Spatial multimode theory of cooperative Raman scattering

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(Submitted 1 March 1988)
Zh. Eksp. Teor. Fiz. **94**, 19–26 (September 1988)

A generalized system of quantum equations that describes the space-time development of cooperative Raman scattering in the multimode case is obtained. It is shown that the process is spatially inhomogeneous, and allowance for propagation effects leads to the appearance of amplitude-phase modulation of the pulses of the scattered fields. The spatial and angular distributions of the intensities of the scattered fields are investigated numerically.

1. INTRODUCTION

The dynamics of the development of cooperative Raman scattering (CRS) of light is mainly described at the present time by two approaches: the semiclassical and the quantum. The main advantages of the first are the simplicity of the exposition and the lucid allowance for the spatial development of the CRS. However the semiclassical equations do not describe the generation of the pulses of the scattered fields and therefore require the introduction of external sources of spontaneous noise or the specification of nonzero initial conditions. In Refs. 1–3 equations describing CRS were obtained on the basis of this approach; it was shown that in the nonresonance case there exists in the initial stage of scattering a periodic solution with alternating pulses of Stokes and anti-Stokes radiation. In Ref. 4 resonance CRS was investigated with allowance for the effects of propagation for scattered waves without allowance for the anti-Stokes component.

The main advantage of the quantum theory is that it takes into account the spontaneous sources, and this makes it possible to describe consistently the process of occurrence of CRS in an initially uncorrelated system of atoms (or molecules). On the other hand, the quantum theory uses an expansion of the field with respect to plane waves, so that in this theory it is difficult to describe the spatial development of the CRS. In Refs. 5 and 6, which are based on a systematic quantum approach, a system of equations is obtained that describes the process of CRS with allowance for relaxation, stimulated, spontaneous, and parametric processes, but without allowance for propagation effects.

In the present paper, which is devoted to the kinetics of space-time development of CRS in a system of two-level emitters, the system of equations obtained in Ref. 5 is generalized to the case of transverse and longitudinal multimode scattering. It is shown that the generalized system describes the space-time development of the scattering. The use of this system to describe the process is not restricted to values of the Fresnel number near unity, and this makes it possible to describe CRS in media with any geometrical shape. By means of numerical solution of the system we investigate the spatial distribution of the intensities of the scattered waves at different times and the mode structure of the scattering in a medium with periodic structure. We show that in the quantum theory, as in the semiclassical theory,[4] allowance for the spatial effects leads to strong amplitude and phase modulation of the scattered-field pulses.

2. GENERALIZED SYSTEM OF QUANTUM EQUATIONS

We consider the process of Raman scattering of a given steplike exciting pumping field (frequency ω_p, wave vector $\mathbf{k}_p$) by a system of N two-level (transition frequency ω_0) emitters contained in a volume V. The initial state of the system is incoherent and unexcited. Scattering gives rise to fields at the combination (Raman) frequencies: Stokes field (frequency ω_s, wave vector $\mathbf{k}_s$) and anti-Stokes field (frequency ω_a, wave vector $\mathbf{k}_a$).

The Hamiltonian of the system has the form[5]

$$\hat{H} = \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}} a_{\mathbf{k}}^{+} a_{\mathbf{k}} + \hbar\omega_p b^{+} b + \sum_{j=1}^{N} \frac{\hbar\omega_0}{2}\sigma_3^{j} + \Big(\alpha_s \sum_{\mathbf{k}_s} g_{\mathbf{k}_s}\Big)[a^{+}_{\mathbf{k}_s} b R^{+}_{\mathbf{k}_p-\mathbf{k}_s} + \text{h.c.}] + \Big(\alpha_a \sum_{\mathbf{k}_a} g_{\mathbf{k}_a}\Big)[a_{\mathbf{k}_a} b^{+} R^{+}_{\mathbf{k}_a-\mathbf{k}_p} + \text{h.c.}], \quad (1)$$

