Nonstationary cooperative phenomena in resonance fluorescence

S. G. Rautian and B. M. Chernobrod

Institute of Automation and Electrometry, Siberian Branch of the Academy of Sciences of the USSR, Novosibirsk

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It is shown that the collective interatomic interaction that occurs in resonance fluorescence can lead to nonstationary phenomena similar to those observed in cooperative emission and scattering. An analogy is established between the cooperative processes in resonance fluorescence and Raman light scattering. Analytic relations describing the linear phase of the process are obtained.

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1. INTRODUCTION

The attention of a number of authors has been drawn in recent years to the investigation of the effect of collective interatomic interaction on resonance fluorescence.[1-4] It has been established that, in the case in which the interaction of the atoms with the external monochromatic field is much stronger than the interatomic interaction, the scattering spectrum consists, as in the case of an isolated atom, of three components whose spacing is equal to the magnitude of the field splitting

$$\Omega_g=(4|G_{mn}|^2+\Omega^2)^{1/2},\quad G_{mn}=d_{mn}E/2\hbar,\quad \Omega=\omega-\omega_{mn}$$

(see Fig. 1). In this case the cooperative effects manifest themselves under certain conditions in both the fluorescence intensity[1-3] and the splitting-component widths.[4] The intensity-correlation function exhibits, besides the beats at the frequency Ω_g, which occur in the case of an isolated atom, beats at the doubled frequency $2\Omega_g$.

In the above-cited papers the stationary fluorescence regime is investigated. The purpose of the present paper is to show that the collective interaction leads to nonstationary phenomena similar to those experimentally observed in cooperative emission[5] and cooperative Raman scattering.[6]

Let a light pulse having the form of a step function of the time propagate in a medium composed of two-level atoms. Under the action of an external field the atomic levels split up (see Fig. 1), and there appear in the fluorescence spectrum in first-order perturbation theory in terms of the spontaneous field three lines with frequencies ω, $\omega_3=\omega-2\Omega_g$, and $\omega_p=\omega+\Omega_g$. If all the atoms are initially in the ground state, then a strong field will "mix" them into the energy quasilevels in such a way that the populations of the quasilevels will be equal to (see Ref. 7)

$$\rho_{nn}=n_0\left(\frac{|\Omega|+\Omega_g}{2\Omega_g}\right)^2,\quad \rho_{n'n'}=n_0\left(\frac{|\Omega|-\Omega_g}{2\Omega_g}\right)^2,\quad \rho_{mm}=\rho_{m'm'}=n_0\left|\frac{G_{mn}}{\Omega_g}\right|^2,\tag{1}$$

where n_0 is the total-number density of the atoms. If $|G_{mn}/\Omega_g|\ll 1$, then the population of the n-th state is much higher than that of the m-th state.

The emission of the line with frequency ω_3 can be interpreted as super-Raman scattering, in which two quanta of the strong field of frequency ω are absorbed and a quantum of frequency ω_3 is emitted, the atom going over in the process from the n-th state into the m-th state.[8] If the characteristic time of the exponential intensification of the field at the frequency ω_3 is shorter than the characteristic time T_2 of disturbance of the phase of the atomic polarization, then there will occur an avalance-type transition from the n-th state into the m-th state with the emission of a short (shorter than T_2) pulse at the frequency ω_3. As a result, an inversion population will arise between the m-th and n-th states, and this in turn will induce an avalanche-type emission of the ω_p resonance line, in much the same way as the cooperative emission in a medium of inverted two-level atoms is induced. The atoms will go into the initial state, so that the process described can occur repeatedly.

2. DERIVATION OF THE BASIC EQUATIONS

We shall describe the fluorescence process within the framework of the semiclassical approach, and perform the subsequent analysis in the approximation of a prescribed exciting field having the form of a step function of the time. The derivation of the basic equations describing cooperative resonance fluorescence is similar to the derivation, contained in our previous paper,[9] of the equations for cooperative resonance Raman scattering.

Let us prescribe the fluorescence field in the form of two quasi-harmonics with carrier frequencies corresponding to the transitions between the energy quasilevels arising in the interaction with the exciting field (see Fig. 1):

$$\begin{gathered}E_f=E_3e^{-i\omega_3t}+E_pe^{-i\omega_pt}+\text{c.c.},\\ E_3={}^1/_2[\mathscr{E}_3^+(z,t)e^{i\mathbf{k}_3\mathbf{r}}+\mathscr{E}_3^-(z,t)e^{-i\mathbf{k}_3\mathbf{r}}],\\ E_p={}^1/_2[\mathscr{E}_p^+(z,t)e^{i\mathbf{k}_p\mathbf{r}}+\mathscr{E}_p^-(z,t)e^{-i\mathbf{k}_p\mathbf{r}}].\end{gathered}\tag{2}$$

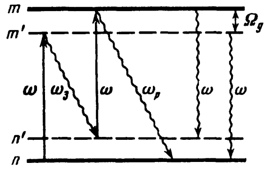

FIG. 1.

