Cooperative and stimulated Raman scattering of light

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(Submitted 20 January 1987)
Zh. Eksp. Teor. Fiz. **93**, 838–844 (September 1987)

A system of quantum equations for the description of the dynamics of cooperative Raman scattering is derived allowing for relaxation, induced, and parametric processes. An equation with a periodic solution is obtained for the case of pure superscattering using a quasistationary approximation for the half-difference between the populations. The condition is found for ignoring the contributions of induced and parametric processes. A study is made of the influence of a four-wave parametric interaction on the cooperative Raman scattering. The results are given of a general numerical solution of the proposed system of equations with different values of characteristic dimensionless parameters.

1. INTRODUCTION

Several theoretical and experimental investigations have been made recently of the cooperative Raman scattering of light.[1,3] An experimental study of the cooperative Raman scattering of light by H_2 molecules was reported in Ref. 3, where the pulsating nature of the scattering and the existence of a delay of the first peak of the Stokes component relative to the leading edge of a pump pulse were established. A semiclassical approach was used in Refs. 1 and 2 to obtain equations describing such cooperative Raman scattering of light without allowance for relaxation processes. A periodic solution of these equations with alternate Stokes and anti-Stokes radiation pulses was obtained. This theoretical analysis ignored the parametric interaction of Stokes and anti–Stokes radiations.

The cooperative Raman scattering of light, like the cooperative spontaneous emission (Dicke superradiance), are examples of collective behavior of a system of spatially separated atoms or molecules, when interatomic correlations induced spontaneously during the process of the scattering in an initially noncoherent system create coherent radiation at characteristic frequencies (Stokes and anti-Stokes frequencies in the Raman scattering case).

A natural question is this: how is the cooperative Raman scattering related to the familiar spontaneous Raman scattering (SPRS) and stimulated Raman scattering (STRS)? An important characteristic of the cooperative effect is a delay of a radiation pulse t_0, scattered by correlated atoms, relative to the leading edge of an exciting pump pulse. The delay time cannot be determined in a consistent manner using a semiclassical theory of the type employed in Refs. 1 and 2.

We shall employ a quantum approach to propose and analyze a system of equations describing the dynamics of the cooperative Raman scattering allowing for relaxation, induced, and parametric processes. This system of equations makes it possible to follow a continuous transition from SPRS and STRS to the cooperative effect. When the relaxation times are much longer than the characteristic times of the collective interaction of atoms with an electromagnetic field, this system of equations yields the equation of Ref. 1 with a characteristic periodic solution. We shall find the conditions under which we can ignore the contributions of induced and parametric processes. We shall show that in the case of finite relaxation times the inclusion of these processes reduces the oscillation period and the delay time and makes the scattering dynamics a damped process. We shall consider qualitatively the influence of the conditions for spatial phase matching on the cooperative Raman scattering.

2. PRINCIPAL EQUATIONS

We shall consider the cooperative Raman scattering for a system of N two-level atoms or molecules subjected to a pump step-like pulse, so that the leading edge of the pump pulse is shorter than all the characteristic times of the problem. We shall limit our analysis to the single-mode case when the Fresnel number is $F\sim 1$ (only this case is mainly encountered in experiments). The initial state of the system is noncoherent and the Raman scattering creates fields the Stokes (frequency ω_s, wave vector $\mathbf{k}_s$) and anti-Stokes (frequency ω_a, wave vector $\mathbf{k}_a$) Raman components. It is assumed that initially the atoms are unexcited and occupy a volume v of cylindrical shape elongated along the z axis (cross-section area A, length L).

The Hamiltonian of the system is similar to the Dicke Hamiltonian which is used widely in the theory of superradiance (see, for example, Ref. 4) and it can be obtained from the latter Hamiltonian by replacing the matrix element of the dipole moment operator of an atom with $\varkappa_{\alpha\beta}\mathscr{E}_{p\beta}$, where $\varkappa_{\alpha\beta}$ are the matrix elements of the scattering tensor:

