Transverse effects in stimulated Raman scattering

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We formulate the quantum theory of stimulated Raman scattering in three dimensions. The spatial coherence function of the Stokes pulse is found as a function of the Fresnel number. The pulse-energy distribution function is calculated and compared with recent measurements.

Stimulated Raman scattering (SRS) is one of the best known nonlinear optical processes. It amounts to the transformation of the pump field into Stokes photons which have a different (smaller) frequency and propagate in the direction parallel to the pump field photons. An interesting feature of the SRS is the interplay of the spontaneous emission of the Stokes photons in the initial stage of the process with the stimulated emission which dominates when the density of Stokes photons becomes sizable. These two processes lead to large observable effects which have been called macroscopic quantum fluctuations. The quantum fluctations of the electromagnetic field in the vacuum state, which are responsible for the spontaneous emission of the Stokes photons, are amplified during the process and manifest themselves as fluctuations of the total Stokes pulse energy. This effect has been discussed by Raymer, Rzażewski, and Mostowski[1] and Rzażewski, Lewenstein, and Raymer.[2] These papers predicted large ($\sim 100\%$) fluctuations of the output Stokes pulse energy in the transient regime and smaller fluctuations in the steady-state regime. The distribution of the Stokes pulse energies in the transient regime was the subject of a recent study of Walmsley and Raymer[3] who found qualitative agreement with the Raymer *et al.*[1] theory.

The Raymer *et al.*[1] theory is restricted to a particular shape of the amplifier, namely such that the Fresnel number is equal to 1. Only in such case can the application of the one-dimensional theory of the SRS as formulated by Mostowski and Raymer[4] be justified. For larger and smaller F the wave front of the Stokes pulse cannot be treated as homogeneous, so the full three-dimensional theory of the Stokes field propagation is needed. It should be noted that even for $F=1$ the wave front is only approximately homogeneous and a three-dimensional theory is also required in this case, although it leads only to small corrections.

In the present paper we will study the Fresnel number dependence of the total Stokes pulse energy distribution in the transient regime. Our analysis is restricted to the $F \gtrsim 1$ region since we will neglect waveguide effects which play a role for $F \lesssim 1$.

As has been shown by Mostowski and Raymer,[4] the quantum theory of SRS has its counterpart in the quantum theory of superfluorescence initiation, formulated by Haake, Haus, and Glauber[5] and Polder, Schuurmans, and Vrehen.[6] In fact, all equations of the SRS case can be translated to the superfluorescence case by a suitable substitution of constants. Such a comparison can be found in Ref. 4.

Recently, we have formulated a three-dimensional theory of superfluorescence initiation.[7] This theory is a direct extension of the Haake *et al.*[5] theory to three dimensions. This theory can be now translated, by the same substitutions as in the one-dimensional case, to the SRS case.

The Raman equations read

$$\frac{d}{dt}Q(t) = -i\kappa_1^* E_L(\vec{r},t)E_s^+(\vec{r},t)$$

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)[E_S(\vec{r},t)\,e^{i(\omega_s t - k_S z)}] = \frac{4\pi n\hbar}{c^2}\kappa_1^*\frac{\partial^2}{\partial t^2}[E_L(\vec{r},t)Q^+(\vec{r},t)\,e^{i(\omega_S t - k_S z)}] \tag{1}$$

where Q is the two-photon polarization field (see Ref. 4), E_S, E_L are the positive frequency components of the Stokes and the pump field, respectively, n is the number density of atoms and

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

Here d_{ij} denotes a transition dipole moment in the active atom, ω_{ij} is a corresponding transition frequency, ω_L and ω_S are the laser pump and the Stokes field frequencies. Such a definition of the coupling constant κ_1 was used in Refs. 1–4.

