

- ¹L. D. Landau and E. M. Lifshitz, *Kvantovaya Mekhanika* (Quantum Mechanics), Nauka, M., 1974 (English translation Pergamon Press, Oxford, 1975).
- ²C. S. Chang and P. Stehle, *Phys. Rev. A* **4**, 641 (1971).
- ³N. B. Delone and V. P. Krainov, *Atom v sil'nom svetovom pole* (Atoms in Strong Light Fields), Atomizdat, M., 1978.
- ⁴B. R. Mollow, *Phys. Rev. A* **5**, 2217 (1972).
- ⁵S. Swain, *J. Phys. A* **5**, 1587 (1972).
- ⁶A. Guccione-Gush and H. P. Gush, *Phys. Rev. A* **10**, 1474 (1974).
- ⁷D. Proznitz, D. W. Wildman, and E. V. George, *Phys. Rev. A* **16**, 1165 (1977).

- ⁸A. O. Mel'kyan and K. Kh. Simonyan, in: "Tezisy dokladov Vsesoyuznogo soveshchaniya po nelineinomu preobrazovaniyu chastoty" (Abstracts of Papers read to the All-Union Conf. on Nonlinear Frequency Conversion), Krasnoyarsk State University, 1977, p. 72.
- ⁹A. E. Kaplan, *Zh. Eksp. Teor. Fiz.* **65**, 1416 (1973) [*Sov. Phys. JETP* **38**, 704 (1974)].
- ¹⁰V. P. Krainov, *Zh. Eksp. Teor. Fiz.* **70**, 1197 (1976) [*Sov. Phys. JETP* **43**, 622 (1976)].

Translated by S. Chomet

Superradiance in Raman scattering of light

V. I. Emel'yanov and V. N. Seminogov

Moscow State University

(Submitted 6 June 1978)

Zh. Eksp. Teor. Fiz. **76**, 34-46 (January 1979)

A multimode theory of superradiance in Raman scattering (SRRS) of light in atomic and molecular systems is developed. The process of formation of the superradiant state from an initially incoherent state via exchange of spontaneously emitted photons between the atoms is considered in explicit form. The time dynamics of the populations in the waveform of SRRS pulse as well as the angular structure of the radiation are investigated. The influence of the depletion of the pump is estimated and an additional condition is derived for the density of the number of scattering atoms, namely, n has an upper bound besides the lower bound, $n_{\min} < n < n_{\max}$. It is noted that, as a result, the observation of SRRS is most probable in gaseous media.

PACS numbers: 42.50. + q, 42.65.Cq

1. INTRODUCTION

The effect of collective spontaneous emission of a sum of two-level atoms (the Dicke superradiance¹) was by now investigated quite fully both theoretically¹⁻⁶ and experimentally.^{7,8} Much less investigated is the analog of this effect in Raman (RS) of light in molecular and atomic systems—the effect of superradiant Raman scattering (SRRS). The paper devoted to this question can be divided into two classes.

The first includes papers^{9,10} dealing with RS in a medium excited beforehand by a coherent field. The macroscopic polarization induced by this field leads to the onset of a nonstationary RS, whose intensity is proportional to the square of the number N of the scattering particles. The interatomic interactions due to the radiation field of the atoms themselves are not important in this case. An effect of this type was observed in experiment in Ref. 11.

In a study belonging to the second class¹² a single-mode model was used to consider the onset of SRRS in an initially incoherent system of atoms via spontaneous induction of interatomic correlations. The analysis in Ref. 2 is in the given-pump-field approximation. In this approximation, the problem turns out to be similar to that of superradiance of a system of two-level atoms.¹⁻⁶ The SRRS takes in this case the form of a pulse of duration τ_p , whose maximum is observed

at the instant t_m (delay time). The SRRS intensity at the instant t_m is proportional to N^2 . Just as in the case of resonant superradiance, the condition for the observation of the SRRS is of the form $t_m \sim 1/n < T_2$ (T_2 is the transverse-relaxation time and, n is the density of the number of the scattering atoms). This means that at a given pump intensity I_L there is a lower bound of the density of the medium, $n > n_{\min}(I_L)$.

The single-mode model used in Ref. 12 does not make it possible to consider a large number of important characteristics of SRRS (including the very condition of the applicability of the single-mode approximation). In the present paper, using the given-pump-field approximation, we develop a multimode theory of SRRS for a medium of arbitrary geometric shape. This makes it possible to consider in explicit form the process of formation of the superradiant state from an initially incoherent state via exchange of spontaneously emitted Stokes phonons by the atoms. This process determines the delay time t_m , which depends substantially on the geometry of the medium. We investigate the angular directivity of the radiation in SRRS. The results of the present paper are applicable also to the case of resonant superradiance in a system of initially inverted two-level atoms.

The expression obtained for t_m differs from the corresponding formulas of Refs. 3 and 12. The reason is that in Refs. 3 and 12 the dynamics of the popula-

tions in all the stages of the time evolution, starting with $t=0$, was deduced by using the law of conservation of the length of the Bloch vector, but this law does not hold at short times (when account is taken of multi-mode spontaneous emission).