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

whereas

$$\mathscr{E}_p=E_p\exp(-i\omega_p t+i\mathbf{k}_p\mathbf{r})+\text{c.c.}$$

is the pump field which can be regarded as a c number. The Hamiltonian is

$$H=\hbar\omega_s a_s^+a_s+\hbar\omega_a a_a^+a_0+\sum_{j=1}^{N}\hbar\omega_0\sigma_{3j}+\sum_{j=1}^{N}[g_s^*e^{-i\omega_p t}a_s^+\sigma_j^+e^{i(\mathbf{k}_p-\mathbf{k}_s)\mathbf{r}_j}+\text{H.c.}]+\sum_{j=1}^{N}(g_a e^{i\omega_p t}a_a\sigma_j^+e^{-i(\mathbf{k}_p-\mathbf{k}_a)\mathbf{r}_j}+\text{H.c.}] \quad (1)$$

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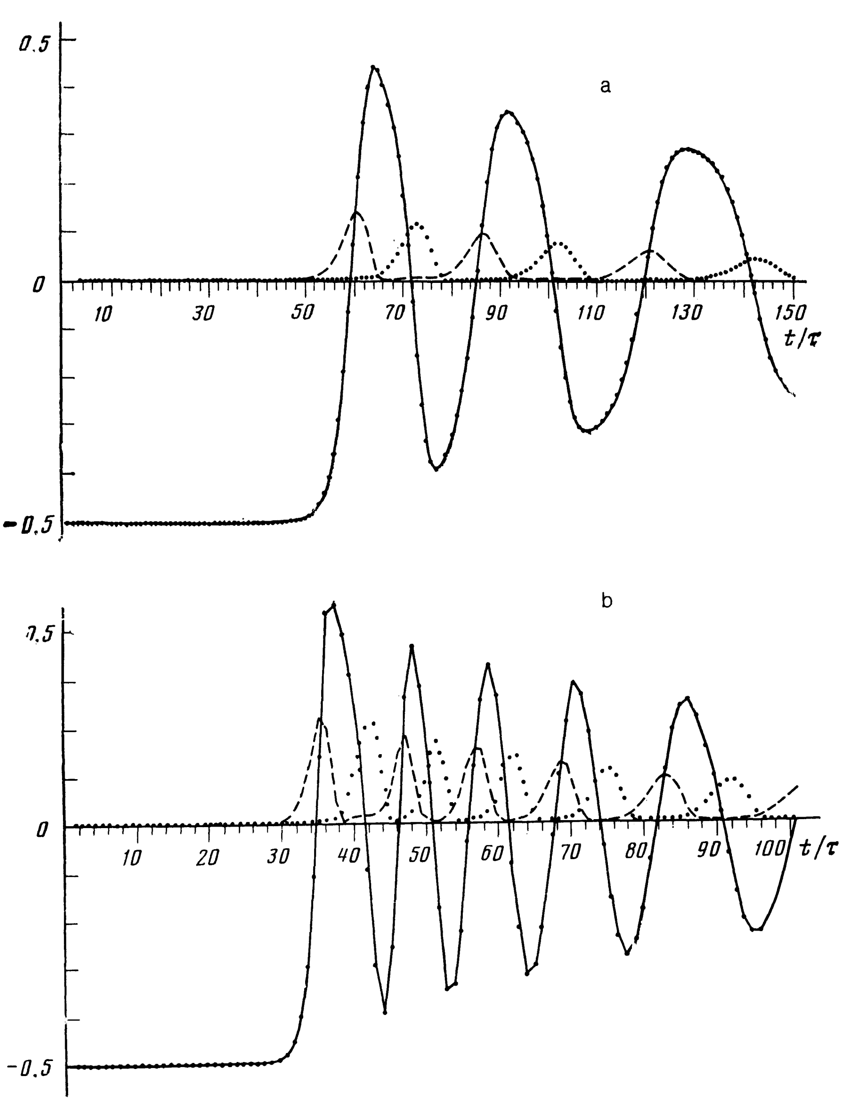

FIG. 1. Normalized intensities of the Stokes and anti-Stokes radiations and normalized values of the half-difference between the populations: a) $\tau/T_2 = 3\times10^{-2}$, $\tau/\tau_c = 0.375$, $\zeta = 0.1$, $N = 10^{12}$; b) $\tau/T_2 = 3\times10^{-2}$, $\tau/\tau_c = 0.75$, $\zeta = 0.05$, $N = 10^{12}$. The continuous curve represents the time dependence of R/N whereas the dashed and dotted curves represent the time dependences of n_s/N and n_a/N.

