Stokes emission into the small forward solid angle $d\Omega=A/L^2 \ll 1$ obeys approximately the following phenomenological rate equation:

$$\frac{d}{dz}I_S(z)=I_L\frac{d\sigma}{d\Omega}d\Omega\, NA+gI_S, \qquad (43)$$

where the first term describes the spontaneous scattering and the second term describes the stimulated growth, with the steady-state gain coefficient g given by Eq. (32). With the initial condition $I_S(z=0)=0$, corresponding to no Stokes photons at the input of the gain medium, the solution of Eq. (43) is

$$I_S(z)=\left(I_L\frac{d\sigma}{d\Omega}d\Omega\, NA/g\right)(e^{gz}-1). \qquad (44)$$

This solution may be rewritten, using Eqs. (28) and (32) as

$$I_S(z)=(A/\lambda_S L)^2(\Gamma/2)(e^{gz}-1), \qquad (45)$$

showing that the "source term," proportional to $(e^{gz}-1)$, is independent of laser intensity I_L and spontaneous scattering cross section $d\sigma/d\Omega$. In fact, to within a factor of π, the source term just corresponds to the number of field modes of a single polarization contained within the volume AL, solid angle A/L^2, and bandwidth 2Γ, multiplied by $\frac{1}{2}$ photon.[6]

When the Fresnel number is equal to one, Eq. (45) reduces to

$$I_S(z)=\tfrac{1}{2}\Gamma(e^{gz}-1), \qquad (46)$$

which is plotted in Fig. 4, and is seen to be in fairly close agreement with the present result Eq. (40). This qualitative agreement strengthens our confidence in the results of the present theory.

IV. SPECTRUM OF RAMAN SCATTERING IN STEADY STATE

The power spectrum of Raman scattering is an important physical quantity which depends strongly on the quantum-statistical nature of the generated radiation, as well as on the statistical properties of the (classical) incident laser field. The power spectrum $P_S(\omega)$ will be defined as

$$P_S(\omega) = \frac{1}{\pi} \operatorname{Re} \int_{-\infty}^{\infty} e^{-i\omega s}(Ac/2\pi\hbar\,\omega_S) \times \langle \hat{E}_S^{(-)}(z, \tau+s)\hat{E}_S^{(+)}(z, \tau)\rangle ds\,, \quad (47)$$

where $\langle \hat{E}_S^{(-)}(z, \tau+s)\hat{E}_S^{(+)}(z, \tau)\rangle$ is the stationary autocorrelation function of the Stokes field, and ω is the frequency as measured from the frequency ω_S of the carrier wave. The brackets $\langle\,\rangle$ now indicate both a quantum expectation value and a classical average over the ensemble of laser phases. The integral of the power spectrum over all frequencies is normalized to yield the steady-state Stokes intensity

$$\int_{-\infty}^{\infty} P_S(\omega)d\omega = I_S(z, \infty)\,. \quad (48)$$

The power spectrum of the laser, as determined by the phase-diffusion autocorrelation function in Eq. (30), is

$$P_L(\omega) = \frac{Ac|E_L|^2}{2\pi\hbar\,\omega_L}\,\frac{\Gamma_L/\pi}{\omega^2+\Gamma_L^2}\,, \quad (49)$$

where here ω is the frequency as measured from ω_L.

In the steady-state limit ($\Gamma\tau \gg 1$) only the term proportional to $\hat{F}^\dagger(z', \tau')$ in the expression (19) for $\hat{E}_S^{(-)}(z, \tau)$ contributes to the spectrum $P_S(\omega)$, since we are assuming that no Stokes radiation is externally incident on the gain medium at $z=0$. Using Eqs. (19), (18), and (30) the Stokes spectrum is found to be

$$P_S(\omega) = (\alpha\Gamma/2\pi) \operatorname{Re} \int_0^\infty ds\, e^{-(i\omega+\Gamma+\Gamma_L)s} \times \int_0^\tau dt \int_0^z dx\, e^{-2\Gamma t} I_0([\alpha x(t+s)]^{1/2}) I_0((\alpha x t)^{1/2})\,, \quad (50)$$

where $\alpha = 4\kappa_1\kappa_2|E_L|^2 = 2\Gamma g$, and we have used $t=\tau-\tau'$ and $x=z-z'$. The integration in x may be carried out[25] to yield

$$P_S(\omega) = (\Gamma/\pi) \operatorname{Re} \int_0^\infty ds\, e^{-(i\omega+\Gamma+\Gamma_L)s} \times \int_0^\infty dt\, e^{-2\Gamma t} \frac{1}{s} \{[\alpha z(t+s)]^{1/2} I_1([\alpha z(t+s)]^{1/2}) I_0((\alpha z t)^{1/2}) - (\alpha z t)^{1/2} I_1((\alpha z t)^{1/2}) I_0([\alpha z(t+s)]^{1/2})\}\,. \quad (51)$$

This expression will be used to evaluate the Stokes spectrum in several limiting cases.