The formation of the SRRS is also influenced by transverse relaxation. It is shown in this paper that allowance for the finite time T_2 leads to an increase of the value of t_m . Under the condition $t_m \gg T_2$, SRRS is the analog of resonant superradiance in weakly amplifying media.¹³

The results obtained in the present paper for the multimode case are valid in the given-pump-field approximation. We have also calculated the single-mode SRRS model with account taken of the depletion of the pump. We present here only a qualitative estimate, which agrees with the calculated results. It turns out that allowance for the depletion of the pump leads to another condition for the observation of the SRRS, namely besides the lower bound on the density of the medium, there exists also an upper bound, $n_{\text{min}}(I_L) < n < n_{\text{max}}$. The reason is that the superscattering state arises at the instant when the populations of the working levels become equalized on account of the absorption of the pump energy. In a sufficiently dense medium, the pump is depleted before this occurs, and there is no superradiance. We note that allowance for the motion of the populations is the principal aspect that distinguishes the SRRS from the regime of non-stationary RS, which is usually described with the change of the populations neglected.¹⁴

2. THE MODEL. INITIAL EQUATIONS

We consider a system of N multilevel atoms (molecules) contained in a volume V of arbitrary geometric shape. The system is acted upon by an electromagnetic field, which we specify in the form of a plane wave:

$$\begin{aligned} E_L(\mathbf{R}, t) &= E_L^-(\mathbf{R}, t) + E_L^+(\mathbf{R}, t) \\ &= e_L(E_L \exp\{-i(\omega_L t - \mathbf{k}_L \mathbf{R})\} + E_L^* \exp\{i(\omega_L t - \mathbf{k}_L \mathbf{R})\}), \end{aligned} \quad (1)$$

where $E_L = 0$ at $t < 0$ and E_L is constant at $t > 0$ (the field pump is regarded as classical).

We assume that at the initial instant $t=0$ all the atoms are in the ground state, and the average polarization of the medium is zero. The RS produces in the medium a Stokes field at the frequency $\omega_s = \omega_L - \omega_{ab}$, where $\omega_{ab} > 0$ is the frequency of the transition of a selected pair of levels:

$$\begin{aligned} E_s(\mathbf{R}, t) &= E_s^-(\mathbf{R}, t) + E_s^+(\mathbf{R}, t) \\ &= (E_{sj}^- e^{-i\omega_{sj} t} + E_{sj}^+ e^{i\omega_{sj} t}). \end{aligned} \quad (2)$$

We write down the equations for the atomic and field operators. For a nonmagnetic medium, in the absence of free currents, the Hamiltonian of the "atoms plus field" system takes in the dipole approximation the form¹⁵

$$\hat{H} = \sum_{\alpha, j} \varepsilon_{\alpha} \sigma_{\alpha\alpha}^j + \frac{1}{8\pi} \int (\mathbf{H}^2 + \mathbf{D}^2) d\mathbf{R} - \int \mathbf{P} \mathbf{D} d\mathbf{R} + 2\pi \int (\mathbf{P}^\perp)^2 d\mathbf{R}. \quad (3)$$

Here \mathbf{H} is the magnetic-field intensity vector, $\mathbf{D} = \mathbf{E}$

+ $4\pi\mathbf{P}^\perp$ is the induction vector, $\mathbf{E} = \mathbf{A}_s + \mathbf{E}_L$,

$$\begin{aligned} \mathbf{P}(\mathbf{R}, t) &= \sum_{j, \alpha, \beta} (\mathbf{d}_{\alpha\beta} \sigma_{\beta\alpha}^j + \sigma_{\alpha\beta}^j \mathbf{d}_{\beta\alpha}) \delta(\mathbf{R} - \mathbf{R}_j), \\ \sigma_{\alpha\beta}^j &= a_{\beta j}^+ a_{\alpha j}, \end{aligned}$$

a_α^+ and a_α are the operators for the creation and annihilation of an atom in a state with energy ε_α , $\mathbf{d}_{\alpha\beta}$ is the dipole-moment matrix element, and

$$\mathbf{P}^\perp(\mathbf{R}, t) = \mathbf{P}(\mathbf{R}, t) + \frac{1}{4\pi} \int \frac{\text{grad}_{\mathbf{R}} \text{div} \mathbf{P}(\mathbf{R}', t)}{|\mathbf{R}' - \mathbf{R}|} d\mathbf{R}' \quad (4)$$

is the transverse polarization. The atomic operators $\sigma_{\alpha\beta}^j$ and $D_j = \sigma_{\alpha\alpha}^j - \sigma_{\beta\beta}^j$ satisfy the commutation relations

$$\begin{aligned} [\sigma_{\alpha\beta}^j, \sigma_{\beta\alpha}^j] &= -D_j \delta_{ij}, \quad [\sigma_{\beta\alpha}^j, D_j] = -2\sigma_{\beta\alpha}^j \delta_{ij}, \\ [\sigma_{\alpha\beta}^j, D_j] &= 2\sigma_{\alpha\beta}^j \delta_{ij}, \quad \sigma_{\alpha\beta}^j \sigma_{\beta\alpha}^j + \sigma_{\beta\alpha}^j \sigma_{\alpha\beta}^j = 1. \end{aligned}$$

Starting from (3), we write down the equation of motion for $\sigma_{\alpha\beta}^j$ and D_j . Using the averaging method of Ref. 16, we obtain the system

$$\begin{aligned} \frac{\partial D_j}{\partial t} &= -\frac{i\mathbf{r}_{\alpha\beta}}{\hbar} \{E_L^-(\mathbf{R}_j, t) E_s^+(\mathbf{R}_j, t) \sigma_{\beta\alpha}^j(t) + \sigma_{\beta\alpha}^j(t) E_L^-(\mathbf{R}_j, t) E_s^+(\mathbf{R}_j, t)\} + \text{H.c.} \end{aligned} \quad (5)$$

$$\begin{aligned} \frac{\partial \sigma_{\alpha\beta}^j}{\partial t} + i \left(\omega_{\alpha\beta} - \frac{i}{T_2} \right) \sigma_{\alpha\beta}^j &= \frac{i\mathbf{r}_{\alpha\beta}}{2\hbar} \{E_L^-(\mathbf{R}_j, t) E_s^+(\mathbf{R}_j, t) D_j(t) \\ &+ D_j(t) E_L^-(\mathbf{R}_j, t) E_s^+(\mathbf{R}_j, t)\}, \end{aligned} \quad (6)$$

where

$$\mathbf{r}_{\alpha\beta} = \frac{1}{\hbar} \sum_p \left\{ \frac{(\mathbf{d}_{\beta p} e_L) \mathbf{d}_{p\alpha}}{\omega_{\beta p} + \omega_L} + \frac{\mathbf{d}_{\beta p} (\mathbf{d}_{p\alpha} e_L)}{\omega_{\beta p} - \omega_L} \right\},$$

the line broadening is assumed homogeneous, and T_2 is a phenomenologically introduced relaxation time.