Here, a_s (a^s_+) and a_a (a_a^+) are the annihilation (creation) operators of Stokes and anti-Stokes photons;

$$g_s=-i(2\pi\hbar\omega_s/v)^{1/2}(\mathbf{e}_{s\alpha}\varkappa_{\alpha\beta}E_{p\beta})$$

(and a similar definition applies to g_a) are the constants representing the coupling of the atoms to the fields at the Stokes and anti-Stokes frequencies in the presence of a pump, $\mathbf{e}_s$ (and similarly $\mathbf{e}_a$) are the unit vectors of the polarization of the radiation; ω_0 is the transition frequency. Repetition of the indices α and β implies summation.

The matrices σ_j^- and $\sigma_j^+ = (\sigma_j^-)^+$ are the annihilation and creation operators of an excitation of a jth atom located at a point $\mathbf{r}_j$. They are related simply[4] to the Pauli matrices:

$$\sigma^\pm=(\sigma_1\pm i\sigma_2)/2.$$

Using the Hamiltonian of Eq. (1) and allowing for the interaction with the heat bath, we obtain a system of equations similar to the corresponding system of equations in the case of superradiance:

$$\frac{dn_s}{dt}+\frac{n_s}{\tau}=F_s, \qquad \frac{dn_a}{dt}+\frac{n_a}{\tau}=F_a,$$
$$\frac{dF_s}{dt}+\frac{1}{2}\left(\frac{1}{\tau}+\frac{1}{T_2}\right)F_s=\frac{1}{T_0^2}[-2n_sR+S_s+S_{s0}-\zeta QR],$$
$$\frac{dF_a}{dt}+\frac{1}{2}\left(\frac{1}{\tau}+\frac{1}{T_2}\right)F_a=\frac{1}{T_0^2}[2n_aR+S_a+S_{a0}+\zeta QR],$$
$$\frac{dS_s}{dt}+\frac{S_s}{T_2}=2R(\zeta F_a-F_s),$$
$$\frac{dS_a}{dt}+\frac{S_a}{T_2}=2R(-\zeta F_s+F_a), \tag{2}$$

In these equations $n_s = \langle a_s^+ a_s\rangle$ represents the average number of the Stokes photons and n_a is the average number of the anti-Stokes photons. The constant $\tau = (c/L + c/L_0)^{-1}$ is the photon lifetime in the active region (L_0 is the absorption length). The quantities F_s and F_a are defined as the correlation functions of the fields and dipole moments at the Stokes and anti-Stokes frequencies:

$$\frac{dQ}{dt}+\frac{Q}{\tau}=-F_s-F_a,$$
$$\frac{dR}{dt}+\frac{N+R}{T_1}=F_s-F_a.$$

$$F_s=i\hbar^{-1}\left[\sum_{j=1}^{N}g_s\langle\sigma_j^-a_s\rangle e^{i\omega_pt}e^{-i(\mathbf{k}_p-\mathbf{k}_s)\mathbf{r}_j}-\text{c.c.}\right],$$

$$F_a=i\hbar^{-1}\left[\sum_{j=1}^{N}g_a\langle\sigma_j^+a_a\rangle e^{i\omega_pt}e^{-i(\mathbf{k}_p-\mathbf{k}_a)\mathbf{r}_j}-\text{c.c.}\right].$$

The constants T_1 and T_2 are the longitudinal and transverse relaxation times,

$$R=\sum_{j=1}^{N}\langle\sigma_{3j}\rangle$$

is the half-difference between the populations of the active levels of the atoms. The quantity $T_0^{-2}=2|g_a|^2/\hbar^2$ represents the characteristic time of the interaction between the atoms and the field. Here, S_s and S_a are the correlation functions of the dipole moments

$$S_s=\sum_{j=1}^{N}\sum_{i\neq j}\langle\sigma_i^-\sigma_j^+\rangle e^{i(\mathbf{k}_p-\mathbf{k}_s)\mathbf{r}_{ji}},$$

$$S_a=\sum_{j=1}^{N}\sum_{i\neq j}\langle\sigma_i^+\sigma_j^-\rangle e^{i(\mathbf{k}_p-\mathbf{k}_a)\mathbf{r}_{ji}},\qquad \mathbf{r}_{ji}=\mathbf{r}_j-\mathbf{r}_i,$$