where

$$\alpha_{s(a)} = \varkappa^{s(a)}\mathscr{E}_p, \quad \mathscr{E}_p = (2\pi\hbar\omega_p/V)^{1/2}, \quad \mathbf{g}_{\mathbf{k}} = (2\pi\hbar ck/V)^{1/2}\mathbf{e}_{\mathbf{k}},$$

$$\varkappa_{\alpha\beta}^{s(a)} = \frac{1}{\hbar}\sum_q \Big[\frac{(\mathbf{d}_{\beta q}\mathbf{e}_{s(a)})\mathbf{d}_{q\alpha}}{\omega_{\beta q}+\omega_p} + \frac{(\mathbf{d}_{\beta q}, \mathbf{e}_p)\mathbf{d}_{q\alpha}}{\omega_{\beta q}-\omega_{s(a)}}\Big]$$

are the matrix elements of the operator of the scattering tensor, $\mathbf{g}_{\mathbf{k}}$ are the constants of the coupling of the atoms to the scattering fields in the presence of pumping, $\mathbf{e}_{s(a)}$ and $\mathbf{e}_p$ are the unit polarization vectors of the photons of the scattered fields and the pumping field, $a^{+}_{\mathbf{k}_s}$ + $(a_{\mathbf{k}_s})$, $a^{+}_{\mathbf{k}_a}$ $(a_{\mathbf{k}_a})$, and b^{+} (b) are the operators of creation (and annihilation) of the Stokes and anti-Stokes fields and the pumping fields, respectively:

$$R_{\mathbf{k}}^{\pm} = \sum_{j=1}^{N} \sigma_{\pm}^{j} \exp(\pm i\mathbf{k}\mathbf{r}_j), \qquad \sigma_{\pm}^{j} = (\sigma_1^{j} \pm i\sigma_2^{j})/2,$$

where σ_1^j, σ_2^j, and σ_3^j are the Pauli spin operators of atom j.

The collective atomic operators $R_{\mathbf{k}}^{+}$ and $R_{\mathbf{k}}^{-}$ satisfy the commutation relations

$$[R_{\mathbf{k}}^{+}; R_{\mathbf{k}'}^{-}] = 2R_{3,\mathbf{k}-\mathbf{k}'}, \qquad [R_{\mathbf{k}}^{\pm}; R_{3,\mathbf{k}'}] = \mp R^{\mp}_{\mathbf{k}\pm\mathbf{k}'}, \quad (2)$$

where

$$R_{3,\mathbf{k}} = \sum_{j=1}^{N} \frac{1}{2}\sigma_3^{j} \exp(i\mathbf{k}\mathbf{r}_j).$$

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From the equation for the operators in the Heisenberg representation

$$i\hbar\frac{\partial\hat{f}}{\partial t}=[\hat{f},\hat{H}]$$

with the Hamiltonian (1) and the commutation relations (2) we obtain then the following system of equations for the intensities of the scattered fields:

$$\frac{dn_{\mathbf{k}_s',\mathbf{k}_s}}{dt}+\frac{n_{\mathbf{k}_s',\mathbf{k}_s}}{\tau_\mathbf{k}}$$

$$=i(\omega_{\mathbf{k}_s'}-\omega_{\mathbf{k}_s})n_{\mathbf{k}_s',\mathbf{k}_s}+F_{\mathbf{k}_s',\mathbf{k}_s}+F^+_{\mathbf{k}_s,\mathbf{k}_s'},$$

$$\frac{dn_{\mathbf{k}_a',\mathbf{k}_a}}{dt}+\frac{n_{\mathbf{k}_a',\mathbf{k}_a}}{\tau_\mathbf{k}}$$

$$=i(\omega_{\mathbf{k}_a'}-\omega_{\mathbf{k}_a})n_{\mathbf{k}_a',\mathbf{k}_a}+E_{\mathbf{k}_a',\mathbf{k}_a}+E^+_{\mathbf{k}_a,\mathbf{k}_a'};$$

