Quantum theory of spatial and temporal coherence properties of stimulated Raman scattering

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A quantum theory of stimulated Raman scattering is presented that takes into account three-dimensional propagation and collisional dephasing, allowing the study of the spatial and temporal coherence properties of the generated Stokes light. Maxwell-Bloch equations for the Stokes field operator and the collective atomic operators are solved analytically under low-signal-gain conditions, where the laser field and the atomic ground states remain undepleted. The intensity and the space-time autocorrelation function of the Stokes field are calculated. The Stokes field is expanded into a set of statistically independent "coherence modes," which are determined explicitly for the case of a cylindrically shaped pumped volume. The Stokes pulse energy W is found to fluctuate from pulse to pulse. The probability distribution function for pulse energies $P(W)$ is calculated for a range of Fresnel numbers of the excited volume and collisional dephasing rates. For small values of Fresnel number and dephasing rate, $P(W)$ is a negative exponential distribution. For large values of either, $P(W)$ narrows and approaches a Gaussian-shaped distribution. This occurs because many independent modes contribute to the Stokes emission, making it spatially and/or temporally incoherent.

I. INTRODUCTION

When a pulse of laser light passes through a medium of Raman-active atoms or molecules, intense Stokes-shifted light can be generated via stimulated Raman scattering (SRS). This occurs through the spontaneous scattering of a few photons followed by amplification during their subsequent propagation through the active volume. The effect was first observed in 1962[1] and explained theoretically[2] soon thereafter in terms of a photon rate-equation model.[3] In such a model the stimulated buildup of the photon density is assumed to be much like that in the Einstein A- and B-coefficient model of blackbody radiation; that is, no account is taken of the coherence properties of the radiation. Later, a single-mode quantum model was studied,[4] which has the advantage of correctly describing the buildup of SRS from spontaneous emission, but does not fully describe the situation under consideration since it assumes a light intensity that is uniform along the propagation axis.

An improved quantum theory was introduced[5–7] that includes both spontaneous initiation of the scattering and spatial buildup along the propagation axis, and allows for consideration of the quantum coherence properties of the generated light. The dynamics of the Stokes light buildup was found to pass through a transient regime at short times, where collisional dephasing is unimportant, to a steady-state regime, where collisional dephasing dominates the spectrum and the temporal coherence properties. On the other hand, in this treatment the Stokes light was assumed to be spatially coherent since it was generated in a pencil-shaped volume with a Fresnel number $\mathscr{F}=A/\lambda_S L$ near unity (A and L are the cross-sectional area and length of the pumped volume and λ_S is the Stokes wavelength). This condition implies roughly that diffraction washes out any initial spatial incoherence associated with spontaneous emission. Thus the field was assumed to be in a single spatial mode. Under these assumptions a one-dimensional wave equation is sufficient to describe the spatial propagation along the direction of the intensity buildup.

Using the one-dimensional theory, it was predicted[8] that the energy of generated Stokes pulses fluctuates by large amounts due to the quantum noise intrinsic to the buildup process. This effect, which was subsequently observed,[9–11] occurs even when the driving laser is perfectly stabilized, if the gain remains unsaturated. Typically, a mean number of 10^9 Stokes photons may be produced, with 100% fluctuations around the mean. The Stokes pulse energy, denoted by W, was predicted[8] to be distributed according to a negative exponential probability density function $P(W)=\langle W\rangle^{-1}\exp(-W/\langle W\rangle)$, where $\langle W\rangle$ is the mean energy. This was predicted to hold in the transient regime of SRS, which could be attained by using laser pulses of short duration. However, the most accurate experimental measurements to date[10] showed that in the transient regime the probability distribution function is not a pure exponential, but rather has a maximum occurring at an energy about 10% of the mean energy. This experiment met all of the above criteria for transient, unit-Fresnel number, unsaturated Stokes generation. It was thus concluded that the one-dimensional propagation assumption was inadequate to describe the experiment.

This conclusion was independently supported by the development of a three-dimensional solution[12] of the quantum propagation equations, which includes diffraction and the initial spatial incoherence associated with spontaneous emission. This theory predicted a peak in $P(W)$ at around 10% of the mean energy, in agreement with the measurements. This theory was limited, though, by the assumptions of a square laser temporal shape and the absence of dephasing collisions, which are important when experiments are carried out using longer laser pulses.[9] The effects of collisions on $P(W)$ have been considered in the one-dimensional model, in both steady-state[13] and pulsed[10] situations, but not, so far, in a three-dimensional theory.

The purpose of the present paper is to present a new, more general solution to the problem of Stokes generation that brings all of the above-mentioned phenomena into a single formulation, thus allowing a fuller understanding of the problem as well as quantitative comparison with experiments under a wide range of conditions. The three-dimensional operator equations are solved for a cylindrically shaped pumped region, including the effects of diffraction and spatial incoherence, as well as temporal incoherence caused by dephasing collisions, and arbitrary temporal shape of the laser pulse. The Fresnel number of the pumped region is allowed to vary, but not to become much less than unity, due to the difficulty of taking into account waveguiding effects. The gain is assumed to be unsaturated. The mean Stokes intensity and the Stokes pulse-energy distribution function $P(W)$ are calculated. In doing this the concepts of "spatial coherence modes" and "temporal coherence modes" are utilized. These modes, which are determined explicitly, are statistically independent fields that are added incoherently to produce the total field. The number of significantly excited modes determines the form of $P(W)$. These concepts are known in the literature of classical coherence theory of partially coherent light.[14,15] This paper shows that the same concepts can arise naturally in the quantum treatment of a nonlinear optical process such as SRS.

II. THREE-DIMENSIONAL MAXWELL-BLOCH EQUATIONS

In this section the equations of motion corresponding to the physical model will be formulated and solved. More details of the formulation can be found in Refs. 7 and 12.

A. Formulation of equations

Consider a medium of identical atoms or molecules initially in their ground states $|1\rangle$, with average number density N. A pulse of laser light with electric field $\mathscr{E}_L(\mathbf{r},t)$ travels in the z direction through the medium. It is assumed that the field $\mathscr{E}_L(\mathbf{r},t)$ is uniform over a circular disk of area A, and zero outside the disk, so that a cylindrical volume with cross-sectional area A and length L is pumped uniformly. The laser light, with frequency ω_L, is scattered to produce photons at the Stokes frequency $\omega_S=\omega_L-\omega_{31}$, where $\hbar\omega_{31}$ is the energy of the final state $|3\rangle$ above the ground state $|1\rangle$. We will assume that the laser field is, to a good approximation, unaffected by its interaction with the medium and can be treated classically. On the other hand, the radiation with frequencies near the Stokes frequency will be treated quantum mechanically to allow for spontaneous initiation of the Raman scattering.

It is assumed that the laser frequency is far from any resonances with intermediate states $|m\rangle$ and that the atoms remain predominantly in their ground states $|1\rangle$. In this case the atomic dynamics are described by the evolution of the collective transition operator $\hat{Q}(\mathbf{r},t)$, which acts like a harmonic oscillator raising operator for the atoms at position $\mathbf{r}$. Before the laser pulse arrives, the atomic operators are not correlated for different atoms, i.e.,

$$\langle \hat{Q}^{\dagger}(\mathbf{r},0)\hat{Q}(\mathbf{r}',0)\rangle = N^{-1}\delta^3(\mathbf{r}-\mathbf{r}') , \tag{1}$$

where the expectation value is taken in the initial ground state. The time evolution of $\hat{Q}(\mathbf{r},t)$ is determined by the Heisenberg equation of motion, including collisional damping,[7]

$$\frac{\partial}{\partial t}\hat{Q}(\mathbf{r},t) = -\Gamma\hat{Q}(\mathbf{r},t) - i\kappa_1^* E_L(\mathbf{r},t)\hat{E}_S^{(+)}(\mathbf{r},t) + \hat{F}(\mathbf{r},t) , \tag{2}$$

where $E_L(\mathbf{r},t)$ is the slowly varying laser-field amplitude, and $\hat{E}_S^{(+)}(\mathbf{r},t)$ is the positive frequency part of the slowly varying Stokes field operator, which propagates in the $+z$ direction. It is assumed that there is negligible dispersion, that is, $k_L=\omega_L/c$ and $k_S=\omega_S/c$. The coupling constant κ_1 is given by

$$\kappa_1 = \hbar^{-2}\sum_m d_{3m}d_{m1}\left[\frac{1}{\omega_{m1}-\omega_L}+\frac{1}{\omega_{m1}+\omega_S}\right] , \tag{3}$$

where $d_{ij}=\langle i|\hat{d}|j\rangle$ is the dipole matrix element. The terms $\Gamma\hat{Q}$ and $\hat{F}$ describe the damping and fluctuations, respectively, induced in the atomic operator $\hat{Q}$ by collisions. $\hat{Q}$ is associated with the coherence between levels $|1\rangle$ and $|3\rangle$. (In the case of a molecular vibrational transition, $\hat{Q}$ is the vibrational coordinate.) $\hat{F}$ is a quantum Langevin operator, whose presence guarantees that the commutation relations for $\hat{Q}(\mathbf{r},t)$ are maintained at all times.[16] Γ is the rate of collisional dephasing, and it is also the half-width at half maximum of the spontaneous Raman transition, ignoring Doppler broadening. $\hat{F}$ and Γ are related by

$$\langle \hat{F}^{\dagger}(\mathbf{r},t)\hat{F}(\mathbf{r}',t')\rangle = 2\Gamma N^{-1}\delta(t-t')\delta^3(\mathbf{r}-\mathbf{r}') . \tag{4}$$

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

$$\left[\nabla^2-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right]\hat{E}_S^{(+)}(\mathbf{r},t)e^{-i(\omega_S t-k_S z)} = \frac{2\kappa_2^*}{c\omega_S}\frac{\partial^2}{\partial t^2}E_L^*(\mathbf{r},t)\hat{Q}(\mathbf{r},t)e^{-i(\omega_S t-k_S z)} , \tag{5}$$

where $\kappa_2=2\pi N\hbar\omega_S\kappa_1^*/c$.