From the Hamiltonian (3) follows also an equation for the Stokes field

$$\frac{\partial^2 \mathbf{E}_s(\mathbf{R}, t)}{\partial t^2} - c^2 \Delta \mathbf{E}_s(\mathbf{R}, t) = -4\pi \frac{\partial^2 \mathbf{P}_s^\perp(\mathbf{R}, t)}{\partial t^2}; \quad (7)$$

\mathbf{P}_s^\perp is the transverse part of the polarization \mathbf{P}_s at the Stokes frequency. We obtain an expression for \mathbf{P}_s , using Ref. 16, in the form

$$\mathbf{P}_s(\mathbf{R}, t) = \sum_j \mathbf{P}_{sj} \delta(\mathbf{R} - \mathbf{R}_j) = - \sum_j \{ \mathbf{r}_{\alpha\beta} E_L^-(\mathbf{R}_j, t) \sigma_{\beta\alpha}^j(t) + \text{H.c.} \} \delta(\mathbf{R} - \mathbf{R}_j). \quad (8)$$

The solution of Eqs. (7) and (8) can be represented as a sum of the solution of the homogeneous and inhomogeneous equations, and the solution of the latter in the "wave zone" can be written out, with (4) taken into account, in explicit form¹⁷

$$\mathbf{E}_s(\mathbf{R}, t) = \frac{1}{c^2} \sum_j \frac{1}{R_{ij}} \left[\left[\frac{\partial^2}{\partial t'^2} \mathbf{P}_{sj}(t') \mathbf{n}_{ij} \right] \mathbf{n}_{ij} \right] + E_{s0}(\mathbf{R}, t), \quad (9)$$

where $t' = t - R_j/c$, $\mathbf{n}_{ij} = \mathbf{R}_{ij}/|\mathbf{R}_{ij}|$, and $\mathbf{R}_{ij} = \mathbf{R}_i - \mathbf{R}_j$.

Since there is no external field at the Stokes-wave frequency, the homogeneous solution E_{s0} corresponds to the vacuum field

$$\begin{aligned} E_{s0}(\mathbf{R}, t) &= i \sum_{\mathbf{k}} (2\pi\hbar kc/V)^{1/2} e_{\mathbf{k}} [a_{\mathbf{k}} \exp\{-i(kct - \mathbf{k}\mathbf{R})\} \\ &- a_{\mathbf{k}}^+ \exp\{i(kct - \mathbf{k}\mathbf{R})\}], \end{aligned} \quad (10)$$

here $a_{\mathbf{k}}^+$ and $a_{\mathbf{k}}$ are the operations of creation and

annihilation of the field quanta, with

$$\langle a_{\mathbf{k}} a_{\mathbf{k}'}^+ \rangle = \delta_{\mathbf{k}\mathbf{k}'}, \quad \langle a_{\mathbf{k}}^+ a_{\mathbf{k}'} \rangle = 0. \quad (11)$$

The angle brackets here and below denote average of the operators over the vacuum state of the field and over the ensemble.

The change to slowly varying variables ρ_{abj} :

$$\sigma_{ab}^j = \rho_{abj}(t) \exp \{-i\omega_{ab}t + ik_L R_{ij}\}. \quad (12)$$

Using (1) and (2) and neglecting the derivatives of the slowly varying density matrix ρ_{abj} , we obtain from (9) an expression for the amplitude of the Stokes field produced at an arbitrary point R_j , by the radiation of the atoms:

$$\begin{aligned} E_{ij}^+ &= \frac{\omega_s^2}{c^2} \sum_i \frac{1}{R_{ij}} \rho_{abj}(t') [[\mathbf{d}_s \times \mathbf{n}_{ij}] \times \mathbf{n}_{ij}] \exp \left(-i \frac{\omega_s}{c} R_{ij} \right), \\ E_{ij}^- &= (E_{ij}^+)^+, \end{aligned} \quad (13)$$

here $\mathbf{d}_s = \mathbf{r}_{ab} \mathbf{E}_L$.

We substitute (12) and (9) in (5), with (2) and (13) taken into account, and average the resultant operator equation:

$$\begin{aligned} \frac{\partial \langle D_j \rangle}{\partial t} &= \frac{\partial \langle D_j \rangle_0}{\partial t} - \left(\frac{i\omega_s}{\hbar c^3} \sum_i \frac{\exp \{-i\omega_s R_{ij}/c\}}{\omega_s R_{ij}/c} \right. \\ &\quad \left. \times \kappa_{ij} \langle \rho_{abj}(t') \rho_{baaj}(t) + \rho_{baaj}(t) \rho_{abj}(t') \rangle + \text{c. c.} \right), \end{aligned} \quad (14)$$

where $\kappa_{ij} = (\mathbf{d}_s \cdot [\mathbf{d}_s^* \times \mathbf{n}_{ij}] \times \mathbf{n}_{ij})$ and

$$\frac{\partial \langle D_j \rangle_0}{\partial t} = -\frac{i\mathbf{d}_s}{\hbar} \langle \mathbf{E}_{s0}^+(\mathbf{R}_j, t) \rho_{baaj}(t) + \rho_{baaj}(t) \mathbf{E}_{s0}^+(\mathbf{R}_j, t) \rangle e^{-i\omega_s t} + \text{H.c.} \quad (15)$$

The term $\partial \langle D_j \rangle_0 / \partial t$ in (14) describes the influence of the zero-point field fluctuations on the dynamics of the population difference. The remaining terms with $i \neq j$ take into account the influence of the correlations produced between the different atoms by the interaction via the dipole-radiation field.

