Two-level atom in a bichromatic resonance field

S. P. Goreslavskiĭ and V. P. Kraĭnov

Engineering-Physics Institute, Moscow

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An analysis is given of the interaction between a two-level atom and two monochromatic classical electromagnetic fields of arbitrary amplitude and frequency close to the frequency of the atomic transition. Published work on this problem is reviewed. An analytical solution is obtained for the case of identical fields, and equal and opposite detuning from resonance. It is shown that, in contrast to the single field, the time average of the upper-level population in the case of two resonance fields may exceed 50% for certain detuning to amplitude ratios, and may reach approximately 70% in the optimum case. The case of separated resonances is examined. It is shown that there are satellite resonances that can be physically interpreted as Raman scattering events with three, five, or more photons. Stark shifts of resonance peaks of the type mentioned above are determined. Analytic expressions are obtained for the width of single-photon resonances and multiphoton combination resonances. The opposite case of close external-field frequencies is also analyzed. The general case is investigated numerically.

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§1. INTRODUCTION

The behavior of an atomic system in a strong electromagnetic field has assumed considerable theoretical and experimental importance since the advent of powerful lasers. In this paper, we consider the case where only two atomic states participate in the interaction with high-intensity radiation. The atom is thus replaced by a two-level system which exhibits a dipole interaction with a strong classical electromagnetic field. All the relaxation processes are ignored, and the levels are assumed to be nondegenerate.

The behavior of a two-level system in a single monochromatic resonance field has been investigated in detail, since the work of Rabi, in the resonance approximation (rotating wave approximation, see, for example, Ref. 1). The case of two monochromatic fields, i.e., the so-called bichromatic field, has attracted much less attention because of its considerable complexity. The general formalism for the description of the interaction between a two-level system and several quantized electromagnetic fields was examined by Change and Stehle.[2] In particular, they examined the case of the bichromatic field. However, they reported specific results only for the case where the second field was much weaker than the first, and its effect could be reduced to a nonresonance shift of the resonance peak due to the first field.

The case where one of the fields (the probe field) was infinitely weak was discussed within the framework of perturbation theory by Delone and Kraĭnov[3] and by Mollow,[4] taking into account spontaneous radiative transitions within the two-level system. Both fields were assumed to be resonance fields, i.e., their frequencies were taken to be close to the level separation. A calculation was made of the transition probability in this system in the presence of the probe field as a function of its frequency.

The interaction of a two-level system with two modes of a quantized electromagnetic field was considered by Swain.[5] The modes were assumed to have equal and opposite detuning, and their amplitudes were taken to be equal. The essential assumption introduced by Swain[5] was that the following initial conditions were satisfied: the atom was in an excited state and the two modes of the quantized electromagnetic field were in the vacuum state, i.e., did not contain photons. It is important to note that this formulation of the problem completely excludes Raman processes with three, five, or more photons. For example, in three-photon Raman scattering, the atom should undergo a transition to the lower state and this should be accompanied by the emission of two photons of one of the modes and the annihilation of one photon of the other mode. This process is forbidden in the formulation given by Swain[5] because of the absence of photons corresponding to the second mode in the vacuum state.

We shall use similar conditions for the detuning and mode amplitudes in Sec. 3 to consider classical electromagnetic fields, i.e., fields containing a large number of photons corresponding to both modes. Raman scattering with three, five, and more photons will be possible and, in general, relatively important. The results of the calculations described in Sec. 3 are, therefore, different from those reported by Swain.[5] It is clear from the foregoing that, in the case of a single mode, for which Raman scattering of the above kind is absent, the quantum and classical approaches to the description of the electromagnetic field lead to the same result, namely, the Rabi solution. This is a well-known fact.

Guccione-Gush and Gush[6] have given a numerical solution for the interaction between two resonance fields and a two-level system for certain definite values of field parameters. They assumed that the frequencies ω_1 and ω_2 were close to the level separation ω_{ba}, and maintained that the number of effectively participating quasienergy harmonics in the wave function was small and that the situation was analogous to that prevailing in the case of a monochromatic field. They performed a numerical solution of the problem on the basis of this assumption. We shall see later (Sec. 3) that this assumption is not valid.