Equations (2) and (5) make up the operator Maxwell-Bloch equations for the SRS problem in three dimensions. An important quantity that enters into the solutions is the Raman gain coefficient

$$g = 2\kappa_1\kappa_2\Gamma^{-1} |E_L(\mathbf{r},t)|^2 . \quad (6)$$

In a simple rate-equation treatment of steady-state SRS with a constant laser intensity, the Stokes intensity grows as $I_{S0}\exp(gz)$, where I_{S0} is the Stokes intensity at $z=0$.

B. Solution of equations

The laser pulse inside the cylinder is a function only of the local time $\tau = t - z/c$, and if its leading edge is defined to be at $t = z/c$, or $\tau = 0$, the atomic operator $\hat{Q}(\mathbf{r},t)$ is left unperturbed for times such that $\tau < 0$. It is thus convenient to change variables from $(\mathbf{r},t)$ to $(\mathbf{r},\tau)$. Then Eqs. (1) and (4) become

$$\langle \hat{Q}^\dagger(\mathbf{r},\tau=0)\hat{Q}(\mathbf{r}',\tau=0)\rangle = N^{-1}\delta^3(\mathbf{r}-\mathbf{r}') , \quad (7a)$$

$$\langle \hat{F}^\dagger(\mathbf{r},\tau)\hat{F}(\mathbf{r}',\tau')\rangle = 2\Gamma N^{-1}\delta(\tau-\tau')\delta^3(\mathbf{r}-\mathbf{r}') . \quad (7b)$$

The boundary condition for the Stokes field is that $\hat{E}_S^{(+)}(\mathbf{r},\tau)$ is equal to the free-field operator at the input face of the medium ($z=0$). We will consider only the case that no Stokes photons are incident on the input face from the outside. This means that all terms in the solution that are proportional to the free field will give zero contribution to all normally ordered expectation values, such as Stokes intensity $\langle \hat{E}_S^{(-)}(\mathbf{r},\tau)\hat{E}_S^{(+)}(\mathbf{r},\tau)\rangle$. The free-field terms will thus be omitted from the solution below.

In order to solve Eqs. (2) and (5) we make the assumption that reflections from the outside surface of the cylinder, due to dispersion near the Raman transition, are negligible. This means we can neglect the field boundary conditions at this surface, as discussed in detail for two-level superfluorescence in Ref. 17. It was shown that in that case this approximation is valid when the Fresnel number $\mathcal{F} = A/\lambda L$ is greater than unity, which guarantees that the typical angles of incidence on the cylinder surface are not in the regime of total internal reflection. The criterion for neglecting reflections in the transient SRS case is the same, i.e., $\mathcal{F} = A/\lambda_S L \gtrsim 1$. In the steady-state case an analogous argument leads, in the high-gain limit, to $\mathcal{F} \geq (gL/4)^{1/2}$. For $gL = 16$, this gives $\mathcal{F} \geq 2$, of the same order as in the transient case.

Under these assumptions the coupled Eqs. (2) and (5) have the following solution, derived in Appendix A,

$$\hat{E}_S^{(+)}(\mathbf{r},\tau) = \int d^3r'\, K(\mathbf{r},\mathbf{r}',\tau)\hat{Q}(\mathbf{r}',0) + \int d^3r' \int_0^\tau d\tau'\, H(\mathbf{r},\mathbf{r}',\tau,\tau')\hat{F}(\mathbf{r}',\tau') , \quad (8a)$$

where the integral kernels are given by

$$K(\mathbf{r},\mathbf{r}',\tau) = \frac{k_S\kappa_2^*}{2\pi} E_L^*(\tau)\frac{e^{-\Gamma\tau}}{z-z'}\exp\left[-i\frac{k_S}{2}\frac{|\boldsymbol{\rho}-\boldsymbol{\rho}'|^2}{(z-z')}\right] \times I_0([4\kappa_1\kappa_2 p(\tau)(z-z')]^{1/2}) , \quad (8b)$$

and

$$H(\mathbf{r},\mathbf{r}',\tau,\tau') = \frac{k_S\kappa_2^*}{2\pi} E_L^*(\tau)\frac{e^{-\Gamma(\tau-\tau')}}{z-z'}\exp\left[-i\frac{k_S}{2}\frac{|\boldsymbol{\rho}-\boldsymbol{\rho}'|^2}{(z-z')}\right] \times I_0(\{4\kappa_1\kappa_2[p(\tau)-p(\tau')](z-z')\}^{1/2}) , \quad (8c)$$

where

$$p(\tau) = \int_0^\tau |E_L(\tau')|^2 d\tau' , \quad (8d)$$

$\boldsymbol{\rho}$ is the radial vector (x,y) and $I_0(x)$ is the modified Bessel function of zero order. The r' integral in Eq. (8a) is over the cylinder volume excited by the laser. This solution is obtained in the paraxial approximation, which is valid for light traveling at small angles to the z axis, and is useful for a cylinder whose length is much longer than its diameter. Because of this approximation the factor $(z-z')^{-1}$ appears in the solution instead of $|\mathbf{r}-\mathbf{r}'|^{-1}$, leading to a divergence near $z'=z$. The divergence is overcome by noting that, in stimulated scattering with high gain, the region near $z'=0$ contributes more strongly than that near $z'=z$. So the integrals in Eq. (8a) need not be extended to the region near $z'=z$. On the other hand, the solution in Eq. (8a) does not properly describe the low-gain scattering (spontaneous emission) regime, because in that regime the region near $z'=z$ must be included, which leads to a divergence.

It should be pointed out that it is possible to solve Eqs. (2) and (5) without the paraxial approximation if the laser pulse is assumed to have a square temporal shape. This avoids the divergence problem at $z'=z$. See Appendix B.

III. INTENSITY OF THREE-DIMENSIONAL STOKES GENERATION

Restricting ourselves to the high-gain regime, we will evaluate the average intensity of the generated Stokes wave from the paraxial solution, Eq. (8). The intensity, in units of photons/cm^2 sec, at the output face $z=L$, is given by

$$I_S(\boldsymbol{\rho},\tau) = \frac{c}{2\pi\hbar\omega_S}\langle \hat{E}_S^{(-)}(\boldsymbol{\rho},L,\tau)\hat{E}_S^{(+)}(\boldsymbol{\rho},L,\tau)\rangle , \quad (9)$$

where the expectation value is taken in the initial state with no Stokes photons and all atoms in their ground states. Using the δ-correlated properties of $\hat{Q}$ and $\hat{F}$ given by Eq. (7), the intensity is seen to be

$$I_S(\boldsymbol{\rho},\tau) = \frac{c}{2\pi\hbar\omega_S}\left[N^{-1}\int d^3r'\, |K(\mathbf{r},\mathbf{r}',\tau)|^2 + 2\Gamma N^{-1}\int d^3r' \int_0^\tau d\tau'\, |H(\mathbf{r},\mathbf{r}',\tau,\tau')|^2\right] . \quad (10)$$

This result will be evaluated in the transient and steady-state limits.

A. Transient Stokes intensity

When the collisional dephasing rate Γ is sufficiently small to have no effect during the Stokes pulse, the scattering is said to be in the transient regime. This is certainly true when the laser pulse duration τ_L satisfies $\Gamma\tau_L \ll 1$. A more accurate criterion is given by $\Gamma\tau_L < gL$, where g is the gain coefficient given in Eq. (6) and L is the medium length. This arises from the requirement that dephasing be negligible for the duration of the Stokes pulse. For a square laser pulse of duration τ_L, the Stokes intensity grows in time as $\exp[(8gL\Gamma\tau_L)^{1/2}]$,[7] leading to a Stokes duration τ_S (full width at half maximum) of approximately $(\tau_L/16\Gamma gL)^{1/2}$. Then, the condition $\Gamma\tau_S \ll 1$ is equivalent to $\Gamma\tau_L \ll 16gL$, which can be stated conservatively as $\Gamma\tau_L < gL$. This is consistent with the fact that the time needed for the Stokes intensity to reach its steady-state value is of order of gL/Γ.[7]

In the transient regime, the first term in Eq. (10) dominates the intensity. Using the asymptotic formula for the Bessel function

$$I_n(x) \sim \frac{\exp(x)}{(2\pi x)^{1/2}}, \quad x \gg 1 \tag{11}$$

the transient intensity at the output face ($z=L$) is found to be, in the high-gain limit,

$$I_{\rm TR}(\rho,\tau) = \frac{\mathcal{F}^2}{8\pi A}\frac{|E_L(\tau)|^2}{p(\tau)}\exp\{[16\kappa_1\kappa_2 L p(\tau)]^{1/2}\} . \tag{12}$$

Note that in this treatment the average Stokes intensity is constant over the output face, with area A. The result Eq. (12) is equivalent to the previous one-dimensional result [Eq. (37), Ref. 7], except for the factor $\mathcal{F}^2$. This is an intuitively expected result since $\mathcal{F}^2$ is roughly the number of spatial modes of the Stokes field that can fit within the cylindrically shaped pumped volume.