A. Spectrum of spontaneous Raman scattering

The spontaneous, or low-gain, limit occurs when $gz = \alpha z/2\Gamma \ll 1$. In this case the exponential functions $e^{-2\Gamma t}$ and $e^{-\Gamma s}$ in Eq. (51) are sharply peaked at $t=s=0$, compared to the Bessel functions which behave as

$$I_0(x) \simeq 1\,, \quad I_1(x) \simeq x/2 \quad (52)$$

for $x \ll 1$. Using these approximations in Eq. (51) leads to

$$P_S(\omega) = \tfrac{1}{2}\Gamma g z \frac{(\Gamma+\Gamma_L)/\pi}{\omega^2+(\Gamma+\Gamma_L)^2}\,, \quad (53)$$

which shows the known result[31] that spontaneous Raman scattering has a spectral width given by the sum of the Raman linewidth Γ and the laser bandwidth Γ_L.

B. Spectrum of SRS with a narrow-band laser

When the laser bandwidth Γ_L is much smaller than the Raman linewidth Γ, Eq. (53) indicates that in the low-gain limit the spontaneous scattering has a Lorentzian half width of Γ. We show here that when the gain becomes high, gain narrowing distorts the Lorentzian line shape by amplifying the center part of the Stokes line more strongly than the line wings.

Consider the case that $\Gamma_L \ll \Gamma$ and $gz = \alpha z/2\Gamma \gg 1$. Using the first term in Eq. (36) for the Bessel functions of large argument and the variable change $r=t+s$, Eq. (51) is approximated as

$$P_S(\omega) = \frac{\Gamma}{2\pi^2} \operatorname{Re} \int_0^\infty dr \int_0^r dt\, e^{i\omega(t-r)} \times \frac{e^{(\alpha z r)^{1/2}-\Gamma r} e^{(\alpha z t)^{1/2}-\Gamma t}}{(rt)^{1/2}(r^{1/2}+t^{1/2})}\,, \quad (54)$$

where the integral is meant to be carried out only over the region near the point $r=t=\alpha z/4\Gamma^2$, where the integrand is sharply peaked. Since the factor $[(rt)^{1/2}(r^{1/2}+t^{1/2})]^{-1}$ is slowly varying in this region, it can be pulled out of the integral and replaced by its value at the peak, $2\Gamma^2/\alpha z$. The remaining integral can be performed by noting that the integrand is invariant under interchange of r and t, combined with complex conjugation, allowing $P_S(\omega)$ to be written as

$$P_S(\omega)=\frac{\Gamma^3}{2\pi^2\alpha z}\left|\int_0^\infty dr\, e^{-i\omega r}\, e^{(\alpha z r)^{1/2}-\Gamma r}\right|^2. \quad (55)$$

This integral may be evaluated approximately by extending the lower limit to $-\infty$, to give

$$P_S(\omega)=\frac{\Gamma^3}{2\pi(\omega^2+\Gamma^2)^{3/2}}\, e^{gz}\exp\left(-gz\frac{\omega^2}{\omega^2+\Gamma^2}\right)$$

$$\simeq\frac{1}{2\pi}\, e^{gz}\exp\left(-gz\frac{\omega^2}{\Gamma^2}\right), \quad (56)$$

where the last step follows because the halfwidth $[\Gamma^2/gz]^{1/2}$ of the exponential factor is much less than Γ when gz is large. Using Eq. (41) for the steady-state high-gain Stokes intensity $I_S(z,\infty)$, this result is rewritten as

$$P_S(\omega)=I_S(z,\infty)\left(\frac{gz}{\pi\Gamma^2}\right)\exp\left(-\frac{gz}{\Gamma^2}\omega^2\right), \quad (57)$$

which obeys the normalization condition (48), and which describes a gain-narrowed line profile. This result is similar to that found in the semiclassical treatment of a Raman amplifier with a very broad-band input Stokes wave.[13]

C. Spectrum of SRS with broad-band laser

When the laser bandwidth Γ_L is much larger than the Raman linewidth Γ, Eq. (53) indicates that in the low-gain limit the spontaneous scattering has a width of Γ_L. We show here that this result holds also for high gains.