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Let us represent the amplitudes of the states in the form

$$a_n=b_n(z,t)\exp[i_2(|\Omega|-\Omega_g)t/2]+b_{n'}(z,t)\exp[i(|\Omega|+\Omega_g)t/2],$$
$$a_m=b_m(z,t)\exp[-i\omega t+i(|\Omega|+\Omega_g)t/2)+b_{m'}(z,t)\exp[-i\omega t+i(|\Omega|-\Omega_g)t/2]. \tag{3}$$

Let us assume that the characteristic scattering time τ_0 is much longer than the oscillation period of the amplitudes of the states, i.e., that $\tau_0\Omega_g \gg 1$. Then the amplitudes $b_{n,n'}$, $b_{m,m'}$ can be assumed to be slowly-varying functions of the time as compared to the exponential functions $\exp[i(\Omega \pm \Omega_g)t]$. In this approximation the equations for the amplitudes $a_{n,m}$ of the states can be uncoupled, and we can go over to the equations for the amplitudes $b_{n,n'}$, $b_{m,m'}$ and, moreover, express the amplitudes, $b_{n'}$ and $b_{m'}$, corresponding to the energy quasilevels lying at distances equal to Ω from the real levels when $|G_{mn}/\Omega| \ll 1$ (see Fig. 1) explicitly in terms of the amplitudes, b_n and b_m, corresponding to the quasilevels lying at distances equal to $|G_{mn}|^2/|\Omega|$ from the real levels.

Going over from the equations for the amplitudes to the equations for the $\hat{\rho}$-matrix elements, we finally obtain the following system of equations:

$$\frac{\partial n}{\partial t}=4\,\mathrm{Re}\left[i\left(\frac{d_{nm}E_p^*}{2\hbar}-\frac{2d_{mn}E_3G_{nm}^2}{\hbar(|\Omega|+\Omega_g)^2}\right)\rho_{mn}\right], \tag{4a}$$

$$\frac{\partial\rho_{mn}}{\partial t}=\left[\frac{d_{mn}E_{\bar{p}}}{2\hbar}-\frac{2d_{nm}E_3^*G_{mn}^2}{\hbar(|\Omega|+\Omega_g)^2}\right]n, \tag{4b}$$

$$\left(\frac{\partial}{\partial z}\pm\frac{1}{v_{3z}}\frac{\partial}{\partial t}\right)\mathscr{E}_3^{\pm}=\frac{16\pi i\omega_3 d_{mn}G_{nm}^2}{v_{3z}(|\Omega|+\Omega_g)^2}\rho_{mn}\exp(\mp ik_{3z}z), \tag{4c}$$

$$\left(\frac{\partial}{\partial z}\pm\frac{1}{v_{pz}}\frac{\partial}{\partial t}\right)\mathscr{E}_p^{\pm}=\frac{4\pi i\omega_p d_{nm}}{v_{pz}}\rho_{mn}\exp(\mp ik_pz), \tag{4d}$$

where

$$n=\sum_j(|b_n^j|^2-|b_m^j|^2)\delta(\mathbf{r}-\mathbf{r}_j),\qquad \rho_{mn}=\sum_j b_mb_n^*\delta(\mathbf{r}-\mathbf{r}_j),$$

and j is the number of the atom.

It is assumed that only the exciting field is incident on the medium. We choose as the initial value of the population difference the steady-state value established in the interaction of the atoms with the exciting field:

$$n(0)=[(|\Omega|+\Omega_g)^2-4|G_{mn}|^2]n_0/4\Omega_g^2. \tag{5}$$

The expression (5) is justified in view of the fact that the characteristic rise time of the field amplitudes $\mathscr{E}_3$ and $\mathscr{E}_p$ is long compared to the oscillation period (i.e., the fact that $\tau_0\Omega_g \gg 1$).