The equation for the operator ρ_{baaj} is obtained from (6) in similar form

$$\begin{aligned} \frac{\partial \rho_{baaj}(t)}{\partial t} &= \frac{i\mathbf{d}_s}{2\hbar} \{ \mathbf{E}_{s0}^+(\mathbf{R}_j, t) D_j(t) + D_j(t) \mathbf{E}_{s0}^+(\mathbf{R}_j, t) \} e^{-i\omega_s t} \\ &+ \frac{i\omega_s^3}{2\hbar c^3} \sum_i \exp \left(-i \frac{\omega_s}{c} R_{ij} \right) \left(\frac{\omega_s}{c} R_{ij} \right)^{-1} \kappa_{ij} (\rho_{abj}(t') D_j(t) + D_j(t) \rho_{abj}(t')). \end{aligned} \quad (16)$$

In the derivation of (16) we have put $T_2 = \infty$, assuming that $t_m \ll T_2$. For a discussion of the case $t_m \gtrsim T_2$ see formula (46) below.

As seen from (14) and (16), the interaction between the atoms takes place with a delay due to the finite speed of light. We assume that the maximum linear dimension of the system L is such as to satisfy the condition

$$L/c \ll \tau_p, \quad (17)$$

where τ_p is the characteristic superradiance process (the characteristic time of the variation of D or ρ_{abj}). We can now put $t' = t - R_{ij}/c \approx t$ in (14) and (16). It is seen from (14) that the dynamics of $\langle D \rangle$ is determined

by the correlations $\langle \rho_{abj}(t) \rho_{baaj}(t) \rangle$. The correlators with $i = j$ make a contribution that describes the spontaneous emission of the individual atoms. The correlators with $i \neq j$ are responsible, as we shall verify later, for the onset of the superradiation.

The equation for the correlation with $i \neq j$

$$P_{ij}(t) = \langle \rho_{abj}(t) \rho_{baaj}(t) + \rho_{baaj}(t) \rho_{abj}(t) \rangle \quad (18)$$

that determines the superradiant dynamics of $D_j(t)$ is obtained from (15):

$$\frac{\partial P_{ij}}{\partial t} = \left(\frac{\partial P_{ij}}{\partial t} \right)_0 + \frac{\omega_s^3}{\hbar c^3} \sum_{k \neq i} C_{ki} \kappa_{ik} P_{kj} \langle D_i \rangle + \frac{\omega_s^3}{\hbar c^3} \sum_{k \neq j} C_{kj} \kappa_{jk} P_{ik} \langle D_j \rangle \quad (i \neq j), \quad (19)$$

where

$$C_{ij} = \sin \left(\frac{\omega_s}{c} R_{ij} \right) / \left(\frac{\omega_s}{c} R_{ij} \right).$$

In the derivation of (19) we have neglected the terms that describe the effect of the shift of the frequency of the atomic transition in superradiance

$$\begin{aligned} \left(\frac{\partial P_{ij}}{\partial t} \right)_0 &= \frac{i\mathbf{d}_s}{\hbar} \langle \mathbf{E}_{s0}^+(\mathbf{R}_i, t) \rho_{baaj}(t) + \rho_{baaj}(t) \mathbf{E}_{s0}^+(\mathbf{R}_i, t) \rangle \langle D_i \rangle e^{-i\omega_s t} \\ &- \frac{i\mathbf{d}_s^*}{\hbar} \langle \rho_{abj}(t) \mathbf{E}_{s0}^-(\mathbf{R}_j, t) + \mathbf{E}_{s0}^-(\mathbf{R}_j, t) \rho_{abj}(t) \rangle \langle D_j \rangle e^{i\omega_s t}. \end{aligned} \quad (20)$$

The terms $\partial \langle D_j \rangle_0 / \partial t$ (15) and $(\partial P_{ij} / \partial t)_0$ (20) are important at the initial time of the evolution, before the superradiance process has managed to establish itself. We therefore calculate then by using for the zeroth approximation of the solution of (16) the expression

$$\rho_{baaj}(t) = \frac{i\mathbf{d}_s}{2\hbar} \int_0^t \{ \mathbf{E}_{s0}^+(\mathbf{R}_j, \tau) D_j(\tau) + D_j(\tau) \mathbf{E}_{s0}^+(\mathbf{R}_j, \tau) \} e^{-i\omega_s \tau} d\tau. \quad (21)$$

We substitute (21) in (15) and (20), using for \mathbf{E}_{s0} the expansion (10). Then, after averaging over the ensemble, over the orientations of \mathbf{d}_s , and over the vacuum state of the field with allowance for (11), and also after summing over all the modes of the field \mathbf{k} , we obtain

$$\partial \langle D_j \rangle_0 / \partial t = -\Gamma \langle D_j \rangle, \quad (22)$$

$$(\partial P_{ij} / \partial t)_0 = \Gamma C_{ij} \langle D_i \rangle \langle D_j \rangle \quad (i \neq j), \quad (23)$$

where $\Gamma = 2\omega_s^3 |\mathbf{r}|^2 |E_L|^2 / 5\hbar c^3$, and

$$|\mathbf{r}| = \frac{1}{\hbar} \left| \sum_p \left(\frac{d_{bp} d_{pa}}{\omega_{bp} + \omega_L} + \frac{d_{bp} d_{pa}}{\omega_{bp} - \omega_L} \right) \right| \quad (24)$$