Assume that the pump field E_L is a constant over the cross section of the cylinder and constant in time. If the waveguide effects are neglected, the solution of these equations reads

$$E_S(\vec{r},t) = \int K(\vec{r},\vec{r}',t)Q^+(0,\vec{r}') \ , \tag{2}$$

where

$$K = \frac{\sqrt{2\pi^3}n\hbar\omega_S^3\kappa_1^* E_L}{c^3|\vec{r}-\vec{r}'|}\exp\left(i[(z-z')-|\vec{r}-\vec{r}'|]\frac{\omega_S}{c} - i\frac{\alpha}{8}|\vec{r}-\vec{r}'|\right)I_0\left\{\left[\alpha\left(\tau + \frac{z-z'}{c} - \frac{|\vec{r}-\vec{r}'|}{c}\right)|\vec{r}-\vec{r}'|\right]^{1/2}\right\} ,$$

I_0 is the modified Bessel function, $\alpha = 16\pi\hbar|\kappa_1 E_L|^2\omega_s/c$, $\tau = t - z/c$.

This is the solution of the Heisenberg equations for the Stokes field operator.

The constant α is closely related to the steady-state Raman gain constant g. If Γ denotes the relaxation rate then we have $g = \frac{1}{2}\Gamma^{-1}\alpha$. In this paper, however, we consider transient effects only, in other words all relaxation times are longer than the duration of the pumping pulse. Therefore, the characteristic time scale is not Γ^{-1} but rather the "Raman time" $(\alpha L)^{-1}$

and we do not use the concept of the steady-state gain constant g.

The average intensity of the Stokes pulse can be found directly from (2) by taking the average value of $|E_S|^2$ for $\vec{r}$ at the right-hand side surface of the sample. Making use of the commutation relation

$$[Q^+(\vec{r}_1,0),Q(\vec{r}_2,0)]=\frac{1}{n}\delta^{(3)}(\vec{r}_1-\vec{r}_2)]$$

we find

$$I(\vec{r},t)=\frac{2\pi^3\hbar^2 n\omega_s^6|\kappa_1^* E_L|^2}{c^6}\int d_3r'\frac{1}{|\vec{r}-\vec{r}'|^2}I_0^2\left\{\left[\alpha\left(\tau+\frac{z-z'}{c}-\frac{|\vec{r}-\vec{r}'|}{c}\right)|\vec{r}-\vec{r}'|\right]^{1/2}\right\} . \quad (3)$$

In the high-gain limit this reduces to

$$I(\vec{r},t)=\frac{F}{2\alpha\tau L}\left[\frac{n\hbar\omega_S^4|\kappa_1^* E_L|}{c^5}\right]^2\exp[(\alpha\tau L)^{1/2}] , \quad (4)$$

where $\vec{r}=(x,y,L)$.

This formula, in fact, has been derived by Raymer and Mostowski (Ref. 4) with the help of the one-dimensional theory and some hand-waving arguments. Here it has been rigorously derived.

The three-dimensional theory can, in fact, predict new features of the Stokes pulse. An interesting quantity is the correlation function $G(\vec{r}_1,\vec{r}_2,t)=\langle E_S^+(\vec{r}_1,t)E(\vec{r}_2,t)\rangle$ calculated for $\vec{r}_1$ and $\vec{r}_2$ on the right-hand side surface of the cylinder. This function is a measure of the field variations over the output area. The formula for $G(\vec{r}_1,\vec{r}_2,t)$ is rather complicated but in the high-gain limit it simplifies to

$$G(\vec{r}_1,\vec{r}_2,t)=\langle I(t)\rangle\frac{2J_1[(F/2a)|\vec{r}_1-\vec{r}_2|]}{(F/2a)|\vec{r}_1-\vec{r}_2|}\times\exp\left[-i\frac{F}{a^2}(|\vec{\rho}_1|^2-|\vec{\rho}_2|^2)\right] , \quad (5)$$

where J_1 is the Bessel function, a is the radius of the output surface, and $\vec{\rho}_1=(x_i,y_i)$ for $i=1,2$.