$$\frac{dF_{\mathbf{k}_s,\mathbf{k}_s'}}{dt}+\frac{1}{2}\left(\frac{1}{\tau_\mathbf{k}}+\frac{1}{T_2}\right)F_{\mathbf{k}_s,\mathbf{k}_s'}$$

$$=i(\omega_p-\omega_0-\omega_{\mathbf{k}_s})F_{\mathbf{k}_s,\mathbf{k}_s'}$$

$$+\frac{1}{T_0^2}\Big[-\sum_{\mathbf{k}_s''}n_{\mathbf{k}_s'',\mathbf{k}_s}R_{3,\mathbf{k}_s'-\mathbf{k}_s''}+S_{\mathbf{k}_s',\mathbf{k}_s}+{}^1/_2S^0_{\mathbf{k}_s',\mathbf{k}_s}$$

$$-\sum_{\mathbf{k}_a}Q_{\mathbf{k}_s,\mathbf{k}_a}R_{3,\mathbf{k}_a+\mathbf{k}_s'-2\mathbf{k}_p}\Big], \qquad (3)$$

$$\frac{dE_{\mathbf{k}_a,\mathbf{k}_a'}}{dt}+\frac{1}{2}\left(\frac{1}{\tau_\mathbf{k}}+\frac{1}{T_2}\right)E_{\mathbf{k}_a,\mathbf{k}_a'}$$

$$=i(\omega_p+\omega_0-\omega_{\mathbf{k}_a})E_{\mathbf{k}_a,\mathbf{k}_a'}$$

$$+\frac{1}{T_0^2}\Big[\sum_{\mathbf{k}_a''}n_{\mathbf{k}_a'',\mathbf{k}_a}R_{3,\mathbf{k}_a'-\mathbf{k}_a''}+A_{\mathbf{k}_a',\mathbf{k}_a}+{}^1/_2A^0_{\mathbf{k}_a',\mathbf{k}_a}$$

$$+\sum_{\mathbf{k}_s}Q_{\mathbf{k}_s,\mathbf{k}_a}R_{3,\mathbf{k}_a+\mathbf{k}_s-2\mathbf{k}_p}\Big],$$

$$\frac{dS_{\mathbf{k}_s',\mathbf{k}_s}}{dt}+\frac{S_{\mathbf{k}_s',\mathbf{k}_s}}{T_2}$$

$$=-\sum_{\mathbf{k}_s''}[F_{\mathbf{k}_s'',\mathbf{k}_s'}R_{3,\mathbf{k}_s''-\mathbf{k}_s}+F^+_{\mathbf{k}_s'',\mathbf{k}_s}R_{3,\mathbf{k}_s'-\mathbf{k}_s''}]$$

$$+\sum_{\mathbf{k}_a}[E_{\mathbf{k}_a,2\mathbf{k}_p-\mathbf{k}_s}R_{3,\mathbf{k}_a+\mathbf{k}_s'-2\mathbf{k}_p}+E^+_{\mathbf{k}_a,2\mathbf{k}_p-\mathbf{k}_s'}R_{3,2\mathbf{k}_p-\mathbf{k}_s-\mathbf{k}_a}],$$

$$\frac{dA_{\mathbf{k}_a',\mathbf{k}_a}}{dt}+\frac{A_{\mathbf{k}_a',\mathbf{k}_a}}{T_2}$$

$$=\sum_{\mathbf{k}_a''}[E_{\mathbf{k}_a'',\mathbf{k}_a'}R_{3,\mathbf{k}_a''-\mathbf{k}_a}+E^+_{\mathbf{k}_a'',\mathbf{k}_a}R_{3,\mathbf{k}_a'-\mathbf{k}_a''}]$$