B. Steady-state Stokes intensity

When the laser pulse is sufficiently long, the Stokes intensity becomes constant in time. This occurs when $\Gamma\tau_L \gg gL$, that is, when dephasing prevents the further growth of the collective dipole. In this case the second term in Eq. (10) dominates the intensity, which becomes, for $\tau\to\infty$, in the high-gain limit

$$I_{\rm SS} = \frac{\mathcal{F}^2}{A(4\pi gL)^{1/2}}e^{gL} . \tag{13}$$

Again, this is equivalent to the one-dimensional result,[7] except for the factor $\mathcal{F}^2$.

C. General case

In the case of arbitrary values of $\Gamma\tau$, the intensity from Eq. (10) can be shown to be given by

$$I_S(\rho,\tau) \cong \frac{\mathcal{F}^2}{A} I_S^{\rm 1D}(L,\tau) , \tag{14}$$

where $I_S^{\rm 1D}(L,\tau)$ is the intensity in the one-dimensional case ($\mathcal{F}=1$), which is given explicitly by Eq. (21) and Fig. 2 in Ref. 7. Again, this is valid only in the high-gain limit.

IV. STATISTICAL PROPERTIES OF STOKES EMISSION

In this section the spatial and temporal coherence properties of the Stokes output field are examined through the field correlation function. The field is expanded into a set of statistically independent "coherence modes." Using this expansion, the probability distribution $P(W)$ for Stokes pulse energy W is found.

A. Stokes field correlation function

The correlation function of the Stokes field in the plane of the output face ($z=L$) is defined to be

$$G(\rho_1,\tau_1;\rho_2,\tau_2) = \frac{c}{2\pi\hbar\omega_S}\langle \hat{E}_S^{(-)}(\rho_1,L,\tau_1)\hat{E}_S^{(+)}(\rho_2,L,\tau_2)\rangle , \tag{15}$$

where again $\rho=(x,y)$ is the radial vector. Using Eqs. (7) and (8), this can be evaluated explicitly. In general the result is very complex. It can be shown explicitly that in the limit of high gain the correlation function factorizes into the product of a spatial part times a temporal part

$$G(\rho_1,\tau_1;\rho_2,\tau_2) \cong G_\rho(\rho_1,\rho_2)G_\tau(\tau_1,\tau_2) . \tag{16}$$

The spatial part is found to be

$$G_\rho(\rho_1,\rho_2) = A^{-2}\int_A d^2\rho' \times \exp\left[-i\frac{k_S}{2L}(|\rho_1-\rho'|^2-|\rho_2-\rho'|^2)\right] , \tag{17}$$

where the integral is over the output face of area A, and the temporal part is found to be

$$G_\tau(\tau_1,\tau_2) = \frac{\mathcal{F}^2 Ac}{2\pi\hbar\omega_S}\langle \hat{E}_S^{(-)}(L,\tau_1)\hat{E}_S^{(+)}(L,\tau_2)\rangle_{\rm 1D} . \tag{18}$$

Except for the factor $\mathcal{F}^2$ the correlation function in Eq. (18) is exactly the same as that which arises in the one-dimensional theory of SRS,[10] which is approximately valid for Fresnel numbers near unity. The factorization of the correlation function will be seen below to greatly simplify the determination of the statistical properties of the Stokes light.

The spatial part of the correlation function, Eq. (17), can be evaluated to give[18]

$$G_\rho(\rho_1,\rho_2) = \frac{1}{A}\frac{2J_1(2\mathcal{F}|\rho_1-\rho_2|/a)}{2\mathcal{F}|\rho_1-\rho_2|/a} \times \exp\left[-i\frac{\mathcal{F}}{a^2}(|\rho_1|^2-|\rho_2|^2)\right] , \tag{19}$$

where $J_1(x)$ is the Bessel function of order 1 and a is the radius of the cylinder (i.e., $A=\pi a^2$). Equation (19) is the well-known measure of spatial coherence of light emitted incoherently from a disk with radius a in the plane $z=0$ and observed in the plane $z=L$.[18] The coherence area of the light at the $z=L$ plane can be seen from Eq. (19) to be approximately $\pi a^2/\mathscr{F}^2$, or $(\lambda_S L)^2/\pi a^2$, which shows that the farther the light travels from its source at $z=0$, the more spatially coherent it becomes. If $\mathscr{F}\cong 1$, only one coherence mode (spatially coherent field) can pass from the source through the area πa^2 at $z=L$, while if $\mathscr{F}\gg 1$, then roughly $\mathscr{F}^2$ modes can pass through the area. Thus, for small $\mathscr{F}$ the effective aperture of the pumped gain region acts as a spatial filter for the Stokes light. This further justifies the approximate one-dimensional (single-mode) treatment of SRS for the case $\mathscr{F}=1$ in Ref. 7.

The temporal correlation function, Eq. (18), can be evaluated to give

$$G_\tau(\tau_1,\tau_2)=\mathscr{F}^2\frac{2\kappa_1\kappa_2 E_L(\tau_1)E_L^*(\tau_2)}{q(\tau_1,\tau_2)}e^{-\Gamma(\tau_1+\tau_2)}\times\left[f(\tau_1,\tau_2)+2\Gamma\int_0^T e^{2\Gamma\tau'}g(\tau')d\tau'\right], \quad (20a)$$

where

$$q(\tau_a,\tau_b)=4\kappa_1\kappa_2 L\int_{\tau_b}^{\tau_a}|E_L(\tau')|^2 d\tau',$$
$$f(\tau_1,\tau_2)=[q(\tau_1,0)]^{1/2}I_1([q(\tau_1,0)]^{1/2})\times I_0([q(\tau_2,0)]^{1/2})-(1\leftrightarrow 2), \quad (20b)$$
$$g(\tau')=[q(\tau_1,\tau')]^{1/2}I_1([q(\tau_1,\tau')]^{1/2})\times I_0([q(\tau_2,\tau')]^{1/2})-(1\leftrightarrow 2),$$

where $(1\leftrightarrow 2)$ indicates interchange of τ_1 and τ_2, and T is the lesser of τ_1 and τ_2. The coherence time τ_c can be defined by $G_\tau(\tau-\tau_c,\tau)=\frac{1}{2}G_\tau(\tau,\tau)$. In the transient, high-gain limit, where only the term containing $f(\tau_1,\tau_2)$ is significant, it can be shown that $\tau_c\cong(\tau/16\Gamma gL)^{1/2}$. This is essentially equal to the duration of the Stokes pulse, as expected for a transform-limited pulse. In the steady-state, high-gain limit, where only the term containing $g(\tau')$ is significant, it can be shown [indirectly from Eq. (57) of Ref. 7] that $\tau_c\cong(gL)^{1/2}/\Gamma$. The light becomes more temporally coherent as the gain increases due to gain narrowing of the emission line.

B. Expansion of field in coherence modes

Following classical coherence theory we will expand the Stokes field in a set of modes, or fields, each of which is coherent, but uncorrelated with any other mode. Such an expansion is known as a Karhunen-Loeve expansion.[14] It was independently developed via functional integration techniques in Ref. 12. A related expansion was recently developed and applied to study the coherence of laser modes.[15] A different mode expansion for the SRS problem was developed in Ref. 19, where statistical features were not studied.

It will be shown below that the result of the mode expansion is to put the spatial autocorrelation function into the form

$$G_\rho(\rho_1,\rho_2)=\sum_n \beta_n\phi_n(\rho_1)\phi_n^*(\rho_2). \quad (21)$$

Each term in the sum is in the factorized form $\phi_n(\rho_1)\phi_n^*(\rho_2)$, which describes a field that is completely spatially coherent, in that its complex degree of coherence[18] attains its maximum value, unity. The $\phi_n(\rho)$ are the spatial coherence modes.

One of the desirable consequences of the mode expansion leading to Eq. (21) is that the probability distribution $P(W)$ for Stokes pulse energy W can be expressed simply in terms of the β_n, once they are known. This will be seen in Sec. IV C.