whereas $S_{s0}=(N/2)-R$ and $S_{a0}=(N/2)+R$ are the populations of the upper and lower levels governing the intensity of the spontaneous scattering. The quantity $Q=\langle a_a a_s\rangle+\text{c.c.}$ determines the correlation functions of the Stokes and anti-Stokes photons. Finally, ζ is the correlation coefficient for all the atoms in the sample:

$$\zeta=\frac{1}{N}\sum_{j=1}^{N}\exp[i(\mathbf{k}_s+\mathbf{k}_a-2\mathbf{k}_p)\mathbf{r}_j].\tag{3}$$

This coefficient gives a measure of the coupling between the Stokes and anti-Stokes fields, and it can be regarded as real if we assume that the atoms occupy a volume with coordinates z satisfying the inequality $-L/2\leqslant z\leqslant L/2$.

Equation (2) is derived on the assumption that

$$\sum_{j=1}^{N}\langle\sigma_{3j}\rangle e^{i\mathbf{k}\mathbf{r}_j}\approx\langle\sigma_{3j}\rangle\sum_{j=1}^{N}e^{i\mathbf{k}\mathbf{r}_j}\approx R\zeta(\mathbf{k}).$$

Physically this assumption means that the field amplitude varies little within the dimensions of the sample.

The parameter ζ is unity if the phase-matching condition is satisfied for the parametric process $\mathbf{k}_s+\mathbf{k}_a-2\mathbf{k}_p=0$; otherwise, it is less than unity. We can calculate this coefficient in the general case by changing in Eq. (3) from summation over all the atoms in the cylindrical sample to integration over the whole volume of the sample, which gives[5]

$$\zeta=4\frac{\sin[L(\mathbf{k}_s+\mathbf{k}_a-2\mathbf{k}_p)_\parallel/2]}{L(\mathbf{k}_s+\mathbf{k}_a-2\mathbf{k}_p)_\parallel}\,\frac{J_1[\rho(\mathbf{k}_s+\mathbf{k}_a-2\mathbf{k}_p)_\perp]}{\rho(\mathbf{k}_s+\mathbf{k}_a-2\mathbf{k}_p)_\perp},\tag{4}$$

where J_1 is a Bessel function of the first kind and of the first order, ρ is the radius of the transverse cross section of the sample, and $\parallel$ and $\perp$ denote the longitudinal and transverse components of a vector.

3. ANALYSIS AND SOLUTION OF DERIVED EQUATIONS

We shall now consider in greater detail the right-hand sides of the third and fourth equations in the system (2). The variable F_s (F_a) is the rate of transfer of energy stored in the atomic system to a Stokes (anti-Stokes) mode of the scattered field, which takes place via the induced processes n_sR (n_aR), spontaneous processes S_{s0} (S_{a0}), cooperative processes S_s (S_a), and QR processes involving a four-wave parametric interaction between the scattered fields. We shall now compare the contribution of the induced, cooperative, and parametric processes to the scattering intensity. The first, fifth, and seventh equations of the system (2) yield

$$\begin{aligned}-2n_sR+S_s-\xi QR=\int_0^t dt'\{&R(t)e^{-(t-t')/\tau}\\&\times[-2F_s(t')-\zeta(F_s(t')+F_a(t'))]\\&+2R(t')e^{-(t-t')/T_2}[\zeta F_a(t')-F_s(t')]\}.\end{aligned}\tag{5}$$

Hence it follows that in systems with $\tau\ll T_2$ the main contribution to the scattered light intensity comes from the cooperative spontaneous processes S_s (S_a), whereas in the $\tau\gg T_2$ case it is the induced and parametric processes that predominate (when considered on the order-of-magnitude scale, they differ only by the presence of the parameter ζ in the latter case).

We shall consider the case of superscattering, when $\tau\ll\tau_c\ll T_2,T_1$, where τ_c is the characteristic time of the collective processes. In this case we can ignore the relaxation processes and then the fifth, sixth, and eighth equations of the system (2) give the law of conservation, similar to the law of conservation of the length of the Bloch vector in the case of superradiance:

$$S_s+S_a+(1+\zeta)R^2=\text{const}\equiv c^2,\tag{6}$$

where c^2 is found from the initial conditions [when the system of atoms is initially in an unexcited noncoherent state, we find that $S_s(0)=S_a(0)=0$, $R(0)=-N/2$, and c^2 $(1+\zeta)N^2/4$].