$$-\sum_{\mathbf{k}_s}[F_{\mathbf{k}_s,2\mathbf{k}_p-\mathbf{k}_a}R_{3,\mathbf{k}_a'+\mathbf{k}_s-2\mathbf{k}_p}+F^+_{\mathbf{k}_s,2\mathbf{k}_p-\mathbf{k}_a'}R_{3,2\mathbf{k}_p-\mathbf{k}_a-\mathbf{k}_s}],$$

$$\frac{dQ_{\mathbf{k}_s,\mathbf{k}_a}}{dt}+\frac{Q_{\mathbf{k}_s,\mathbf{k}_a}}{\tau_\mathbf{k}}$$

$$=i(2\omega_p-\omega_{\mathbf{k}_s}-\omega_{\mathbf{k}_a})Q_{\mathbf{k}_s,\mathbf{k}_a}-F_{\mathbf{k}_s,2\mathbf{k}_p-\mathbf{k}_a}-E_{\mathbf{k}_a,2\mathbf{k}_p-\mathbf{k}_s},$$

$$\frac{dR_{3,\mathbf{k}}}{dt}+\frac{N+R_{3,\mathbf{k}}}{T_1}$$

$$=\sum_{\mathbf{k}_s}(F_{\mathbf{k}_s,\mathbf{k}_s+\mathbf{k}}+F^+_{\mathbf{k}_s,\mathbf{k}_s-\mathbf{k}})-\sum_{\mathbf{k}_a}(E_{\mathbf{k}_a,\mathbf{k}_a+\mathbf{k}}+E^+_{\mathbf{k}_a,\mathbf{k}_a-\mathbf{k}}),$$

where $n_p=\langle b^+b\rangle$; $n_{\mathbf{k}',\mathbf{k}}=\langle a^+_{\mathbf{k}'}a_\mathbf{k}\rangle$, $n_p\gg n_{\mathbf{k}',\mathbf{k}}$ (we ignore the depletion of the pumping), $\tau_\mathbf{k}=(c/L(\mathbf{k})+c/L_0)^{-1}$ is the lifetime of a photon in the active region ($L(\mathbf{k})$ is the length of the sample in the direction of mode $\mathbf{k}$, L_0 is the photon mean free path), T_1 and T_2 are the times of longitudinal and transverse relaxation, respectively, and

$$\frac{1}{T_0^2}=2n_p\left|\frac{\alpha_{s(a)}g_\mathbf{k}}{\hbar}\right|^2$$

gives the characteristic time of interaction between the atoms and the field. The quantities $F_{\mathbf{k}_s,\mathbf{k}_s'}$ and $E_{\mathbf{k}_a,\mathbf{k}_a'}$ are defined as the correlation functions of the operators of the fields and the dipole moments of the atoms:

$$F_{\mathbf{k}_s,\mathbf{k}_s'}=i\hbar^{-1}(\alpha_s,g_{\mathbf{k}_s})\langle a_{\mathbf{k}_s}b^+R^-_{\mathbf{k}_p-\mathbf{k}_s'}\rangle,$$

$$E_{\mathbf{k}_a,\mathbf{k}_a'}=i\hbar^{-1}(\alpha_a,g_{\mathbf{k}_a})\langle a_{\mathbf{k}_a}b^+R^+_{\mathbf{k}_a'-\mathbf{k}_p}\rangle.$$

The correlation functions of the dipole moments, $S_{\mathbf{k}_s',\mathbf{k}_s}$ and $A_{\mathbf{k}_a',\mathbf{k}_a}$, describe the cooperative processes:

$$S_{\mathbf{k}_s',\mathbf{k}_s}=\frac{1}{2}\langle R^-_{\mathbf{k}_p-\mathbf{k}_s'}R^+_{\mathbf{k}_p-\mathbf{k}_s}\rangle,$$

$$A_{\mathbf{k}_a',\mathbf{k}_a}=\frac{1}{2}\langle R^+_{\mathbf{k}_a'-\mathbf{k}_p}R^-_{\mathbf{k}_a-\mathbf{k}_p}\rangle.$$