Consider the case that $\Gamma_L \gg \Gamma$, αz. In this case the integrand in Eq. (51) is significantly different from zero only along the line $s=0$, where the Bessel functions are slowly varying in s compared to $\exp[-(i\omega+\Gamma_L)s]$, allowing them to be pulled out of the s integral at $s=0$. This leads to

$$P_S(\omega)=(\Gamma\alpha z/2\pi)\,\mathrm{Re}\int_0^\infty ds\, e^{-(i\omega+\Gamma_L)s}\int_0^\infty dt\, e^{-2\Gamma t}[I_0^2((\alpha z t)^{1/2})-I_1^2((\alpha z t)^{1/2})], \quad (58)$$

which is simple to evaluate by recognizing the t integral as exactly the steady-state Stokes intensity as given in Eq. (39). Hence,

$$P_S(\omega)=I_S(z,\infty)\frac{\Gamma_L/\pi}{\omega^2+\Gamma_L^2}. \quad (59)$$

This result shows that SRS has the same spectrum as the laser [see Eq. (49)] when the spectrum of the laser is broader than that of the molecular transition.

V. DISCUSSION

In summary, we have presented a quantum-mechanical theory of stimulated Raman scattering which unifies the treatment of the spontaneous initiation and the spatial propagation of SRS. Using a collective atomic operator formalism and a one-dimensional propagation approximation, we have derived an expression, Eq. (19), for the Stokes field operator $\hat{E}_S^{(-)}(z,\tau)$, and have obtained from it Stokes intensities and power spectra for various circumstances.

It is interesting to compare the present results for Raman *generation* with earlier semiclassical results for Raman *amplification*. The familiar semiclassical theory[4,5] gives in steady state for the output Stokes intensity

$$I_S(z)=I_S(0)e^{gz}, \quad (60)$$

where $I_S(0)$ is the intensity at the input of the amplifying medium. The standard connection between single-atom spontaneous scattering and Raman amplification is made by comparing Eq. (60) with the result (45) obtained from photon rate equations. When the Fresnel number $\mathcal{F}=A/\lambda_S L$ is equal is unity, Eq. (46) indicates that one may identify

$$I_S(0)\to\tfrac{1}{2}\Gamma \quad (61)$$

to be an effective source term for the initiation of Raman scattering at high gains in the absence of an input Stokes field. This source term should be compared with the present result found from Eq. (41):

$$I_S(0)\to\tfrac{1}{2}\Gamma(\pi gz)^{-1/2}, \quad (62)$$

which is seen to be a factor $(\pi gz)^{1/2}$ less than the standard result. Although this factor is relatively unimportant when $\exp(gz)$ is very large, its form is interesting since it is very similar to that obtained in a semiclassical treatment of the amplification of a broad-band input Stokes wave (with bandwidth $\Gamma_S \gg \Gamma$) in the presence of a narrow-band pump laser ($\Gamma_L=0$)[32]

$$I_S(z) = I_S(0)(\Gamma/\Gamma_S)(\pi g z)^{-1/2} e^{gz}. \tag{63}$$

One may thus conjecture that the factor $(\pi g z)^{-1/2}$ arises in the present treatment because the spontaneous Stokes scattering, which acts as the source term, has a nonzero bandwidth, equal [as seen in Eq. (53)] to the Raman linewidth Γ. In fact, if $I_S(0)$ is taken to be given by the standard result (61) and Γ_S is taken to be equal to Γ, then the semiclassical amplifier intensity (63) agrees exactly with the generator intensity (41). A similar equivalence holds in the transient case ($\Gamma\tau \ll 1$).[33]

Another point, which has received a lot of attention,[5,9,13-15] is the dependence of the Raman gain on the bandwidth Γ_L of the laser. It was shown in Ref. 13 that at low gains the *amplifier* gain is suppressed by a large phase-diffusion laser bandwidth, while at high gains the effects of the bandwidth are negligible. In contrast, it has been found here (Sec. III B) that the *generator* gain is exactly independent of such a bandwidth for arbitrary gains. One may rationalize this by saying that the spontaneous noise which initiates the process has all possible frequency components, and so that part of the noise which has the proper phase relationship with the broad-band laser is "picked out" and amplified.[5,13,14] The study of the effects of laser bandwidth arising from amplitude fluctuations is made difficult by the complex way in which the amplitude $|E_L(\tau)|$ appears in Eqs. (24) and (20).