1. Spatial coherence modes

Consider expanding the Stokes field operator in the output plane $z=L$ as

$$\hat{E}_S^{(-)}(\rho,L,\tau)=\left[\frac{2\pi\hbar\omega_S}{c}\right]^{1/2}\sum_n \hat{a}_n^\dagger(\tau)\phi_n(\rho), \quad (22a)$$

where $|\rho|$ is in the domain 0 to a, and τ is in the domain $-\infty$ to $+\infty$. The factor in front is for convenience. The spatial coherence modes $\phi_n(\rho)$ are assumed to be orthonormal,

$$\int_A d^2\rho\,\phi_n(\rho)\phi_m^*(\rho)=\delta_{nm}, \quad (22b)$$

and therefore the operator coefficients $\hat{a}_n(\tau)$ are given by

$$\hat{a}_n^\dagger(\tau)=\left[\frac{c}{2\pi\hbar\omega_S}\right]^{1/2}\int_A d^2\rho\,\hat{E}_S^{(-)}(\rho,L,\tau)\phi_n(\rho). \quad (23)$$

The field correlation function, Eq. (15), can be expressed as

$$G(\rho_1,\tau_1;\rho_2,\tau_2)=\sum_{n,m}\langle\hat{a}_n^\dagger(\tau_1)\hat{a}_m(\tau_2)\rangle\phi_n(\rho_1)\phi_m^*(\rho_2). \quad (24)$$

Now a great simplification can be made by using the property, Eq. (16), that the correlation function factorizes into space and time parts. Using this, Eq. (24) can be written

$$G(\rho_1,\tau_1;\rho_2,\tau_2)=G_\tau(\tau_1,\tau_2)\sum_{n,m}\left[\frac{\langle\hat{a}_n^\dagger(\tau_1)\hat{a}_m(\tau_2)\rangle}{G_\tau(\tau_1,\tau_2)}\right]\times\phi_n(\rho_1)\phi_m^*(\rho_2), \quad (25)$$

where the term in large parentheses must be independent of τ_1 and τ_2. We can set it equal to a constant β_{nm} and thus write

$$\langle\hat{a}_n^\dagger(\tau_1)\hat{a}_m(\tau_2)\rangle=\beta_{nm}G_\tau(\tau_1,\tau_2). \quad (26)$$

A further simplification comes about by imposing the condition that the mode amplitudes $\hat{a}_n$ be uncorrelated, i.e., that $\langle\hat{a}_n^\dagger(\tau_1)\hat{a}_m(\tau_2)\rangle$ equals zero if $m\neq n$. This means that Eq. (26) becomes

$$\langle\hat{a}_n^\dagger(\tau_1)\hat{a}_m(\tau_2)\rangle=\beta_n\delta_{nm}G_\tau(\tau_1,\tau_2), \quad (27)$$

where the second subscript on the constants β_{nm} was dropped for simplicity. The values of the constants β_n will be determined below. Note that using Eq. (27) in (24) immediately leads to the desired form of the spatial correlation function given in Eq. (21).

The physical interpretation of the $\hat{a}_n$ and the β_n can be found by noting that each spatial mode $\phi_n(\rho)$ has temporal coherence described by Eq. (27) with $m=n$. Also, at $\tau_2=\tau_1=\tau$ this gives the mean number of photons in the mode, using Eqs. (14) and (18), as

$$\langle \hat{a}_n^\dagger(\tau)\hat{a}_n(\tau)\rangle=\beta_n G_\tau(\tau,\tau)=\beta_n A I_S(\boldsymbol{\rho},\tau) , \quad (28)$$

which is equal to β_n times the total number of photons per second being emitted through the output face at time τ. Thus β_n is seen to be the fraction of photons emitted into spatial mode $\phi_n(\boldsymbol{\rho})$. Evidently, then, it must be true that

$$\sum_n \beta_n=1. \quad (29)$$

This can be proved easily from Eq. (21), from which it follows that

$$\sum_n \beta_n=\int_A d^2\rho\, G_\rho(\boldsymbol{\rho},\boldsymbol{\rho})=1 , \quad (30)$$

the last step following from Eq. (19).

In order to determine the values of the β_n and the explicit forms of the $\phi_n(\boldsymbol{\rho})$, note that, using Eqs. (23), (15), and (16), we can derive from Eq. (26) the result

$$\int_A d^2\rho \int_A d^2\rho'\, G_\rho(\boldsymbol{\rho},\boldsymbol{\rho}')\phi_n^*(\boldsymbol{\rho})\phi_m(\boldsymbol{\rho}')=\beta_n\delta_{nm} . \quad (31)$$

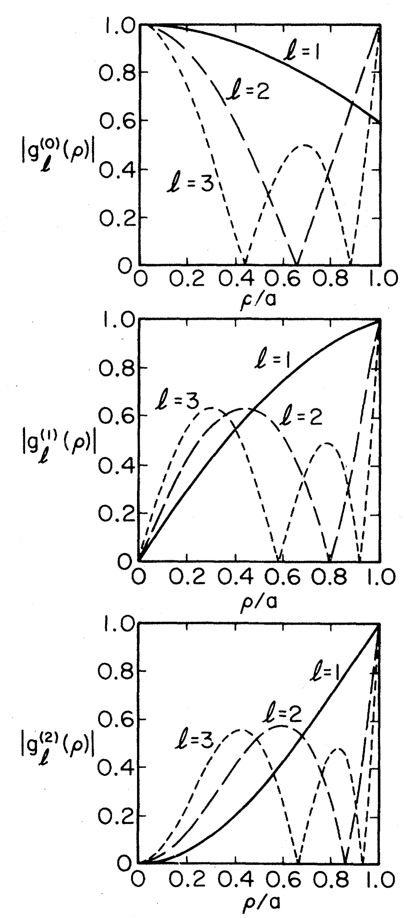

FIG. 1. Absolute value of radial eigenfunctions $g_l^{(k)}(\rho)$ for the case $\mathscr{F}=1$, from Eq. (34). Each eigenfunction is plotted so that its maximum value is unity.

TABLE I. Radial eigenvalues $\beta_l^{(k)}$ corresponding to excitation strengths of spatial coherence modes $g_l^{(k)}(\rho)$, determined from Eq. (34). For Fresnel number $\mathscr{F}$ equal to unity, a small number of spatial modes are significantly excited, while for $\mathscr{F}=3$, a large number are excited.

$\mathscr{F}$	k	l	$\beta_l^{(k)}$
1	0	1	0.630
		2	0.672(−2)[a]
		3	0.192(−5)
		4	0.981(−10)
	1	1	0.161
		2	0.185(−3)
		3	0.190(−7)
	2	1	0.191(−1)
		2	0.405(−5)
		3	0.170(−9)
3	0	1	0.111
		2	0.923(−1)
		3	0.903(−2)
		4	0.486(−4)
	1	1	0.109
		2	0.477(−1)
		3	0.929(−3)
		4	0.223(−5)
	2	1	0.994(−1)
		2	0.129(−1)
		3	0.726(−4)
		4	0.938(−7)
	3	1	0.710(−1)
		2	0.215(−2)
		3	0.484(−5)
	4	1	0.350(−1)
		2	0.257(−3)
		3	0.285(−6)
	5	1	0.117(−1)
		2	0.255(−4)
		3	0.152(−7)
	6	1	0.275(−2)
		2	0.215(−5)
		3	0.123(−8)

[a]The number enclosed in parentheses indicates the exponent of the multiplicative factor of 10.

This equation can be satisfied, noting the orthogonality of the $\phi_n(\boldsymbol{\rho})$, if we require that

$$\int_A d^2\rho'\, G_\rho(\boldsymbol{\rho},\boldsymbol{\rho}')\phi_m(\boldsymbol{\rho}')=\beta_m\phi_m(\boldsymbol{\rho}) . \quad (32)$$

This integral eigenvalue problem with the kernel $G_\rho(\boldsymbol{\rho},\boldsymbol{\rho}')$ given by Eq. (19), has an infinite number of solutions $\phi_m(\boldsymbol{\rho})$, with associated eigenvalues β_m, which can be found by numerical means. If the position $\boldsymbol{\rho}$ is expressed in polar coordinates, $\boldsymbol{\rho}=(\rho\cos\theta,\rho\sin\theta)$, it is shown in Appendix C that the eigenfunctions can be labeled with two integers, k and l, and can be written as

$$\phi_l^{(k)}(\boldsymbol{\rho})=e^{ik\theta}g_l^{(k)}(\rho)\exp\left[-i\frac{\mathscr{F}}{a^2}\rho^2\right], \tag{33}$$

where $k=0,\pm1,\pm2,\ldots$ and $l=1,2,3,\ldots$. The factor $\exp(-i\mathscr{F}\rho^2/a^2)$ describes a paraxial spherical wave originating at $\mathbf{r}=0$. The radial eigenfunctions $g_l^{(k)}(\rho)$ obey the equation

$$\int_0^a d\rho'\, U^{(k)}(\rho',\rho)g_l^{(k)}(\rho')=\beta_l^{(k)}g_l^{(k)}(\rho), \tag{34}$$

where the new kernel $U^{(k)}(\rho',\rho)$ is given in Appendix C. For $k=0$ the eigenvalues are nondegenerate, while for $|k|\geq1$ the eigenvalues are doubly degenerate, corresponding to $k=\pm|k|$. Equation (34) is solved numerically by discretizing the ρ' variable and using a standard matrix diagonalization routine.