The terms

$$S^0_{\mathbf{k}_s',\mathbf{k}_s}=\frac{N}{2}\gamma(\mathbf{k}_s'-\mathbf{k}_s)-R_{3,\mathbf{k}_s'-\mathbf{k}_s},$$

$$A^0_{\mathbf{k}_a',\mathbf{k}_a}=\frac{N}{2}\gamma(\mathbf{k}_a'-\mathbf{k}_a)+R_{3,\mathbf{k}_a'-\mathbf{k}_a}$$

describe the spontaneous processes (in the case $\mathbf{k}'_{s(a)}=\mathbf{k}_{s(a)}$ we obtain $S^0=N/2-R_3$ and $A^0=N/2+R_3$, the populations of the lower and upper levels, where $R_3=\frac{1}{2}\sum_{j=1}^{N}\langle\sigma_3^j\rangle$ is the half-difference of the populations of the working levels of the atoms). The quantity

$$Q_{\mathbf{k}_s,\mathbf{k}_a}=\frac{1}{n_p}\langle a_{\mathbf{k}_s}a_{\mathbf{k}_a}b^+b^+\rangle$$

determines the correlation function of the Stokes and anti-Stokes fields and the pumping field. The terms of the type $\sum Q_{\mathbf{k}_s,\mathbf{k}_a}R_{3,\mathbf{k}-\mathbf{k}'}$ describe processes of four-wave parametric interaction, and the terms $\sum_{\mathbf{k}''}n_{\mathbf{k}'',\mathbf{k}}R_{3,\mathbf{k}'-\mathbf{k}''}$ describe the stimulated processes.

The intensities of the scattered fields are given by

$$I_{s(a)}=\sum_{\mathbf{k}_{s(a)}}\sum_{\mathbf{k}'_{s(a)}}\frac{2\pi\hbar c^2}{V}\frac{(\mathbf{e}_{\mathbf{k}_{s(a)}}\mathbf{e}_{\mathbf{k}'_{s(a)}})}{\omega_{\mathbf{k}_{s(a)}}\omega_{\mathbf{k}'_{s(a)}}}n_{\mathbf{k}_{s(a)},\mathbf{k}'_{s(a)}}$$

$$\times\exp\left[i\left(\mathbf{k}_{s(a)}-\mathbf{k}'_{s(a)}\right]\mathbf{r}_j\right].$$

The correlation coefficients $\gamma(\mathbf{k}'-\mathbf{k})$ give a measure of the coupling between the different modes of the scattered fields:

$$\gamma(\mathbf{k}'-\mathbf{k})=\frac{1}{N}\sum_{j=1}^{N}\exp[i(\mathbf{k}'-\mathbf{k})\mathbf{r}_j].$$

The calculation of these coefficients is analogous to the calculation made in Ref. 6. In the case of a medium with periodic structure we obtain for a sample of rectangular shape with measurements L_x, L_y, L_z

$$\gamma(\mathbf{k}'-\mathbf{k})=\frac{W}{V}\frac{\sin(L_x\Delta\mathbf{k}_x/2)}{\sin(d_x\Delta\mathbf{k}_x/2)}\frac{\sin(L_y\Delta k_y/2)}{\sin(d_y\Delta\mathbf{k}_y/2)}\frac{\sin(L_z\Delta\mathbf{k}_z/2)}{\sin(d_z\Delta\mathbf{k}_z/2)},$$

$$W=d_xd_yd_z;\ \Delta\mathbf{k}=\mathbf{k}'-\mathbf{k},\tag{4}$$

where d_x, d_y, d_z are the distances between neighboring emitters along the corresponding axes. For a sample of cylindrical shape, we initially expand the collective atomic operators along the z coordinate (the z axis is along the sample axis) with respect to plane waves and over the transverse section with respect to cylindrical functions (in contrast to a rectangular sample, for which plane-wave expansions are made with respect to all coordinates). We then obtain a system of equations of exactly the same form as (3) but with correlation coefficients