This result shows that the output Stokes field wave front is not homogeneous, even for $F=1$. The coherence function is just the same as if the left-hand side of the amplifier were a classical incoherent radiation source.[8] The larger the F the smaller the ratio of the correlation length to the radius of the perpendicular cross section of the amplifier.

The notion of diffraction modes is connected with the correlation function G. If the field is purely coherent, i.e., emitted in one mode, the correlation function factorizes: $G(\vec{r}_1,\vec{r}_2,t)=E(\vec{r}_1,t)E^+(\vec{r}_2,t)$. If the field is emitted in several modes, the correlation function takes the form $G(\vec{r}_1,\vec{r}_2,t)=\sum_i E_i(\vec{r}_1,t)E_i^+(\vec{r}_2,t)$. Generalizing, by the diffraction mode we will mean an eigenfunction of $G(\vec{r}_1,\vec{r}_2,t)$ treated as an integral kernel.

In our case these eigenfunctions and their eigenvalues depend on F. If for some F the kernel has N nonzero eigenvalues we will say that emission takes place in N diffraction modes. As we will show the eigenvalues do tend to zero rapidly for $1\lesssim F\lesssim 10$, therefore the notion of the number of the diffraction modes has a rather precise meaning.

In Ref. 7 we have presented a method which reduces the problem of diagonalizing the kernel G to the diagonalization of finite (and small) matrices. As an example we give in Table I the eigenvalues of the kernel $2J_1[(F/2a)|\vec{r}-\vec{r}'|]/[(F/2a)|\vec{r}-\vec{r}'|]$ for some values of F. We see that for $F=1$ there is one leading eigenvalue, i.e., the emission takes place mostly in one mode. For larger F, however, the emission takes place in several modes.

The number of diffraction modes in which the emission takes place has a direct impact on the pulse energy statistics. The pulse energy distribution function is defined as

$$p(w)=\left\langle\delta\left[w-\int_0^T dt\int d_2r' E_S(\vec{r}',t)E_S^+(\vec{r}',t)\right]\right\rangle . \quad (6)$$

This is a direct generalization of the formula (7) of Raymer *et al.* We assume that the intensity is understood as the ratio of the total flux to the area (this is the origin of the extra integration over the area).

Inserting the expression for E_S [Eq. (2)] we find

$$p(w)=\left\langle\delta\left[w-\int H(\vec{r}_1,\vec{r}_2,T)\times Q^+(\vec{r}_1,0)Q(\vec{r}_2,0)d_3r_1d_3r_2\right]\right\rangle , \quad (7)$$

with $H=\int_0^T dt\int K(\vec{r}',\vec{r}_1)K(\vec{r}_1',\vec{r}_2)d_2r'$. This distribution function can be expressed in terms of the eigenvalues $\lambda_1,\lambda_2,\ldots$ of the kernel H. In the one-dimensional case it was possible to diagonalize this kernel numerically, but otherwise exactly. Here we will restrict ourselves to the high-gain limit only. In this limit the integration can be performed and we find that

$$H(\vec{r}_1,\vec{r}_2,T)=K^{(1)}(z_1,z_2,T)\frac{2J_1[(F/2a)|\vec{\rho}_1-\vec{\rho}_2|]}{(F/2a)|\vec{\rho}_1-\vec{\rho}_2|} , \quad (8)$$

where $K^{(1)}$ is the kernel from the one-dimensional theory (Ref. 1). It has been shown by Raymer, Rzażewski, and Mostowski[1] that $K^{(1)}$ has in the high-gain limit only one

TABLE I. Eigenvalues of the kernel $2J_1(F|\vec{\rho}-\vec{\rho}'|/2a)/(F|\vec{\rho}-\vec{\rho}'|/2a)$ for $F=1$ and $F=2.5$. Asterisks denote doubly degenerated eigenvalues. Eigenvalues larger than 10^{-5} are listed only.