We now average (14) and (19) over the orientations of the dipole moment ($\kappa_{ij} \approx - (1/5) |\mathbf{r}|^2 |E_L|^2$) and substitute in them the expressions (22) and (23). Taking into account the commutation relations for the operators D_j and ρ_{abj} , we get

$$\frac{\partial \langle D \rangle}{\partial t} = \Gamma \sum_{i \neq j} C_{ij} P_{ij} + \Gamma (N - \langle D \rangle); \quad (25)$$

$$\frac{\partial P_{ij}}{\partial t} = -\Gamma C_{ij} \{ \langle \rho_{bbj} \rangle \langle D_i \rangle + \langle \rho_{bbi} \rangle \langle D_j \rangle \} - \frac{\Gamma}{2} \sum_{k \neq i, j} C_{ki} P_{kj} \langle D_i \rangle - \frac{\Gamma}{2} \sum_{k \neq i, j} C_{kj} P_{ik} \langle D_j \rangle \quad (i \neq j), \quad (26)$$

where

$$\langle D(t) \rangle = \sum_i \langle D_i(t) \rangle.$$

Eq. (25) describes the dynamics of the total atom population difference. At the initial instant of time $t = 0$, $P_{ij} = 0$, $\langle D \rangle = -N$. The start of the time evolution of $\langle D \rangle$ is determined by the spontaneous RS, to which the last term of (25) corresponds. The first term in the right-hand side of (26) describes the induction of interatomic correlations by spontaneous emission of a Stokes photon from one atom, and the reaction to it by another atom. Without this term, as seen from (26), we find at the specified initial conditions ($P_{ij}(t=0) = 0$) that $P_{ij}(t) = 0$ at all instants of time and there is no superradiance.

By virtue of the condition (17), the spatial dimension of the SRRS pulse is $c\tau_p \gg L$. We can assume therefore in (26) that the differences of the populations of all the atoms are the same:

$$\langle D_i \rangle \approx \langle D \rangle / N \quad (27)$$

(we shall henceforth omit the angle brackets).

To find the solution of (25)–(27), it is convenient to introduce the eigenfunctions $\psi_\lambda(\mathbf{R}_j)$ and the eigenvalues λ of the interatomic-interaction matrix C_{ij} (Ref. 6):

$$\sum_{j=1}^N C_{pj} \psi_\lambda(\mathbf{R}_j) = \lambda \psi_\lambda(\mathbf{R}_p), \quad (28)$$

where

$$C_{pj} = \frac{\sin(\omega_s R_{pj}/c)}{\omega_s R_{pj}/c} = \frac{1}{4\pi} \int \exp(ik\mathbf{R}_{pj}) d\Omega, \quad |k| = \frac{\omega_s}{c}, \quad (29)$$

and $d\Omega$ is the solid-angle element.

Let the eigenfunctions satisfy the completeness and orthonormalization condition:

$$\sum_{j=1}^N \psi_\lambda(\mathbf{R}_j) \psi_{\lambda'}(\mathbf{R}_j) = \delta_{\lambda\lambda'}, \quad \sum_{\lambda} \psi_\lambda(\mathbf{R}_j) \psi_\lambda(\mathbf{R}_p) = \delta_{jp}. \quad (30)$$

As follows from (28)–(30) that

$$C_{ij} = \sum_{\lambda} \lambda \psi_\lambda(\mathbf{R}_i) \psi_\lambda(\mathbf{R}_j). \quad (31)$$

We introduce the collective quantity

$$P(\lambda, t) = \sum_{i \neq j} P_{ij}(t) \psi_\lambda(\mathbf{R}_i) \psi_\lambda(\mathbf{R}_j). \quad (32)$$

Then, using (28)–(31), we obtain from (25)–(27) the system

$$\frac{\partial D}{\partial t} = \Gamma \sum_{\lambda} \lambda P(\lambda, t) + \Gamma(N-D), \quad (33)$$

$$\frac{\partial P(\lambda, t)}{\partial t} = -\frac{\Gamma(N-D)D}{N^2}(\lambda-1) - \frac{\Gamma D}{N}(\lambda-1)P(\lambda, t) + \frac{\Gamma D}{N} \sum_{j \neq k} C_{kj} \psi_\lambda(\mathbf{R}_j) \psi_\lambda(\mathbf{R}_k) P_{kj}(t). \quad (34)$$

We note that by virtue of the definition (32) we have $\sum_{\lambda} P(\lambda, t) = 0$. Summing both halves of (34) over λ , we can easily verify that both halves of (34) vanish.

Eqs. (33) and (34) are valid for arbitrary geometry of the scattering medium.

3. DYNAMICS OF POPULATION DIFFERENCE. SHAPE OF THE SRRS PULSE. ANGULAR DIRECTIVITY PATTERN OF THE RADIATION

We consider the particular case of a cylindrical volume. The characteristic geometrical parameters are

$$H = \omega_s L/c, \quad h = \omega_s R/c \quad (H \gg h \gg 1),$$

where L is the length of the cylinder, and R and S are the radius and the cross-section area. In the limit of small and large Fresnel numbers ($F = S/\lambda_s L$, where λ_s is the Stokes-emission wavelength) all the largest eigenvalues and the corresponding $\psi_\lambda(\mathbf{R}_j)$ are explicitly defined.⁶ For $F \ll 1$ and $F \gg 1$ all the large eigenvalues are degenerate with multiplicity g_0 and are equal to

$$\lambda = \lambda_0 = N\pi/2H \gg 1, \quad g_0 = 1/F \gg 1 \quad (F \ll 1); \quad (35)$$

$$\lambda = \lambda_0 = N/h^2 \gg 1, \quad g_0 = 2F \gg 1 \quad (F \gg 1). \quad (36)$$

The remaining $\lambda \ll \lambda_0$, and the eigenfunctions take the form

$$\psi_\lambda(\mathbf{R}_j) = \frac{1}{\sqrt{N}} (\cos \mathbf{k}_0 \mathbf{R}_j + \sin \mathbf{k}_0 \mathbf{R}_j), \quad \mathbf{k}_0 = \frac{\omega_s}{c} \hat{\mathbf{k}}_0, \quad (37)$$

where \mathbf{k}_0 is a unit vector directed along the cylinder axis.