$$\gamma(\mathbf{k}'-\mathbf{k})=2\frac{d_z}{L_z}\frac{J_1[(\mathbf{k}'-\mathbf{k})_\perp\rho]}{(\mathbf{k}'-\mathbf{k})_\perp\rho}\frac{\sin(L_z(\mathbf{k}'-\mathbf{k})_z/2)}{\sin(d_z(\mathbf{k}'-\mathbf{k})_z/2)},\tag{5}$$

where ρ is the radius of the transverse section, J_1 is a Bessel function of the first kind of the first order, and $\perp$ denotes the projection of the vector onto the plane of the transverse section.

We introduce the collective operator of the half-difference of the populations of the working levels of the atoms:

$$R_{3,\mathbf{K}}=\frac{1}{2}\sum_{j=1}^{N}\langle\sigma_3{}^j\rangle\exp(i\mathbf{K}\mathbf{r}_j),\tag{6}$$

where $\mathbf{K}$ are the natural modes of the medium (for a medium with periodic structure $\mathbf{K}=\sum_i n_i\mathbf{K}_{0i}$, $n_i=0,1,2,\ldots$; $\mathbf{K}_{0i}$ are the fundamental vectors of the reciprocal lattice). From (6) we determine $\langle\sigma_3^j\rangle$, and, substituting in the expression for $R_{3,\mathbf{k}'-\mathbf{k}}$, we obtain

$$R_{3,\mathbf{k}'-\mathbf{k}}=\frac{1}{2}\sum_{j=1}^{N}\langle\sigma_3{}^j\rangle\exp[i(\mathbf{k}'-\mathbf{k})\mathbf{r}_j]=\sum_{\mathbf{K}}R_{3,\mathbf{K}}\gamma(\mathbf{k}'-\mathbf{k}-\mathbf{K}).$$

It can be seen from (4) and (5) that $\gamma(\mathbf{k}'-\mathbf{k}-\mathbf{K})$ is a sharply directed function having peaks at the values

$$\mathbf{k}'-\mathbf{k}=\mathbf{K}.\tag{7}$$

It follows that the main contribution to the scattering intensity will be made by modes for which the condition (7) is satisfied.

3. RESULTS OF A NUMERICAL EXPERIMENT

Even in the case of a finite number of modes the generalized system of equations (3) cannot be solved analytically. In principle, it can be solved by numerical methods for any number of modes and for a medium of any geometrical shape. We present here the results of numerical solution for the most important case, that of a cylindrical sample elongated along its axis (z axis). In this case, as can be seen from the expression for the correlation coefficients (5), the main contribution is made by the first few modes situated near the fundamental mode (which we take to be the mode $\mathbf{k}^0_{s(a)}$ with $(\mathbf{k}^0_{s(a)})_\perp=0$, $(\mathbf{k}^0_{s(a)})_z=\omega_{s(a)}/c$). This assertion is confirmed by the numerical experiment. Then in the system (3) $R_{3,\mathbf{k}'-\mathbf{k}}$ and $R_{3,\mathbf{k}_s+\mathbf{k}_a-2\mathbf{k}_p}$ can be represented in the form

$$R_{3,\,\mathbf{k}'-\mathbf{k}}=N_\perp\gamma(\mathbf{k}'-\mathbf{k})_\perp R_{3,\,(\mathbf{k}'-\mathbf{k})z},$$
$$R_{3,\,\mathbf{k}_s+\mathbf{k}_a-2\mathbf{k}_p}=N_\perp\gamma(\mathbf{k}_s+\mathbf{k}_a-2\mathbf{k}_p)R_{3,\,(\mathbf{k}_s+\mathbf{k}_a-2\mathbf{k}_p)z},\tag{8}$$

where

$$R_{3,\mathbf{k}_z}=\frac{1}{2}\sum_{j=1}^{N_z}\langle\sigma_3{}^j\rangle\exp(i\mathbf{k}_zz_j),\qquad\gamma(\mathbf{k})_\perp=\frac{1}{N_\perp}\sum_{j=1}^{N_\perp}\exp(i\mathbf{k}_\perp\mathbf{r}_{\perp j}),$$

and $N_\perp$ and N_z are the numbers of atoms in the transverse section and along the axis of the sample, respectively.