λ	$F=1$	$F=2.5$
λ_1	1.9780	0.5003
λ_2^*	0.5065	0.4709
λ_3^*	5.9968×10^{-2}	0.3534
λ_4	2.1118×10^{-2}	0.2909
λ_5^*	5.8141×10^{-4}	0.1767
λ_6^*		8.454×10^{-2}
λ_7^*		5.649×10^{-2}
λ_8^*		1.299×10^{-2}
λ_9^*		1.2096×10^{-2}
λ_{10}		1.8229×10^{-3}

nonzero eigenvalue. Therefore the eigenvalues of H are determined by the eigenvalues of the diffraction kernel $2J_1[(F/2a)|\vec{\rho}_1-\vec{\rho}_2|]/(F/2a)|\vec{\rho}_1-\vec{\rho}_2|$.

The distribution $p(w)$ is given by

$$p(w)=\lim_{M\to\infty}\sum_{k=1}^{M}\frac{\lambda_K^{M-2}}{(\lambda_1-\lambda_K)(\lambda_2-\lambda_K)\cdots(\lambda_M-\lambda_K)}\times\exp\left(-\frac{w}{\lambda_K}\right). \quad (9)$$

Contrary to the one-dimensional case several eigenvalues give a comparable contribution to $p(w)$, especially for larger F. Plots of $p(w)$ for $F=1, 2.5, 8$ are given in Fig. 1. Observe that the larger the Fresnel number the narrower the distribution becomes. The distribution functions have value zero at $w=0$, the maxima occur for w significantly larger than zero. This feature of the distribution functions provides the fundamental difference between the present and the one-dimensional approach. One-dimensional theory predicts that the distribution function has a maximum essentially at $w=0$ in the high-gain limit (or at 0.05 of the mean value, at most, in the intermediate-gain case).

In their measurements Walmsley and Raymer (Ref. 3) found the tendency of the distribution function to lie below the theoretical (from one-dimensional theory) values for small values of the argument. A similar tendency was observed in measurements of the Essen group.[9] No convincing explanation was given, although Walmsley and Raymer suggested that the three-dimensional effects could play a role.

Although both experiments were performed for amplifiers with $F=1$, the present three-dimensional theory presents a natural and straightforward explanation of the observed shapes of the Stokes pulse energy distribution functions.

Our theory predicts also a new feature of the distribution function, namely, the narrowing of the distribution for larger Fresnel numbers. A related phenomenon in superfluorescence, the narrowing of the delay times distribution function with increasing Fresnel numbers has been measured by Vrehen and der Weduwe[10] and attributed by Drummond and Eberly,[11] Watson *et al.*,[12] Mostowski and Sobolewska[7,13] to the transverse effects in light propagation. Stokes pulse energy statistics measurements for various Fresnel numbers are called for.

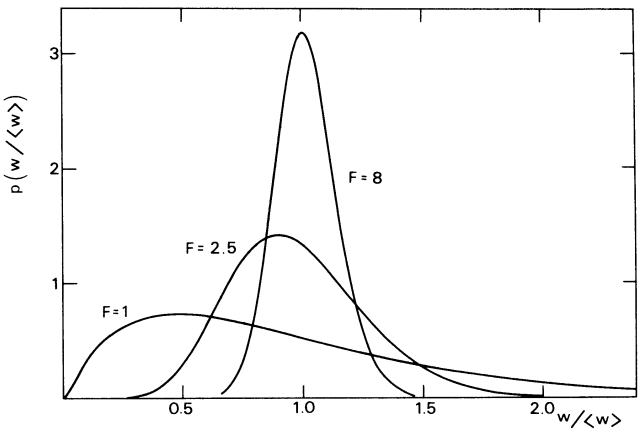

FIG. 1. Pulse energy distribution functions for $F=1, 2.5, 8$.

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