We obtain the solution of the system (33) and (34) for the cases (35) and (36). At $\lambda = \lambda_0$, the last term in (34) is of the order of $g_0/N \ll 1$ of the second term in the right-hand side, and can be neglected. It follows then from (34) that all the $P(\lambda_0, t)$ are the same and we have in (33)

$$\sum_{\lambda} \lambda P(\lambda, t) \approx g_0 \lambda_0 P(\lambda_0, t).$$

Recognizing this, we can rewrite (33) and (34) in the form

$$\frac{\partial D}{\partial t} = \Gamma \lambda_0 g_0 P(\lambda_0, t) + \Gamma(N-D), \quad (38)$$

$$\frac{\partial P(\lambda_0, t)}{\partial t} = -\frac{\Gamma(N-D)D}{N^2} \lambda_0 - \frac{\Gamma D}{N} \lambda_0 P(\lambda_0, t). \quad (39)$$

We note that when the substitutions $D \rightarrow -D$ and $\Gamma \rightarrow \gamma \equiv (4/3) \omega_{ab}^3 |d_{ab}|^2 / \hbar c^3$ are made, Eqs. (38) and (39) describe the process of superradiant emission of an initially inverted system of two-level atoms with transition frequency ω_{ab} .

Eliminating $P(\lambda_0, t)$ we obtain an equation for $D(t)$

$$\frac{\partial^2 D}{\partial t^2} + \frac{\Gamma \lambda_0}{2N} \frac{\partial D^2}{\partial t} - \Gamma \frac{\partial}{\partial t} (N-D) = \frac{\Gamma^2 D(N-D)}{N} \lambda_0 - \frac{\Gamma^2 D(N-D)}{N^2} \lambda_0^2 g_0, \quad (40)$$

with initial conditions

$$D(t=0) = -N, \quad \frac{\partial}{\partial t} D(t=0) = 2\Gamma N.$$

If we neglect the terms of the right-hand side of (40), then the solution takes the form

$$D(t) = -D(0) \operatorname{th} \left(\frac{t - t_m^*}{\tau_p} \right) \quad (41)$$

$$\tau_p = 2/\Gamma\lambda_0, \quad t_m^* = 1/2 \tau_p \ln \lambda_0, \quad (42)$$

where λ_0 is given by (36) for $F \gg 1$ or (35) for $F \ll 1$. From (38) and (39), neglecting the spontaneous terms, we get

$$D^2 + 2Ng_0 P(\lambda_0, t) \approx N^2, \quad (43)$$

whence, taking (41) into account,

$$P(\lambda_0, t) = \frac{N}{2g_0} \left[\operatorname{ch}^2 \left(\frac{t - t_m^*}{\tau_p} \right) \right]^{-1}. \quad (44)$$

At the instant $t = t_m^*$ we have $D(t) = 0$, $P(\lambda_0, t) = \max$, and the system is in a superradiant state. For large F , the phonon for t_m^* agrees with the result of Ref. 12 (with account taken of the correction introduced for the multiple modes). The very same result is obtained also in the theory of resonant superradiance.³

It is impossible to obtain an analytic solution of the complete equation (40). From its general form it follows, however, that the delay time t_m should in fact be larger than predicted by (41), (44), and (42), since the right-hand side of (40) vanishes when the spontaneous component in (39) is increased by a factor $N/\lambda_0 g_0 \gg 1$.

We can estimate t_m approximately in the following manner. Assuming that $D(t) \approx -\text{const}$ at $0 \leq t \leq t_m$ we get from (39) and (43)

$$P(\lambda_0, t_m) \approx 2(e^{2\lambda_0 t_m} - 1) \approx N/2g_0.$$

Hence

$$t_m \approx \frac{1}{\Gamma\lambda_0} \ln \frac{N}{g_0}. \quad (45)$$

The expression for t_m can be represented, taking (35) and (36) into account, in the form

$$t_m = \begin{cases} [\ln \lambda_0 + \ln H]/\Gamma\lambda_0, & F \gg 1 \\ [\ln \lambda_0 + \ln(h^2/\pi)]/\Gamma\lambda_0, & F \ll 1 \end{cases}$$

i.e., $t_m > t_m^*$.

We note that if $g_0 \approx 1$, then formula (45) corresponds to the result of the single-mode model.¹² Thus, the condition for the validity of the single-mode approximation for the description of SRRS is $g_0 \sim 1$, i.e., $F \sim 1$, in agreement with the conclusion obtained in Ref. 6 with respect to resonant superradiance. A numerical solution of the system (38) and (39) confirms the estimate (45). Figs. 1 and 2 show plots of $P(\lambda_0, t)$ and $D(t)$, obtained by numerically solving (38) and (39); they are compared with the plots of P and D corresponding to formulas (44) and (41). The fact that we obtained a delay time t_m longer than t_m^* can be attributed to the following: In Ref. 3 and 12, the law of conservation of the length of the Bloch vector is used to obtain $D(t)$ during all the stages of the time evolution,

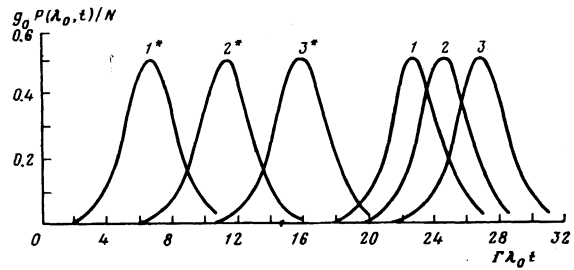


FIG. 1. Shape of SRRS pulse: comparison of the delay times with t_m and t_m^* . Curves 1-3) numerical solution of (38) and (39); 1*-3*) calculation by formula (44). We assumed $L = 10$ cm, $S = 1$ cm², and $N = 10^{14}$. $\lambda_0 = 10^{-5}$ cm for cases 1, 1', 10^{-4} cm for cases 2, 2', and 10^{-2} cm for cases 3, 3'.

starting with $t = 0$. For the multimode mode this law takes in our notation the form (43). Formula (43) follows from (38) and (39) in which the spontaneous terms are neglected, and is therefore not valid at short times. It is easily seen that at the instant $t = 0$ formula (43) gives for the growth rate of the correlator of the collective polarization $(\partial P(\lambda_0, t)/\partial t)$ a value $N/\lambda_0 g_0 \gg 1$ times larger than then the one given by (39).