Figure 1 gives the dependence of the normalized half-difference of the populations, R_3/N, and $V\omega_s^2I_s/2\pi\hbar c^2$, $V\omega_a^2I_a/2\pi\hbar c^2$ on the normalized time in the center of a cylindrical sample in the case of longitudinal pumping. Allowance was made for one transverse mode and five longitudinal modes $((\mathbf{k}_{s(a)})_z=\mathbf{k}^0_{s(a)}$, $\mathbf{k}^0_{s(a)}\pm\frac{\pi}{L_z}$,

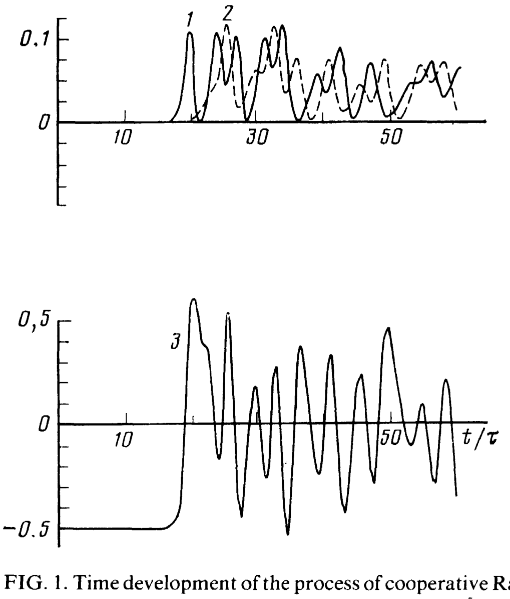

FIG. 1. Time development of the process of cooperative Raman scattering at the center of a cylindrical sample ($\lambda_s=6.8\cdot10^{-5}$ cm, $\lambda_p=5.3\cdot10^{-5}$ cm, $\lambda_a=4.34\cdot10^{-5}$ cm, $L_z=1$ cm, $\rho=2.5\cdot10^{-2}$ cm, $\tau\equiv\tau_k=3\cdot10^{-11}$ sec, $\tau/\tau_c=0.75$, where τ_c is the time of the cooperative processes; $N=10^{16}$, $\tau/T_2=0.03$): 1) $V\omega_s^2I_s/2\pi\hbar c^2$, 2) $V\omega_a^2I_a/2\pi\hbar c^2$, 3) R_3/N.

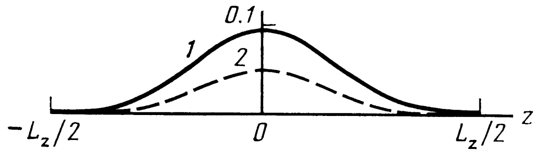

FIG. 2. Spatial distribution of the intensities of the scattered fields ($t/\tau = 32.5$): 1) $V\omega_s^2 I_s/2\pi\hbar c^2$, 2) $V\omega_a^2 I_a/2\pi\hbar c^2$.

$\mathbf{k}^0_{s(a)} \pm 2\pi/L_z$, where π/L_z is the magnitude of the fundamental translation vector of the reciprocal lattice with respect to the coordinate z) for each of the two scattering components.