Figure 1 and Table I give some of the radial eigenfunctions and eigenvalues, determined numerically for the case $\mathscr{F}=1$. The lowest-order mode $g_1^{(0)}(\rho)$ has no radial or angular nodes. It is approximately uniform across the output face and is the dominant excitation, having $\beta_1^{(0)}=0.63$. Nevertheless, we find that even for $\mathscr{F}=1$, the modes $g_1^{(\pm1)}$ are excited with non-negligible excitation strengths $\beta_1^{(\pm1)}=0.16$. These modes have a zero at $\rho=0$, and suitable linear combinations of them have angular nodes at $\theta=0$ and $\theta=\pi$.

Figure 2 and Table I give some of the eigenfunctions and eigenvalues for the case $\mathscr{F}=3$. The shapes of the eigenfunctions are qualitatively similar to the $\mathscr{F}=1$ case, but the eigenvalues, giving the excitation strengths, show that now many modes ($\sim\mathscr{F}^2$) are excited. This is in agreement with the qualitative discussion following Eq. (19).

2. Temporal coherence modes

A procedure formally analogous to the above development of spatial coherence modes can be used to find the temporal coherence modes.[14] The intuitive meaning of these modes is perhaps less clear than in the spatial case, but they nevertheless prove to be useful in determining the probability distribution $P(W)$ in the next section. By analogy with Eq. (21), the operators $\hat{a}_n(\tau)$ can be expanded as

$$\hat{a}_n(\tau)=\beta_n^{1/2}\sum_k \hat{b}_k^{(n)}\Psi_k(\tau), \tag{35}$$

where the $\Psi_k(\tau)$ are orthonormal over the interval $-\infty<\tau+\infty$, and $\beta_n^{1/2}$ is inserted for convenience. Requiring that the temporal coherence modes $\Psi_k(\tau)$ are excited in a statistically uncorrelated fashion implies that $\langle\hat{b}_k^{(n)\dagger}\hat{b}_l^{(m)}\rangle=\lambda_k\delta_{kl}\delta_{nm}$, where the λ_k are the eigenvalues of the equation

$$\int_{-\infty}^{\infty}d\tau'\, G_\tau(\tau,\tau')\Psi_k(\tau')=\lambda_k\Psi_k(\tau). \tag{36}$$

It is convenient to normalize the eigenvalues λ_k by the total mean number of photons (energy) emitted in the Stokes pulse, which is given by

$$\begin{aligned}\langle\hat{W}\rangle&=\int_A d^2\rho\int_{-\infty}^{\infty}d\tau\, I_S(\boldsymbol{\rho},\tau)\\&=\int_{-\infty}^{\infty}d\tau\, G_\tau(\tau,\tau),\end{aligned} \tag{37}$$

where the intensity $I_S(\boldsymbol{\rho},\tau)$ is given in Eq. (9), and use was made of Eqs. (15), (16), and (30). Values of $\langle\hat{W}\rangle$ can be found from Fig. 3 of Ref. 7 (using the relation $\langle\hat{W}\rangle=\mathscr{F}^2N_S$). The correlation function can be written as

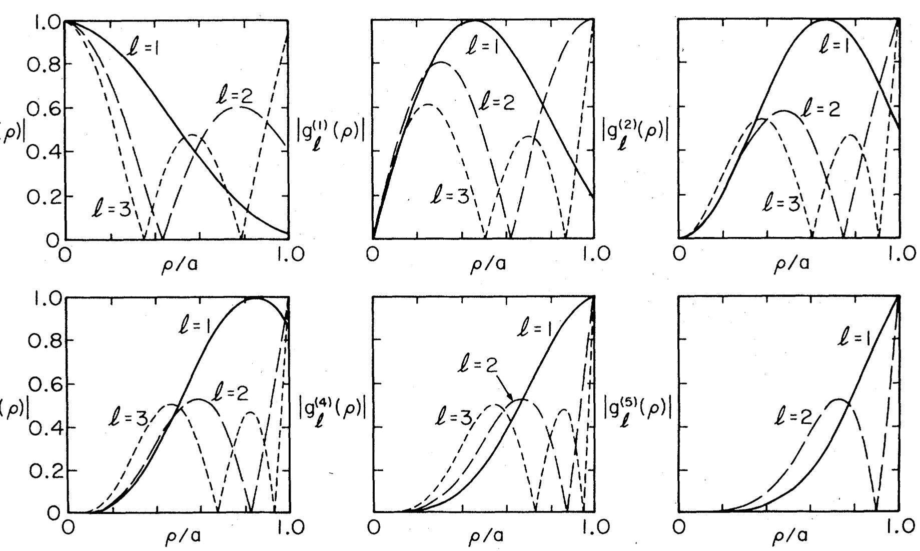

FIG. 2. Absolute value of radial eigenfunctions $g_l^{(k)}(\rho)$ for the case $\mathscr{F}=3$.

$$G_\tau(\tau_1\tau_2)=\sum_k \lambda_k \Psi_k(\tau_1)\Psi_k^*(\tau_2) \tag{38}$$

which, when used in Eq. (37), leads to

$$\langle \hat{W} \rangle = \sum_k \lambda_k \ . \tag{39}$$

Thus λ_k is the mean total energy emitted into temporal mode $\Psi_k(\tau)$. Defining normalized eigenvalues $\tilde{\lambda}_k$ by

$$\tilde{\lambda}_k = \lambda_k / \langle \hat{W} \rangle \tag{40}$$

leads to the interpretation that $\beta_n \tilde{\lambda}_k$ is the fraction of the total energy emitted into the spatial-temporal mode $\phi_n(\rho)\Psi_k(\tau)$.

Table II gives values for some of the normalized eigenvalues $\tilde{\lambda}_k$, determined numerically from Eq. (36) for different values of $\Gamma\tau_L$ and gL. The laser pulse is assumed to be Gaussian in time with full width at half maximum equal to τ_L. The given values of the gain coefficient g [see Eq. (6)] correspond to the intensity at the peak of the laser pulse. It can be seen from Table II that when the ratio $\Gamma\tau_L/gL$ is less than unity, a single temporal mode is dominantly excited, i.e., $\tilde{\lambda}_1$ is much larger than all the other $\tilde{\lambda}_k$. This means that the Stokes light emitted during the laser pulse is temporally coherent. This corresponds to the transient regime of SRS. On the other hand, when $\Gamma\tau_L/gL$ is greater than unity, the number of temporal modes significantly excited scales as $\Gamma\tau_L/gL$. In this case temporal incoherence exists during the Stokes pulse.

TABLE II. Normalized temporal eigenvalues $\tilde{\lambda}_k$ corresponding to excitation strengths of temporal coherence modes $\Psi_k(\tau)$, determined from Eqs. (36) and (40). The number of significantly excited modes increases with increasing $\Gamma\tau_L/gL$.

$\Gamma\tau_L$	gL	$\Gamma\tau_L/gL$	k	$\tilde{\lambda}_k$
1	50	0.02	1	0.997
			2	0.274(−2)[a]
			3	0.484(−4)
			4	0.535(−5)
7	7	1	1	0.757
			2	0.131
			3	0.414(−1)
			4	0.190(−1)
			5	0.108(−1)
			6	0.702(−2)
30	5	6	1	0.274
			2	0.169
			3	0.112
			4	0.775(−1)
			5	0.561(−1)
			6	0.422(−1)
			7	0.327(−1)
			8	0.260(−1)
			9	0.212(−1)
			10	0.176(−1)
			11	0.149(−1)
			12	0.127(−1)
			13	0.111(−1)
			14	0.971(−2)

[a]The number enclosed in parentheses indicates the exponent of the multiplicative factor of 10.

C. Pulse-energy probability distribution function

Using the mode expansion of the field operator developed in Sec. IV B, the probability density function $P(W)$ for observing a Stokes pulse with energy W can be calculated.[8,10,12,14] First, the pulse-energy operator $\hat{W}$ is written as

$$\hat{W}=\frac{c}{2\pi\hbar\omega_S}\int_A d^2\rho \int_{-\infty}^{\infty} d\tau \hat{E}_S^{(-)}(\rho,L,\tau)E_S^{(+)}(\rho,L,\tau)$$

$$=\sum_{n,k} \beta_n \hat{b}_k^{(n)\dagger}\hat{b}_k^{(n)} \ , \tag{41}$$

where the last step results from Eqs. (22a) and (35), and the orthonormality of the eigenfunctions.

The probability density function is given by

$$P(W)=\frac{1}{2\pi}\int_{-\infty}^{\infty} d\xi \exp(-i\xi W)C(\xi) \ , \tag{42}$$

where $C(\xi)$, the characteristic function, may be written as

$$C(\xi)=\langle :\exp(i\xi\hat{W}): \rangle \ . \tag{43}$$

The colons, which indicate normal ordering, have been added to simplify the calculation, and since we are dealing only with fields containing many photons, they do not significantly affect the value of $C(\xi)$.