Finally, the strong connection between the present work and recent theoretical work[11,12] on two-level superfluorescence will be discussed. The operator Maxwell-Bloch equation (16) for the Raman problem is identical in form to that obtained in the superfluorescence problem,[11] when the two-level atoms are assumed to be predominantly in their excited states, making a linearized theory valid. In contrast to the superfluorescence problem, in which the linearized theory breaks down at long times due to the movement of the atoms to their ground states, the Raman problem has a regime which can be described by the linearized theory for all times and arbitrary gains. This occurs in the low-signal gain limit treated in the present work, where it is assumed that level $|1\rangle$ and the laser remain undepleted. This means that stimulated Raman scattering is properly identified as a particular case of superfluorescence, involving collective atomic behavior initiated by quantum fluctuations. In this case the coupling between the effective inverted medium and the spontaneous radiation field may be thought of as being "turned on" by the presence of the laser field. Superfluorescent effects in Raman scattering have been discussed previously,[8,17,34] but with spatial propagation neglected.

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APPENDIX A: SLOWLY VARYING OPERATOR EQUATIONS

Here we give several details of a derivation which leads to Eqs. (5) and (15). From Eq. (3) we explicitly write the equations

$$\frac{d}{dt}\hat{\sigma}_{31} - i\,\omega_{31}\hat{\sigma}_{31} = i\sum_m d_{1m}\hat{E}\hat{\sigma}_{3m} - i\sum_m d_{m3}\hat{E}\hat{\sigma}_{m1}, \tag{A1}$$

$$\frac{d}{dt}\hat{\sigma}_{m1} - i\omega_{m1}\hat{\sigma}_{m1} = -i\,d_{1m}\hat{E}(\hat{\sigma}_{11} - \hat{\sigma}_{mm}) - i\,d_{3m}\hat{E}\hat{\sigma}_{31}, \tag{A2}$$

$$\frac{d}{dt}\hat{\sigma}_{3m} - i\omega_{3m}\hat{\sigma}_{3m} = -id_{m3}\hat{E}(\hat{\sigma}_{mm} - \hat{\sigma}_{33}) + id_{m1}\hat{E}\hat{\sigma}_{31}. \tag{A3}$$

We have neglected terms which correspond to excitation between the intermediate states $|m\rangle$. For simplicity we assume the atom to be at $z=0$ so that the slowly varying atomic operator $Q(t) = \hat{\sigma}_{31}(t)\exp(-i\,\omega_{31}t)$.

To eliminate $\hat{\sigma}_{m1}$ from Eq. (A1), formally integrate Eq. (A2):

$$\hat{\sigma}_{m1}(t) = \hat{\sigma}_{m1}(0)e^{i\omega_{m1}t} - i\int_0^t dt' e^{i\omega_{m1}(t-t')} \times \{d_{1m}\hat{E}(t')[\hat{\sigma}_{11}(t') - \hat{\sigma}_{mm}(t')] + d_{3m}\hat{E}(t')\hat{Q}(t')e^{i\omega_{31}t'}\}. \tag{A4}$$

When this expression is substituted into Eq. (A1), only those terms which oscillate near the frequency ω_{31}

need to be retained. For example, the $\hat{\sigma}_{m1}(0)$ term does not contribute, while the following term has a part which does:

$$-i\sum_m d_{m3}\int_0^t dt' e^{i\omega_{m1}(t-t')}\, d_{1m}\hat{E}(t)\hat{E}(t')[\hat{\sigma}_{11}(t')-\hat{\sigma}_{mm}(t')] \simeq -i\kappa_1^* E_L(t)\hat{E}_S^{(+)}(t)[\hat{\sigma}_{11}(t)-\hat{\sigma}_{mm}(t)]e^{i\omega_{31}t}, \qquad \text{(A5)}$$

where κ_1 is given in Eq. (7). This result is found by noting that of the sixteen terms arising from $\hat{E}(t)\hat{E}(t')$, only two contribute,