It is well known that even in the single-mode case the CRS process has an oscillatory nature[5] due to the presence of two scattered components, in contrast to superradiance. These oscillations strictly follow the oscillations of the population difference, and in the case $T = T = \infty$ the process becomes purely periodic. It can be seen from Fig. 1 that in the multimode case of the quantum theory, as in the semiclassical theory, the allowance for propagation effects has the consequence that in addition to these oscillations there is phase and amplitude modulation of the pulses of the scattered fields. In contrast to the semiclassical theory, the modulations are here deeper, but with increasing number of modes they are partly smoothed. Compared with the single-mode case, the field amplitudes are reduced.

Figure 2 shows the spatial distribution of the scattered-field intensities at a certain time. It follows from the figure that in an extended medium the scattering process is spatially inhomogeneous. In our symmetric problem the field intensities have maxima in the center of the sample, since we take into account both the forward fields, traveling from the left to the right, as well as the reverse waves of both scattering components.

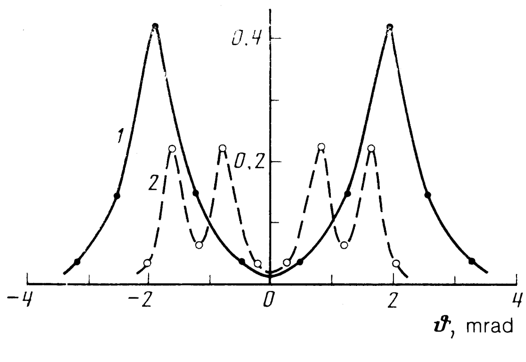

FIG. 3. Angular distribution of the number of photons of the scattered fields ($t/\tau = 47$, $\sin \vartheta_{s(a)} \equiv |(\mathbf{k}_{s(a)}|_\perp/|\mathbf{k}_{s(a)}|)$: 1) $n_{\mathbf{k}_s\mathbf{k}_s}/N$, 2) $n_{\mathbf{k}_a\mathbf{k}_a}/N$.

By means of the generalized system (3) one can also consider the angular dependence of the scattering. This is more conveniently done in the case when there is one longitudinal mode and several transverse modes for each of the scattered components. Then instead of (8) we obtain

$$R_{3,\,\mathbf{k}'-\mathbf{k}} = N_z \gamma(\mathbf{k}'-\mathbf{k})_z R_{3,\,(\mathbf{k}'-\mathbf{k})_\perp},$$
$$R_{3,\,\mathbf{k}_s+\mathbf{k}_a-2\mathbf{k}_p} = N_z \gamma(\mathbf{k}_s+\mathbf{k}_a-2\mathbf{k}_p)_z R_{3,\,(\mathbf{k}_s+\mathbf{k}_a-2\mathbf{k}_p)_\perp};$$

where

$$R_{3,\mathbf{k}_\perp} \equiv \frac{1}{2}\sum_{j=1}^{N_\perp} \langle \sigma_3^j \rangle \exp(i\mathbf{k}_\perp \mathbf{r}_{j\perp}), \qquad \gamma(\mathbf{k})_z \equiv \frac{1}{N_z}\sum_{j=1}^{N_z} \exp(i\mathbf{k}_z z_j).$$

Figure 3 gives the dependence of the normalized number of photons of the scattered fields, $n_{\mathbf{k},\mathbf{k}}/N$, on the scattering angle at a certain time. Investigations showed that the scattering is strongly directed and basically along a layer formed by two conical surfaces having axes that coincide with the axis of the cylindrical sample but different angles. The values of these angles and the thicknesses of the layer are very close to the values obtained earlier in Ref. 6. In contrast to Ref. 6, the optimal scattering angle varies as a function of the time in a narrow interval between two limiting values of the scattering angle, which are equal to the angles of the cones. In the direction of the sample axis there is no scattering.

Thus, the generalized system of equations (3) makes it possible to describe at the quantum level the dynamical development of the process of cooperative Raman scattering of light with allowance for the spontaneous, stimulated, parametric, and relaxation processes and propagation processes. By means of this system one can investigate the spatial distribution, angular distribution, and mode composition of the scattering.

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Translated by Julian B. Barbour