The expectation value in Eq. (43) is taken in the initial state with all atoms in their ground states and no Stokes photons present. It is known[8,12,20] that, since the atoms are being treated as a continuum field of harmonic oscillators, the quantum expectation value of a normally ordered quantity can be represented by a classical average over a set of independent, complex, random variables $b_k^{(n)}$, which are Gaussian distributed with variance given by $\langle |b_k^{(n)}|^2 \rangle = \lambda_k$, i.e.,

$$C(\xi)=\prod_{n,k}\int d^2 b_k^{(n)} P(b_k^{(n)})\exp(i\xi\beta_n |b_k^{(n)}|^2) \ , \tag{44a}$$

where $P(b_k^{(n)})$ is the density function for $b_k^{(n)}$,

$$P(b_k^{(n)})=\frac{1}{\pi\lambda_k}\exp(-|b_k^{(n)}|^2/\lambda_k) \ . \tag{44b}$$

Note that the Gaussian nature of the $b_k^{(n)}$, along with their being uncorrelated, means that they are statistically independent. Equation (44) can be evaluated to give

$$C(\xi)=\prod_{n,k}(1-i\xi\beta_n\lambda_k)^{-1} \ . \tag{45}$$

The probability distribution is then found from Eq. (42) to be

$$P(W)=\lim_{N,K\to\infty}\sum_{n=1}^{N}\sum_{k=1}^{K} C_{nk}^{(N,K)}\exp(-W/\beta_n\lambda_k) \ , \tag{46a}$$

where

$$C_{nk}^{(N,K)}=(\beta_n\lambda_k)^{NK-2}\prod_{m(\neq n)}^{N}\prod_{l(\neq k)}^{K}(\beta_n\lambda_k-\beta_m\lambda_l)^{-1} \ . \tag{46b}$$

This formula is valid when the eigenvalues are nondegenerate. In the present case some of the eigenvalues are twofold degenerate and thus a limit must be taken in Eq. (46) as $\beta_m \lambda_m \rightarrow \beta_n \lambda_k$ (Ref. 17). We will not present the rather lengthy expression for $P(W)$ that results. This result is a generalization of Eqs. (15), (4), and (9) in Refs. 8, 10, and 12, respectively. In the transient limit, the λ_k here are the same as the λ_i in Ref. 8. The β_n here are related to the λ_n in Ref. 12 by $\beta_n = \lambda_n / \pi$.

It should be emphasized that the above approach to obtain $P(W)$ is very similar in some respects to that used in theories of photoelectron counting.[14] In those treatments, the autocorrelation function was assumed to factorize into spatial and temporal parts, as in our Eq. (16), and our Eq. (46) was obtained. The temporal part was usually taken to be stationary in time. In the present treatment we have explicitly calculated the correlation function for SRS and found that it factorizes. The temporal dynamics for SRS is nonstationary. In addition, we have calculated the distribution function $P(W)$ for the energy of pulses with macroscopic amounts of energy, in contrast to the case of photoelectron counting.

V. RESULTS AND DISCUSSION

Figure 3 shows several examples of the probability density function $P(W)$ for different values of $\mathcal{F}$, $\Gamma\tau_L$, and gL. The behavior of $P(W)$ is in agreement with simple ideas of statistics. When $\mathcal{F}$ and $\Gamma\tau_L/gL$ are small ($\lesssim 1$), a single spatial-temporal mode is dominant in the Stokes emission. Then $P(W)$ is nearly a negative exponential. This describes Bose-Einstein statistics in the limit of a large mean number of photons. If the probability $p(n)$ of observing n photons within a certain counting time is given by a Bose-Einstein distribution, then

$$p(n) = \frac{\langle n \rangle^n}{(1+\langle n \rangle)^{1+n}} \rightarrow \frac{1}{\langle n \rangle} \exp\left[-\frac{n}{\langle n \rangle}\right], \quad (47)$$

where the limit holds for $\langle n \rangle \gg 1$. This distribution is also obeyed by a single-mode thermal light source at high temperature, when the photon-counting time is much smaller than the coherence time of the light.[3,22] One may think of the SRS process as amplifying the zero-point radiation field, which is Gaussian distributed, leading to the exponential distribution for instantaneous intensity in Eq. (47).[8] (Note, however, that in the normally ordered treatment used here, the emission actually appears to arise from the zero-point motion of the dipole moments of the atoms, rather than the zero-point field.[21])

When $\mathcal{F}$ is small ($\cong 1$) but $\Gamma\tau_L/gL$ is large ($\gg 1$), a single spatial mode and many temporal modes are excited. The Stokes emission is spatially coherent but temporally incoherent. The distribution $P(W)$ is seen in Fig. 3 to become peaked near the mean and to narrow as $\Gamma\tau_L/gL$ increases. Corresponding behavior occurs for a thermal source when the photon-counting time becomes longer than the coherence time. In this case $p(n)$ for the thermal source is a Poisson distribution,[3] which becomes Gaussian in the limit of large mean photon number $\langle n \rangle$,

$$p(n) = \frac{\langle n \rangle^n}{n!} \exp(-\langle n \rangle) \rightarrow \frac{1}{(2\pi\langle n \rangle)^{1/2}} \exp\left[\frac{(n-\langle n \rangle)^2}{2\langle n \rangle}\right]. \quad (48)$$

This behavior, for both Stokes light and thermal light, can be understood in the following way. Let n_i be the number of photons emitted in the ith coherence time (temporal mode). Then the detected photon number is the sum of the numbers in each mode

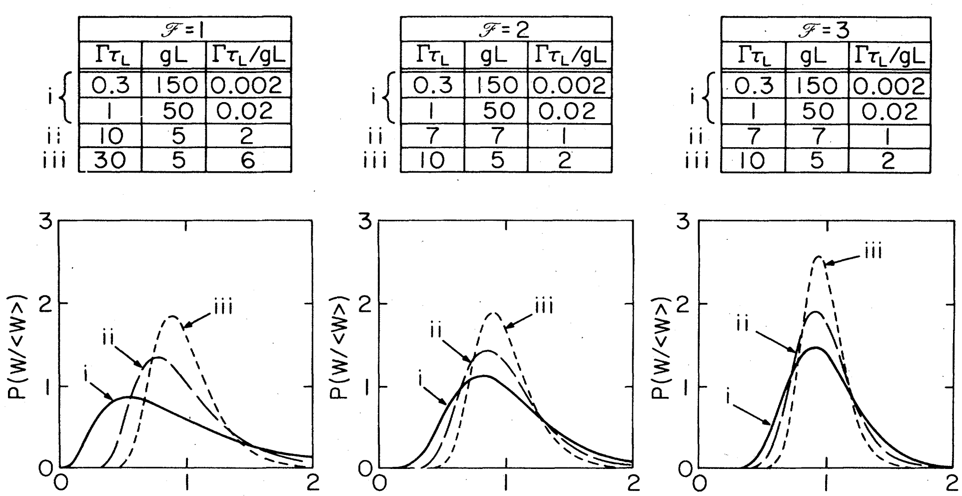

FIG. 3. Probability density function $P(W/\langle W \rangle)$ for Stokes pulse energy W, for various Fresnel numbers $\mathcal{F}$, and various collisional dephasing rates Γ. The effect of Γ enters through the factor $\Gamma\tau_L/gL$, where τ_L is the laser pulse duration and gL is the number of gain lengths in the medium. As either $\mathcal{F}$ or $\Gamma\tau_L/gL$ is increased, $P(W/\langle W \rangle)$ narrows, illustrating that the Stokes pulse-energy fluctuations decrease. This is associated with the decrease of the spatial and/or temporal coherence of the Stokes emission.

$$n = n_1 + n_2 + n_3 + \cdots \; . \tag{49}$$

In the limit of many modes, the well-known central-limit theorem states that $p(n)$ will be a Gaussian distribution centered at $n = \langle n \rangle$, with width decreasing as the square root of the number of modes present. The departures from exponential seen in Fig. 3 are indications of the tendency of $P(W)$ to become Gaussian when many modes are excited.

Physically, one may view each coherence time as an independent chance for the atoms to emit a spontaneous photon, which would be amplified to the macroscopic level. In a pure single-mode case it would be most likely that the atoms do not emit a photon (negative exponential distribution). As more modes are added, it becomes less likely that all modes will fail to emit a photon (departure from exponential).

When $\Gamma\tau_L/gL$ is small ($\ll 1$) but $\mathscr{F}$ is large ($\gg 1$), a single temporal mode and many spatial modes are excited. The Stokes emission is temporally coherent but spatially incoherent. As $\mathscr{F}$ increases, the number of spatial modes increases as $\mathscr{F}^2$, causing the peak of $P(W)$ to move toward the mean energy $\langle W \rangle$. This behavior is analogous to that observed above as the number of temporal modes was increased. Again an equation of the form of Eq. (49) applies, where n_i is the number of photons emitted into the ith spatial mode.