Since $\tau\ll T_2$, it therefore follows (as shown above) that the cooperative processes predominate, so that the contributions of the induced and parametric processes can be ignored. Then, in the quasistationary approximation, ignoring the first terms in the first four equations of the system (2), and using Eq. (6), we obtain the following equation

$$\frac{d^2x}{dt'^2}+\frac{4}{N}\frac{dx}{dt'}=-2x[1-x^2+4/N],\tag{7}$$

where $x=2R/N$ and $t'=t/\tau_c$.

This is the equation for oscillations of a nonlinear oscillator with the quasielastic coefficient dependent on the oscillation amplitude:

$$\alpha(x)=2[1-x^2+4/N].$$

In the zeroth approximation with respect to the parameter $4/N$, Eq. (7) reduces to the familiar equation of Ref. 1 with the periodic solution, but it is of more general form [in contrast to Eq. (7), it was necessary to assume in Ref. 1 that not all the atoms were initially in the ground state]. In this approximation we find from Eq. (7) that

$$\frac{dx}{dt'}=\pm\frac{1}{a}[(1-x^2)(1-a^2x^2)]^{1/2},\tag{8}$$

where $a = (1 + 8/N)^{-1/2}$

The solution of Eq. (8) is in the form of elliptic sines

$$x(\vartheta) = x(0)\,\mathrm{sn}\,(\vartheta - \vartheta_0),$$

where $x(0) = 1$ if the atoms are in the ground state and $x(0) = -1$ if they are in an excited state: $\vartheta = (1/2)\ln 2N$ is the integration constant, found from the initial conditions.

The delay time t_0 is found in terms of the constant ϑ_0:

$$t_0 = a\tau_c\vartheta_0 = \frac{\tau_c}{2}\left[1 + \frac{8}{N}\right]^{-1/2}\ln 2N,$$

whereas the oscillation period is deduced in terms of elliptic Jacobi functions as follows:

$$T = 4a\tau_c K(a) \approx 4t_0.$$

The system of equations (2) was solved for the general case by numerical methods assuming various values of the dimensionless parameters τ/τ_c, τ/T_2, and ζ. In the case when $\tau \ll \tau_c$ and $T_1 = T_2 = \infty$, a purely periodic solution obtained and corresponded Eq. (8). The scattering process begins with an increase in the Stokes component and then the anti-Stokes component appears because of a four-wave parametric interaction. During the first half-period the Stokes component exceeds the intensity of the anti-Stokes component but beyond the maximum the system of atoms becomes inverted. During the second half-period the Stokes and anti-Stokes components, are interchanged and the system assumes a final state which is identical with the initial one.

If relaxation is allowed for, then—as expected—the periodic nature of the process disappears and for $T_2 \sim t_0$ a considerable reduction in the intensity of the scattered fields takes place after two or three peaks. The change in the time T_2 does not affect greatly the delay time t_0.

If $\tau_c \gtrsim \tau$, the role of the induced and parametric processes increases greatly (photons do not have sufficient time to leave the active volume before the interatomic correlations are induced) and an allowance for these processes reduces the oscillation period. The delay time t_0 is directly proportional to the characteristic constant of the collective processes τ_c ($t_0 \sim \tau_c$,) as in the superradiance case. The oscillation period depends weakly on the parameter ζ (when ζ is increased, the period decreases). If $\zeta = 1$, which corresponds to complete spatial phase matching of the modes characterized by $\mathbf{k}_s + \mathbf{k}_a - 2\mathbf{k}_p = 0$, the oscillations of the intensities of the scattered fields disappear. The numerical solution yields one weakly separated and closely spaced Stokes and anti-Stokes peak each, and these relax slowly to zero with time. This means that in the case of a strong coupling between the scattered fields there is no cooperative Raman scattering.

We shall conclude by noting that, for the sake of simplicity, we have considered only the single-mode case and have ignored the process of pump depletion, but the system of equations (3) is readily generalized to the case of multimode radiation and pumping of any form.

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Translated by A. Tybulewicz