As the initial value, $\rho_{mn}(0)$, of the off-diagonal element, we choose the value that corresponds to the spontaneous noncooperative fluorescence. Performing estimates entirely similar to those carried out in Ref. 10, we obtain

$$\rho_{mn}(0)\exp(\mp ik_{p,3z}z)\approx\frac{n_0}{2}\left|\frac{G_{mn}}{\Omega}\right|(2\pi z^2\lambda n_0)^{-1/2}. \tag{6}$$

To estimate the characteristic fluorescence times, let us consider the linear problem in which the population difference is assumed to be a constant in time, a situation which corresponds to the initial phase of the process. Integrating Eq. (4b), and substituting the result into the equations, (4c) and (4d), for the waves propagating in the direction of the exciting field, we obtain the following system of equations:

$$\frac{\partial^2\theta_3}{\partial\tau\,\partial z}=\beta_3(\theta_3-\theta_pe^{-2iqz}),\qquad \frac{\partial^2\theta_p}{\partial\tau\,\partial z}=-\beta_p(\theta_p-\theta_3e^{2iqz}), \tag{7}$$

where

$$\theta_3=\frac{4G_{mn}^2}{(|\Omega|+\Omega_g)^2}\int_0^t dt'\,\frac{d_{nm}\mathscr{E}_3^{+*}}{\hbar}+\theta_0,\qquad \theta_p=\int_0^t dt'\,\frac{d_{mn}}{\hbar}\mathscr{E}_p^{+}+\theta_0,$$

$$\tau=t-z/v,\qquad 2q=2k_z-k_{3z}-k_{pz},\ \beta_3=\frac{8\pi\omega_3|d_{nm}|^2|G_{mn}|^4n(0)}{v\hbar(|\Omega|+\Omega_g)^4}$$
$$\beta_p=\frac{2\pi\omega_p|d_{nm}|^2n(0)}{v\hbar}.$$

In deriving Eqs. (7), we neglected the group-velocity dispersion, assuming that $v_{3z}=v_{pz}=v$.

The system of equations (7) formally coincides with the system of equations that describes nonresonance cooperative Raman scattering. The field emitted at the frequency ω_3 is then equivalent to the Stokes component, while the field emitted at the frequency ω_p corresponds to the anti-Stokes component. The second terms in the parentheses on the right-hand sides of Eqs. (7) describe the four-wave interaction of the fields that corresponds to the conservation a law $2\omega=\omega_3+\omega_p$. In the general case the parametric coupling of the fields may have a significant effect on the temporal and spatial characteristics of cooperative fluorescence. In the present paper we analyze the case (the simplest one) in which there is no spatial coherence, i.e., in which $qz_{ch} \gg 1$, where z_{ch} is the characteristic build-up length for the fields $\mathscr{E}_3$ and $\mathscr{E}_p$.

The Eqs. (7) without the parametric terms are considered in Ref. (10), which is devoted to the investigation of nonresonance cooperative Raman scattering. To the initial and boundary conditions

$$\theta_{3,p}(z,\tau=0)=\theta_0,\qquad \mathscr{E}_{3,p}(z=0,\tau)=0, \tag{8}$$

where $\theta_0 \approx |G_{mn}/\Omega|(2\pi n_0\lambda z^2)^{-1/2}$ correspond the self-similar solutions

$$\theta_3=\theta_0I_0(\beta_3^{1/2}\eta),\qquad \theta_p=\theta_0J_0(\beta_p^{1/2}\eta), \tag{9}$$

where $\eta=2(z\tau)^{1/2}$, J_0 is the zeroth-order Bessel function, and I_0 is the zeroth-order Neumann function of the first kind.

As follows from the expressions (9), the fluorescence at the super-Raman frequency ω_3 intensifies in time (exponentially at large values of the arguments), while the fluorescence at the frequency ω_p attenuates as it oscillates. It is clear that, qualitatively, the formulas (9) describe the behavior of the functions θ_3 and θ_p right up to the point where the population difference n changes its sign. Subsequently, the super-Raman and resonance components exchange roles. The characteristic pulse lengths $\tau_{03,p}$ of the super-Raman and resonance lines and the time lag of the super-Raman line are given by the formulas (see Ref. 10)

$$\tau_{03,p}=\frac{1}{\beta_{3,p}z}\ln\frac{\pi}{2\theta_0},\qquad t_{03}=\frac{\tau_{03}}{4}\ln\frac{\pi}{2\theta_0}. \tag{10}$$

Let us estimate the times $\tau_{03,p}$ and t_{03} with the following parameter values: $\Omega=10\ \mathrm{cm}^{-1}$, $\gamma=10^8\ \mathrm{sec}^{-1}$, $n_0=10^{16}\ \mathrm{cm}^{-3}$, $z=3$ cm, $I=300\ \mathrm{kW/cm}^2$, and $\lambda=0.7$

μm. Then $\tau_{03} \approx 1.3 \times 10^{-10}$ sec, $\tau_{0p} \approx 1.7 \times 10^{-13}$ sec, and $t_{03} \approx 6 \times 10^{-10}$ sec.

Let us note that, for the nonstationary cooperative fluorescence to appear, the front of the exciting pulse should be shorter than the characteristic time of disturbance of the phase of the atomic polarization; in the case of the opposite relationship between the times the stationary cooperative-fluorescence regime, which is investigated in Refs. 1–4, is realized.

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