$$E_L(t)e^{i\omega_L t}\hat{E}_S^{(+)}(t')e^{-i\omega_S t'} + E_L(t')e^{i\omega_L t}\hat{E}_S^{(+)}(t)e^{i\omega_S t}, \qquad \text{(A6)}$$

corresponding to the two time orderings shown in Fig. 1. The integral in Eq. (A5) is carried out adiabatically, since E_L, $\hat{E}_S^{(+)}$, $\hat{\sigma}_{11}$, and $\hat{\sigma}_{mm}$ are slowly varying compared to $\exp(i\omega_{m1}t)$. The term in Eq. (A4) containing $\hat{Q}$ contributes only a Stark shift term which we neglect. Carrying out a similar procedure for $\hat{\sigma}_{3m}$ leads to the adiabatic equation for $\hat{\sigma}_{31}$,

$$\frac{d}{dt}\hat{\sigma}_{31} = i\omega_{31}\hat{\sigma}_{31} - i\kappa_1^* E_L\hat{E}_S^{(+)}e^{i\omega_{31}t}(\hat{\sigma}_{11}-\hat{\sigma}_{33}). \qquad \text{(A7)}$$

The final simplification comes by making a linearization assumption. If the atom never departs significantly from its initial state $|1\rangle$, then $\hat{\sigma}_{11}-\hat{\sigma}_{33}$ may be replaced by the unit operator.[11] Equation (A7) [with the $\exp(-ikz)$ factors reincluded] then leads directly to Eq. (5).

The form of the macroscopic polarization operator given in Eq. (15) follows from a similar adiabatic treatment. The polarization is defined as

$$\hat{P}_S(\vec{r},t) = \sum_\alpha \hat{P}_S^\alpha(t)\delta^3(\vec{r}-\vec{r}^{\,\alpha}), \qquad \text{(A8)}$$

where $\hat{P}_S^\alpha(t)$ is the polarization resulting from an atom located at $\vec{r}^{\,\alpha}$:

$$\hat{P}_S^\alpha = \hbar\sum_m (d_{m1}\hat{\sigma}_{m1}^\alpha + d_{3m}\hat{\sigma}_{3m}^\alpha) + \text{H.a.} \qquad \text{(A9)}$$

Substituting Eq. (A4) for $\hat{\sigma}_{m1}^\alpha$, and a similar result for $\hat{\sigma}_{3m}^\alpha$ into this expression, and retaining only those terms which oscillate at ω_S, leads to

$$\hat{P}_S^\alpha = \hbar\kappa_1 E_L^*\hat{Q}^\alpha e^{i(\omega_S t - k_S z)} + \text{H.a.} \qquad \text{(A10)}$$

A term corresponding to a background refractive index for the Stokes wave has been neglected. The macroscopic polarization, averaged over the volume ΔV of a transverse slice at z may then be found:

$$\begin{aligned}\hat{P}_S(z,t) &= \frac{1}{\Delta V}\int_{\Delta V_z}\hat{P}_S(\vec{r}\,',t)d^3r' \\ &= \frac{1}{\Delta V}\sum_{\{\alpha\}_z}\hat{P}_S^\alpha(t) \\ &= N\hbar\kappa_1^* E_L(z,t)\hat{Q}^\dagger(z,t)e^{i(\omega_S t - k_S z)} \\ &\quad + \text{H.a.},\end{aligned} \qquad \text{(A11)}$$

in agreement with Eq. (15).

APPENDIX B: SOLUTION OF MAXWELL-BLOCH EQUATIONS

Here a method will be presented for solving equations of the form of Eq. (16), i.e.,

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)E(z,t) = -i\kappa_2 Q(z,t)A(z,t), \qquad \text{(B1)}$$

$$\frac{\partial}{\partial t}Q(z,t) = -\Gamma Q(z,t) + i\kappa_1 A^*(z,t)E(z,t) + F(z,t). \qquad \text{(B2)}$$

Changing variables to $\tau = t - z/c$, and assuming that $A(z,t)$ depends only on τ, gives

$$\frac{\partial}{\partial z}E(z,\tau) = -i\kappa_2 Q(z,\tau)A(\tau), \qquad \text{(B3)}$$

$$\frac{\partial}{\partial \tau}Q(z,\tau) = -\Gamma Q(z,\tau) + i\kappa_1 A^*(\tau)E(z,\tau) + F(z,\tau). \qquad \text{(B4)}$$

The values of the function $E(z=0,\tau)$ and $Q(z,\tau=0)$ are assumed known.