When both $\Gamma\tau_L/gL$ and $\mathscr{F}$ are much larger than unity, the Stokes emission is temporally and spatially incoherent. The number of spatial-temporal modes that are significantly excited is approximately given by $\mathscr{F}^2\Gamma\tau_L/gL$. Again the peak of the distribution $P(W)$ moves toward the mean energy as this number increases.

In conclusion, it has been found that microscopic quantum fluctuations associated with spontaneous Raman scattering can give rise to large fluctuations in the total energy of generated Stokes pulses. This occurs for pulses containing macroscopic amounts (say, $1\mu J$, or 10^{13} photons) of energy. The fluctuations are in great excess of the shot noise limit. As the Fresnel number $\mathscr{F}$ and/or the ratio $\Gamma\tau_L/gL$ increases, the Stokes emission becomes spatially and/or temporally incoherent, and the pulse-energy distribution function $P(W)$ narrows. It has been shown how to determine the spatial and temporal coherence modes from knowledge of the Stokes field correlation function, using concepts known in classical coherence theory.[14,15,18]

The present theory is restricted to Fresnel numbers not less than about unity, although the precise range of validity is difficult to ascertain. For smaller Fresnel numbers, numerical simulations of the type in Ref. 23 may prove to be useful.

Recent experimental studies of SRS in molecular hydrogen have illustrated the present predictions concerning the dependence of the pulse-energy fluctuations on Fresnel number and collisional dephasing rate.[24]

ACKNOWLEDGMENTS

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APPENDIX A: PARAXIAL SOLUTION OF MAXWELL-BLOCH EQUATIONS

A method is presented here for solving the coupled equations (2) and (5), which may be written in the moving frame, where $\tau = t - z/c$,

$$\frac{\partial}{\partial\tau}Q(\mathbf{r},\tau) = -\Gamma Q(\mathbf{r},t) - i\kappa_1^* E_L(\tau)E_S^{(+)}(\mathbf{r},\tau) + F(\mathbf{r},\tau) \; , \tag{A1}$$

$$\left[\nabla_T^2 + 2ik_S\frac{\partial}{\partial z}\right]E_S^{(+)}(\mathbf{r},t) = \frac{2\kappa_2^*\omega_S}{c}E_L^*(\tau)Q(\mathbf{r},t) \; . \tag{A2}$$

In obtaining these equations, the paraxial approximation was used, i.e., $\partial^2 E_S^{(+)}/\partial z^2 \ll k_S \partial E_S^{(+)}/\partial z$. Also, use was made of the slowly-varying-envelope approximation, i.e., $\partial G/\partial\tau \ll \omega G$, where G stands for either $E_S^{(+)}$, E_L, or Q, and ω is ω_S, ω_L, or ω_{31}, respectively. Finally, it has been assumed that $E_L(\tau)$ is independent of x and y throughout a cylindrical region in which the equations are to be solved. ∇_T^2 above is the transverse Laplacian $\partial^2/\partial x^2 + \partial^2/\partial y^2$.

To solve Eqs. (A1) and (A2) define the Fourier transform variables by

$$\tilde{G}(\boldsymbol{\beta},k,\tau) = (2\pi)^{-3}\int dz \int d^2\rho \, G(\boldsymbol{\rho},z,\tau) \times \exp[-i(kz + \boldsymbol{\beta}\cdot\boldsymbol{\rho})] \; , \tag{A3}$$

where $\boldsymbol{\rho} = (x,y)$ is the radial vector. In the transform space, Eqs. (A1) and (A2) become

$$\frac{\partial}{\partial\tau}\tilde{Q} = -\Gamma\tilde{Q} - i\kappa_1^* E_L \tilde{E}_S^{(+)} + \tilde{F} \; , \tag{A4}$$

$$(-\beta^2 - 2k_s k)\tilde{E}_S^{(+)} = \frac{2\kappa_2^*\omega_S}{c}E_L^*\tilde{Q} \; . \tag{A5}$$

From Eq. (A5) a solution for $\tilde{E}_S$ is found, which is substituted into Eq. (A4). The result is a differential equation whose solution is

$$\tilde{Q}(\boldsymbol{\beta},k,\tau) = \tilde{Q}(\boldsymbol{\beta},k,0)\exp\left[-\Gamma\tau - \frac{i\,2k_S\alpha(\tau)}{\beta^2 + 2k_S k}\right] + \int_0^\tau d\tau' \, \tilde{F}(\boldsymbol{\beta},k,\tau)\exp\left[-\Gamma(\tau-\tau') - \frac{i\,2k_S[\alpha(\tau)-\alpha(\tau')]}{\beta^2 + 2k_S k}\right] , \tag{A6}$$

where

$$\alpha(\tau) = \kappa_1\kappa_2\int_0^\tau |E_L(\tau')|^2 d\tau' \; . \tag{A7}$$

This solution is then substituted back into Eq. (A5) to obtain the result for $\tilde{E}_S^{(+)}(\boldsymbol{\beta},k,\tau)$. In order to carry out the inverse transform back to $\mathbf{r}$ space, it is convenient to change variables to $p=k+\beta^2/2k_S$. Then the inverse transform can be written as

$$E_S^{(+)}(\rho,z,\tau)=\frac{\kappa_2^* E_L(\tau)}{(2\pi)^3}\int d^3r'\int dp\int d^2\beta\frac{1}{p}\exp[ip(z-z')-i\beta^2(z-z')/2k_S+i\boldsymbol{\beta}\cdot(\boldsymbol{\rho}-\boldsymbol{\rho}')]$$
$$\times\left[Q(\mathbf{r}',0)e^{-\Gamma\tau}\exp[-i\alpha(\tau)/p]+\int_0^\tau d\tau' F(\mathbf{r}',\tau')e^{-\Gamma(\tau-\tau')}\exp\{-i[\alpha(\tau)-\alpha(\tau')]/p\}\right]. \tag{A8}$$

The p and $\boldsymbol{\beta}$ integrals above can be carried out using the calculus of residues to yield[26]

$$E_S^{(+)}(\mathbf{r},\tau)=\frac{k_S\kappa_2^* E_L(\tau)}{2\pi}\int d^3r'\frac{1}{z-z'}\exp\left[\frac{-k_S i\,|\boldsymbol{\rho}-\boldsymbol{\rho}'|^2}{2(z-z')}\right]$$
$$\times\left[Q(\mathbf{r}',0)e^{-\Gamma\tau}I_0([4\alpha(\tau)(z-z')]^{1/2})+\int_0^\tau d\tau' F(\mathbf{r}',\tau')e^{-\Gamma(\tau-\tau')}I_0(\{4[\alpha(\tau)-\alpha(\tau')](z-z')\}^{1/2})\right]. \tag{A9}$$

This solution is exactly that given in Eq. (8) in the text.

APPENDIX B: NONPARAXIAL SOLUTION OF MAXWELL-BLOCH EQUATIONS

The coupled equations (2) and (5) can be solved without making the paraxial approximation if it is assumed that the laser field in the cylindrical region is constant throughout its duration, i.e., $E_L(\mathbf{r},\tau)=A_L\,(0\le\tau\le\tau_L)$. The equations can then be written

$$\frac{\partial}{\partial t}q(\mathbf{r},t)=-(i\omega_S+\Gamma)q(\mathbf{r},t)-i\kappa_1^* A_L A_S(\mathbf{r},t)+f(\mathbf{r},t)\,, \tag{B1}$$

$$\left[\nabla^2-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right]A_S(\mathbf{r},t)=\frac{2\kappa_2^*}{c\omega_S}A_L^*\frac{\partial^2}{\partial t^2}q(\mathbf{r},t)\,, \tag{B2}$$

where the following variables have been defined:

$$A_S(\mathbf{r},t)\equiv E_S^{(+)}(\mathbf{r},t)e^{-i(\omega_S t-k_S z)}\,,$$
$$q(\mathbf{r},t)\equiv Q(\mathbf{r},t)e^{-i(\omega_S t-k_S z)}\,,$$
$$f(\mathbf{r},t)\equiv F(\mathbf{r},t)e^{-i(\omega_S t-k_S z)}\,.$$

Changing to a moving-frame time $\tau=t-z/c$, (B1) and (B2) become

$$\frac{\partial}{\partial\tau}q(\mathbf{r},\tau)=-(i\omega_S+\Gamma)q(\mathbf{r},\tau)-i\kappa_2^* A_L A_S(\mathbf{r},\tau)+f(\mathbf{r},\tau)\,, \tag{B3}$$

$$\left[\nabla^2-\frac{2}{c^2}\frac{\partial^2}{\partial z\partial\tau}\right]A_S(\mathbf{r},\tau)=\frac{2\kappa_2^*}{c\omega_S}A_L^*\frac{\partial^2}{\partial\tau^2}q(\mathbf{r},\tau)\,. \tag{B4}$$