It follows from the derivation that the results given above are valid for the case $t_m \geq T_2$. On the other hand if $t_m \geq T_2$, then it is necessary to add a term $-2T_2^{-1}P(\lambda_0, t)$ to the right-hand side of (39). We then obtain from (38) and (39), in analogy with the procedure used in the derivation of (45)

$$t_m = \frac{\tau_p}{2} \left(1 - \frac{\tau_p}{T_2} \right)^{-1} \ln \frac{N}{g_0} \left(1 - \frac{\tau_p}{T_2} \right). \quad (46)$$

Thus, SRRS is possible also in the case when $t_m \geq T_2$ but $\tau_p < T_2$. This corresponds to the case of a resonant superradiance in weakly amplifying media.¹³

We proceed now to consider the shape of the SRRS pulse and the angular directivity of the radiation. The energy of the scattered field at the point of observation is

$$W_s(\mathbf{R}_p) = \frac{\langle |\mathbf{E}_s(\mathbf{R}_p, t)|^2 \rangle}{4\pi} = \frac{1}{4\pi} \langle \mathbf{E}_{s0}^-(\mathbf{R}_p, t) \mathbf{E}_s^+(\mathbf{R}_p, t) + \mathbf{E}_s^-(\mathbf{R}_p, t) \mathbf{E}_{s0}^+(\mathbf{R}_p, t) \rangle + \frac{1}{4\pi} \langle \mathbf{E}_{sp}^+ \mathbf{E}_{sp}^- + \mathbf{E}_{sp}^- \mathbf{E}_{sp}^+ \rangle. \quad (47)$$

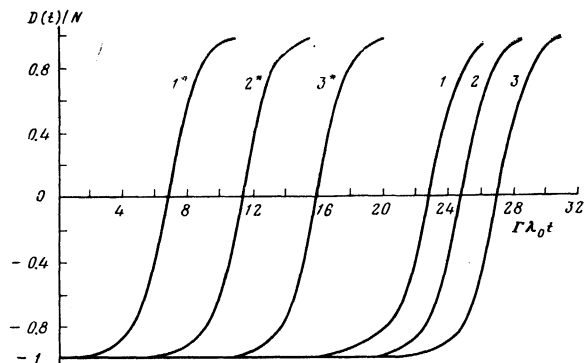


FIG. 2. Diameter of the population difference in SRRS. The parameters and notation are the same as in Fig. 1.

We substitute in the last two terms of (47) the expressions (13), and in the first two terms the expressions (10) and (13), where $\rho_{\alpha\beta i}$ is given by formula (21). After averaging we obtain in the far zone ($|\mathbf{R}_p| \gg L, L_2\omega_s/cR_p < \pi$)

$$W_s(\mathbf{R}_p) = \frac{\hbar\omega_s\Gamma}{4\pi cR_p^2} \rho_{\alpha\beta}(t) + \frac{\hbar\omega_s\Gamma}{8\pi cR_p^2} \sum_{i \neq j} \sum P_{ij}(t) e^{i\mathbf{k}\cdot\mathbf{R}_{ij}}, \quad (48)$$

$$\mathbf{k} = \frac{\omega_s}{c} \frac{\mathbf{R}_p}{|\mathbf{R}_p|} = \frac{\omega_s}{c} \hat{\mathbf{k}}.$$

We introduce the collective variable

$$P(\lambda, \lambda', t) = \sum_{i \neq j} \sum P_{ij} \psi_\lambda(\mathbf{R}_i) \psi_{\lambda'}(\mathbf{R}_j) \quad (49)$$

and the converse

$$P_{ij}(t) = \sum_{\lambda} \sum_{\lambda'} P(\lambda, \lambda', t) \psi_{\lambda'}(\mathbf{R}_i) \psi_{\lambda}(\mathbf{R}_j). \quad (50)$$

An equation for $P(\lambda, \lambda', t)$ is obtained from (26). It is similar in form to (39), with $P(\lambda, t) \rightarrow P(\lambda, \lambda', t)$, and with the first terms in the right-hand sides of (34) and (39) multiplied by $\delta_{\lambda\lambda'}$. Since $P(\lambda, \lambda', 0) = 0$ at $t = 0$, it follows that $P(\lambda, \lambda', t) = \delta_{\lambda\lambda'} P(\lambda, \lambda, t) = \delta_{\lambda\lambda'} P(\lambda, t)$, where $P(\lambda, t)$ is given by formula (32). Taking this into account and using formula (50), we obtain from (48) an expression for the energy scattered in the unit time into a unit solid angle in the direction of the unit vector $\hat{\mathbf{k}}$:

$$I_{s,k}(t) = \frac{\hbar\omega_s\Gamma}{4\pi} \left\{ \frac{N-D(t)}{2} + \frac{1}{2} \sum_{\lambda} P(\lambda, t) \sum_{i \neq j} \sum \psi_{\lambda}(\mathbf{R}_i) \psi_{\lambda}(\mathbf{R}_j) e^{i\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)} \right\}. \quad (51)$$