We will use a Laplace transform technique to solve Eqs. (B3) and (B4). Reimann's method has been employed[4,5] for equations of nearly identical form [without the $F(z,\tau)$ term]. Define $e(x,\tau)$ to be the Laplace transform of $E(z,\tau)$

$$e(s,\tau) = \mathcal{L}\{E(z,\tau)\} = \int_0^\infty e^{-sz}E(z,\tau)\,dz, \qquad \text{(B5)}$$

and similarly

$$q(s,\tau) = \mathcal{L}\{Q(z,\tau)\}, \qquad \text{(B6)}$$

$$f(s,\tau)=\mathcal{L}\{F(z,\tau)\}. \tag{B7}$$

Then Eq. (B3) leads to

$$e(s,\tau)=s^{-1}[E(0,\tau)-i\kappa_2 q(s,\tau)A(\tau)], \tag{B8}$$

and Eq. (B4), along with the above result (B8), leads to

$$\frac{\partial}{\partial\tau}q(s,\tau)=[-\Gamma+s^{-1}\kappa_1\kappa_2|A(\tau)|^2]q(s,\tau)+is^{-1}\kappa_1 A^*(\tau)E(0,\tau)+f(s,\tau), \tag{B9}$$

which may be solved to give

$$q(s,\tau)=q(s,0)e^{-\Gamma\tau}e^{s^{-1}a(\tau)}+\int_0^\tau d\tau' e^{-\Gamma(\tau-\tau')}e^{s^{-1}[a(\tau)-a(\tau')]}\times[is^{-1}\kappa_1 A^*(\tau')E(0,\tau')+f(s,\tau')], \tag{B10}$$

where

$$q(s,0)=\mathcal{L}\{Q(z,0)\}, \tag{B11}$$

and

$$a(\tau)=\kappa_1\kappa_2\int_0^\tau |A(\tau'')|^2 d\tau''. \tag{B12}$$

Substituting this solution (B10) into (B8) gives for $e(s,\tau)$

$$e(s,\tau)=s^{-1}E(0,\tau)-i\kappa_1 A(\tau)e^{-\Gamma\tau}q(s,0)s^{-1}e^{s^{-1}a(\tau)}+A(\tau)\int_0^\tau d\tau' e^{-\Gamma(\tau-\tau')}e^{s^{-1}[a(\tau)-a(\tau')]}\times[s^{-2}\kappa_1\kappa_2 A^*(\tau')E(0,\tau')-is^{-1}\kappa_2 f(s,\tau')]. \tag{B13}$$

This result for $e(s,\tau)$ may be inverse-transformed, with the aid of the Laplace convolution theorem and the known transform of the modified Bessel function[35]

$$\mathcal{L}\{(z/a)^{n/2}I_n((4az)^{1/2})\}=s^{-(n+1)}e^{s^{-1}a} \tag{B14}$$

to yield

$$E(z,\tau)=E(0,\tau)-i\kappa_2 A(\tau)e^{-\Gamma\tau}\int_0^z dz' Q(z'0)I_0([4(z-z')a(\tau)]^{1/2})+(\kappa_1\kappa_2 z)^{1/2}A(\tau)\int_0^\tau d\tau' e^{-\Gamma(\tau-\tau')}A^*(\tau')E(0,\tau')\frac{I_1(\{4z[a(\tau)-a(\tau')]\}^{1/2})}{[a(\tau)-a(\tau')]^{1/2}}-i\kappa_2 A(\tau)\int_0^\tau d\tau'\int_0^z dz' e^{-\Gamma(\tau-\tau')}F(z',\tau')I_0(\{4(z-z')[a(\tau)-a(\tau')]\}^{1/2}). \tag{B15}$$

This verifies the solution given in Eq. (19) of the Maxwell-Bloch equations Eq. (16).

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[33] Equation (37) should be compared to Eq. (22) of Ref. 13, which should be corrected to read

$$\langle\langle |E_S(z,\tau)|^2\rangle\rangle = (\mathcal{E}_{S0}^2/2\pi)[2(\Gamma_L+\Gamma_S)\tau]^{-1} \times \exp[2(\alpha z\tau)^{1/2}] \;.$$

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