To solve these equations, define the Laplace transform variables by

$$\widetilde{G}(\mathbf{r},s)=\int_0^\infty d\tau\, e^{-s\tau}G(\mathbf{r},\tau)\,,$$

where $G(\mathbf{r},\tau)$ stands for A_S, q, or f. Equation (B3) then leads to

$$\widetilde{q}(\mathbf{r},s)=\frac{q(\mathbf{r},0)-i\kappa_1^* A_L\widetilde{A}_S(\mathbf{r},s)+\widetilde{f}(\mathbf{r},s)}{s+i\omega_S+\Gamma}\,, \tag{B5}$$

and Eq. (B4) leads to

$$\left[\nabla^2-\frac{2s}{c}\frac{\partial}{\partial z}\right]\widetilde{A}_S(\mathbf{r},s)=\frac{2\kappa_2^*}{c\omega_S}A_L^*\left[s^2\widetilde{q}(\mathbf{r},s)-sq(\mathbf{r},0)-\frac{\partial}{\partial\tau}q(\mathbf{r},\tau)\bigg|_{\tau=0}\right]-\frac{2}{c}\frac{\partial}{\partial z}A_S(\mathbf{r},0)\,. \tag{B6}$$

Substituting (B5) into (B6) and defining the new variable

$$\mathscr{A}_S(\mathbf{r},s)=e^{-sz/c}\widetilde{A}_S(\mathbf{r},s)\,, \tag{B7}$$

it is found that

$$(\nabla^2+\ell^2)\mathscr{A}_S(\mathbf{r},s)=-4\pi\mathscr{N}(\mathbf{r},s)\,, \tag{B8}$$

where

$$\ell^2=-\frac{s^2}{c^2}+i\frac{2\kappa_1\kappa_2|A_L|^2}{c\omega_S}\frac{s^2}{s+i\omega_S+\Gamma}\,,$$
$$\ell\cong i\frac{s}{c}\left[1-i\frac{\kappa_1\kappa_2|A_L|^2}{\omega_S}\frac{c}{s+i\omega_S+\Gamma}\right], \tag{B9}$$

and

$$\mathscr{N}(\mathbf{r},s)=\frac{e^{-sz/c}}{4\pi}\left[\frac{2\kappa_2^* A_L^*}{c\omega_S}\left[\frac{s(i\omega_S+\Gamma)}{s+i\omega_S+\Gamma}q(\mathbf{r},0)+\frac{s^2}{s+i\omega_S+\Gamma}\widetilde{f}(\mathbf{r},s)-\frac{\partial}{\partial\tau}q(\mathbf{r},\tau)\bigg|_{\tau=0}\right]-\frac{2}{c}\frac{\partial}{\partial z}A_S(\mathbf{r},0)\right]. \tag{B10}$$

The solution of Eq. (B8) [with (B7)] is

$$\tilde{\mathscr{A}}_S(\mathbf{r},s)=\int d^3r'\frac{\exp(i\not{k}\,|\,\mathbf{r}-\mathbf{r}'\,|\,)}{|\,\mathbf{r}-\mathbf{r}'\,|}\mathscr{N}(\mathbf{r}',s)\ . \tag{B11}$$

Neglecting the initial derivatives $\partial q(\mathbf{r},0)/\partial\tau$ and $\partial A_S(\mathbf{r},0)/\partial z$, Eq. (B11) can be inverse Laplace transformed,[26] using $\Gamma \ll \omega_S$, to give

$$\begin{aligned}A_S(\mathbf{r},t)=\frac{k_S\kappa_2^*A_L^*}{2\pi}\int\frac{d^3r'}{|\,\mathbf{r}-\mathbf{r}'\,|}\exp\left[i\frac{\kappa_1\kappa_2\,|\,A_L\,|^2}{c}\,|\,\mathbf{r}-\mathbf{r}'\,|\right]\exp\left[-(i\omega_S+\Gamma)\left[\tau-\frac{|\,\mathbf{r}-\mathbf{r}'\,|}{c}+\frac{z-z'}{c}\right]\right]\\ \times\left\{q(\mathbf{r}',0)I_0\left[\left[4\kappa_1\kappa_2\,|\,A_L\,|^2\,|\,\mathbf{r}-\mathbf{r}'\,|\left[\tau-\frac{|\,\mathbf{r}-\mathbf{r}'\,|}{c}+\frac{z-z'}{c}\right]\right]^{1/2}\right]\right.\\ \left.+\int_0^\tau d\tau' f(\mathbf{r}',\tau')I_0\left[\left[4\kappa_1\kappa_2\,|\,A_L\,|^2\,|\,\mathbf{r}-\mathbf{r}'\,|\left[\tau-\tau'-\frac{|\,\mathbf{r}-\mathbf{r}'\,|}{c}+\frac{z-z'}{c}\right]\right]^{1/2}\right]\right\}.\end{aligned} \tag{B12}$$

The r' integral is over the cylinder volume. Note that this solution contains the retarded time $\tau-|\,\mathbf{r}-\mathbf{r}'\,|/c-(z-z')/c$, which accounts correctly for propagation through the medium. Also, unlike the paraxial solution, there is no divergence of the solution as $z'\to z$, since the correct Green's function, proportional to $1/|\,\mathbf{r}-\mathbf{r}'\,|$, has been used. It is not possible, however, to generalize this method to an arbitrary laser spatial or temporal profile. In the high-gain limit, when only propagation close to the z axis is important, this solution reduces to that of Appendix A.

APPENDIX C: FACTORIZATION OF SPATIAL CORRELATION FUNCTION

Here Eqs. (33) and (34) are derived and the form of the integral kernel $U^{(k)}(\rho',\rho)$ is found. The two-dimensional spatial correlation function is given, from Eq. (19), by

$$G_\rho(\boldsymbol{\rho}_1,\boldsymbol{\rho}_2)=\frac{1}{A}\frac{2J_1(2\mathscr{F}\,|\,\boldsymbol{\rho}_1-\boldsymbol{\rho}_2\,|\,/a)}{2\mathscr{F}\,|\,\boldsymbol{\rho}_1-\boldsymbol{\rho}_2\,|\,/a}\exp\left[-i\frac{\mathscr{F}}{a^2}(\rho_1^2-\rho_2^2)\right], \tag{C1}$$

where $\rho_i\equiv|\,\boldsymbol{\rho}_i\,|$. In polar coordinates the radial vectors are $\boldsymbol{\rho}_i=(\rho_i\cos\theta_i,\rho_i\sin\theta_i)$, for $i=1,2$. Using Gegenbauer's theorem,[27] Eq. (C1) can be written as

$$G_\rho(\boldsymbol{\rho},\boldsymbol{\rho}_2)=\frac{4}{A}\exp\left[-i\frac{\mathscr{F}}{a^2}(\rho_1^2-\rho_2^2)\right]\sum_{n=-\infty}^{\infty}{}' e^{in(\theta_1-\theta_2)}\sum_{j=|n|}^{\infty}(j+1)\frac{J_{j+1}(2\mathscr{F}\rho_1/a)J_{j+1}(2\mathscr{F}\rho_2/a)}{4\mathscr{F}^2\rho_1\rho_2/a^2}, \tag{C2}$$

where the prime indicates that j takes on only even values if $|\,n\,|$ is even and only odd values if $|\,n\,|$ is odd. Thus the integral equation (32),

$$\int_0^a d\rho'\int_0^{2\pi}d\theta'\,\rho' G_\rho(\boldsymbol{\rho},\boldsymbol{\rho}')\phi_m(\boldsymbol{\rho}')=\beta_m\phi_m(\boldsymbol{\rho})\ , \tag{C3}$$

may be simplified by factorizing the eigenfunctions $\phi_m(\boldsymbol{\rho})$ into an angular part and a radial part, labeled by integers l and k, so that

$$\phi_m(\boldsymbol{\rho})=e^{ik\theta}g_l^{(k)}(\rho)\exp\left[-i\frac{\mathscr{F}}{a^2}\rho^2\right]. \tag{C4}$$

The θ' integration in Eq. (C3) is then easily accomplished, leaving the radial equation

$$\int_0^a d\rho'\,U^{(k)}(\rho',\rho)g_l^{(k)}(\rho')=\beta_l^{(k)}g_l^{(k)}(\rho)\ , \tag{C5}$$

where the radial integral kernel is given by

$$U^{(k)}(\rho',\rho)=\frac{2}{\mathscr{F}^2}\sum_{j=|k|}^{\infty}{}'(j+1)\frac{1}{\rho}J_{j+1}\left[\frac{2\mathscr{F}\rho}{a}\right]\times J_{j+1}\left[\frac{2\mathscr{F}\rho'}{a}\right]. \tag{C6}$$

For each value of k, Eq. (C5) has an infinte number of eigensolutions. We determine those with the largest eigenvalues by discretizing the ρ' variable and using a standard numerical technique for matrix diagonalization. The calculation typically converges for a matrix size of between 60×60 and 80×80. Also it should be pointed out that the sum in Eq. (C6) is truncated at $j=6\mathscr{F}$, since terms beyond this are negligible.

If only the eigenvalues, and not the eigenfunctions, are required, a faster numerical technique is that discussed in the Appendix of Ref. 17.

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