In the limiting cases $F \gg 1$ and $F \ll 1$, using (37), we obtain $I_{s,k}(t)$ in explicit form

$$I_{s,k}(t) = \frac{\hbar\omega_s\Gamma}{4\pi} \left\{ \frac{N-D(t)}{2} + \frac{N}{2} g_0 P(\lambda_0, t) \Theta(\mathbf{k}) \right\}, \quad (52)$$

where $D(t)$ and $P(\lambda_0, t)$ are given by formulas (41) and (44) [where we make the substitution $t_m^* \rightarrow t_m$ (45)], and the angular directivity factor of the SRRS is

$$\Theta(\mathbf{k}) = \frac{1}{2N^2} \sum_{i \neq j} \sum \left[\exp\{i(\mathbf{k}_0 + \mathbf{k}) \cdot (\mathbf{R}_i - \mathbf{R}_j)\} + \exp\{i(\mathbf{k}_0 - \mathbf{k}) \cdot (\mathbf{R}_i - \mathbf{R}_j)\} \right] \\ = 2 \sum_{(\pm)} \left[\frac{\sin^{1/2} H(1 \mp \cos \varphi)}{1/2 H(1 \mp \cos \varphi)} \right]^2 \left(\frac{J_1(\hbar \sin \varphi)}{\hbar \sin \varphi} \right)^2. \quad (53)$$

Here $\cos \varphi = \mathbf{k} \cdot \mathbf{k}_0 / |\mathbf{k}|^2$ (we recall that \mathbf{k}_0 is directed along the cylinder axis: $|\mathbf{k}| = |\mathbf{k}_0|$); J_1 is a Bessel function of the first kind.

The first term in (52) describes the isotropic spontaneous RS produced starting with the instant $t = 0$, the second term describes the collective radiation which is formed in the form of a pulse whose maximum is reached at the instant t_m . As follows from (52) and (53), the radiation in SRRS goes into small solid angles and in opposite directions along the cylinder axis. From (52), (42), and (44) it is seen that at the instant t_m the scattering power $I_{s,k}(t_m) \sim N^2$.

We note that when the substitutions $D \rightarrow -D$, $\omega_s \rightarrow \omega_{\alpha\beta}$, and $\Gamma \rightarrow \gamma$ are made, formula (51)–(53) describe the shape of the pulse and the directivity pattern of the resonant superradiance in a system of two-level atoms.

The results of the present paper were obtained in the given-pump-field approximation. This field can be regarded as given if the energy supplied during the radiation time τ_p exceeds the energy drawn from the pump field, i.e.,

$$|E_L|^2 c \tau_p S / 2\pi > n \hbar \omega_L S L,$$

whence, taking (42) and (36), and (24) into account, we get

$$n < \frac{c/L}{\pi (2\omega_s \omega_L)^{1/2} |r|} \equiv n_{\max}. \quad (54)$$

Thus, in contrast to the case of resonant superradiance, in the case of SRRS there exists an upper bound on the density of the number of scattering particles. At $L \sim 1$ cm, $\omega_s \sim \omega_L \sim 10^{15}$ sec⁻¹, and $|r| \sim 10^{-24}$ cgs esu we obtain $n_{\max} \sim 10^{19}$ cm⁻³. Our exact calculation of the SRRS process in the single-mode model, with allowance for the depletion of the pump,¹⁸ confirms the estimate (54). Consequently, observation of SRRS is more probable in gaseous media.

The authors thank S. A. Akhmanov for a discussion of the results.

¹R. H. Dicke, Phys. Rev. **93**, 99 (1954).

²R. Bonifacio, P. Shwendimann, and F. Haake, Phys. Rev. A **4**, 854 (1971).

³N. E. Rehler and J. H. Eberly, Phys. Rev. A **3**, 1735 (1971).

⁴V. I. Emel'yanov and Yu. L. Klimontovich, Opt. Spektrosk. **41**, 913 (1976) [Opt. Spectrosc. (USSR) **41**, 541 (1976)].

⁵A. V. Andreev, Yu. A. Il'inskiĭ, and R. V. Khokhlov, Zh. Eksp. Teor. Fiz. **73**, 1296 (1977) [Sov. Phys. JETP **46**, 682 (1977)].

⁶E. Ressayre and A. Tallet, Phys. Rev. A **15**, 2410 (1977).

⁷N. Skribanowitz, P. P. Herman, J. C. MacGillivray, and M. S. Feld, Phys. Rev. Lett. **30**, 309 (1973).

⁸H. M. Gibbs and O. H. F. Vrehen, Phys. Rev. Lett. **39**, 547 (1977).

⁹H. A. Hopf, R. Shea, and M. O. Scully, Phys. Rev. A **7**, 2105 (1973).

¹⁰T. M. Makhviladze, M. E. Sarychev, and L. A. Shelepin, Zh. Eksp. Teor. Fiz. **69**, 499 (1975) [Sov. Phys. JETP **42**, 255 (1975)].

¹¹R. L. Shoemaker and R. G. Brewer, Phys. Rev. Lett. **28**, 1430 (1972).

¹²S. G. Rautian and B. M. Chernobrod, Zh. Eksp. Teor. Fiz. **72**, 1342 (1977) [Sov. Phys. JETP **45**, 705 (1977)].

¹³A. V. Andreev, Pis'ma Zh. Tekh. Fiz. **3**, 779 (1977) [Sov. Tech. Phys. Lett. **3**, 317 (1977)].

¹⁴S. A. Akhmanov, K. N. Drabovich, A. P. Sukhorokov, and A. S. Chirkin, Zh. Eksp. Teor. Fiz. **59**, 485 (1970) [Sov. Phys. JETP **32**, 266 (1971)].

¹⁵S. Stenhol, Phys. Lett. C **6**, No. 1 (1973).

¹⁶V. S. Butylkin, Yu. G. Khronopulo, and E. I. Yakubovich, Zh. Eksp. Teor. Fiz. **71**, 1712 (1976) [Sov. Phys. JETP **44**, 897 (1976)].

¹⁷L. D. Landau and E. M. Lifshitz, Teoriya polya (Field Theory), Nauka, 1973 [Pergamon 1976].

¹⁸V. I. Emel'yanov and V. N. Seminogov, Kvantovaya Élektron. (Moscow) **6**, No. 4 (1979) [Sov. J. Quantum Electron. **9**, No. 4 (1979)].

Translated by J. G. Adashko