The effect of bichromatic CO_2-laser radiation on a two-level system consisting of the 4^3S and 4^3P states of helium was investigated experimentally by Prosnitz *et al.*[7] In addition, a third, weak, field transferring the particle to a third level from one of the resonating states was also introduced. When the electric field was of the order of 6×10^4 V/cm, the resonance saturation parameter, i.e., the ratio of the perturbation to the resonance detuning, was of the order of unity. The probability of transitions to the third level in the presence of the probe field was calculated on a computer within the framework of the resonance approximation for this particular system. However, numerous additional assumptions relating to the cutoff in the rank of the Hamiltonian matrix in the course of its diagonalization were introduced. Their validity is not clear. No comparison was made between theoretical and experimental data because of the uncertainty in the laser electric field at different points in the active volume of the medium.

The general equations for the case of the bichromatic field are discussed in Sec. 2 below, and the case of symmetric detuning and equal fields, which can be solved exactly, is examined in Sec. 3. Section 4 is devoted to the case where the field frequencies ω_1 and ω_2 are sufficiently different in comparison with their amplitudes. This section also considers different multiphoton combination resonances in the system. Section 5 gives a brief description of another limiting case in which ω_1 and ω_2 are close to one another, and, finally, Sec. 6 discusses a numerical calculation for the intermediate case.

As already mentioned, we shall neglect throughout the width of the upper level, γ, connected with spontaneous transitions to the lower state. This is valid for strong perturbations $V\gg\gamma$, and we shall assume that this is so. It is, of course, also assumed that $V\ll\omega_1$, ω_2.

If, in a realistic situation, spontaneous transitions with widths γ' to some other levels different from the two under examination are important, it must be assumed further that the time of operation of these fields, T, is restricted by the condition $\gamma' T\ll 1$ (no relaxation to other states).

§2. EQUATIONS FOR THE BICHROMATIC RESONANCE FIELDS

Consider the effect of two monochromatic fields, $V\cos\omega_1 t$ and $\alpha V\cos(\omega_2 t+\psi)$, on our two-level system on the assumption that the frequencies ω_1 and ω_2 are close to the level separation ω_{ba}. When this condition is satisfied, we can use the so-called generalized resonance or ladder approximations.[6] This takes into account only graphs in which the absorption of a photon of a particular type is accompanied by the subsequent emission of a photon (not necessarily of the same type) and vice versa.

In this approximation, the equations describing the behavior of the probability amplitudes $C_a(t)$ and $C_b(t)$ for finding a particle in the upper and lower levels are as follows:

$$\begin{aligned} i\,dC_a/d\varphi &= [e^{-i\lambda(x+1)\varphi}+\alpha e^{-i\lambda(x-1)\varphi+i\psi}]C_b,\\ i\,dC_b/d\varphi &= [e^{i\lambda(x+1)\varphi}+\alpha e^{i\lambda(x-1)\varphi-i\psi}]C_a. \end{aligned} \tag{1}$$

where, in dimensionless notation, $\hbar=1$, $\varphi\equiv\frac{1}{2}Vt$, $\lambda\equiv(\omega_2-\omega_1)/V$, and $x=(2\omega_{ba}-\omega_1-\omega_2)/(\omega_2-\omega_1)$.

The diagonal matrix elements (if they are present) lead to rapidly oscillating terms and do not contribute to (1).

Substituting

$$C_a=C_a'\exp(-i\lambda x/2),\quad C_b=C_b'\exp(i\lambda x/2),$$

in (1), we can readily show that C_a' and C_b' satisfy a set of equations with periodic coefficients with period equal to[8] $2\pi/\lambda$. This means that the Floquet theorem is valid for the case of the bichromatic fields (but only for the resonance approximation and not for the exact set of equations as for the single monochromatic field).

We shall now investigate the various exact and approximate solutions of (1) for different values of the parameters x, λ, α, ψ (to be specific, we assume that $0<\alpha<1$).

§3. THE CASE OF EQUAL FIELDS AND EQUAL AND OPPOSITE DETUNING

Consider the case where $\alpha=1$ and $x=0$. We thus assume that the detuning of the two resonances relative to ω_{ba} are equal in magnitude and opposite in direction. The set of equations given by (1) can then be solved exactly. To be specific, we suppose that $C_a(0)=1$ and find that

$$|C_b(t)|^2=\sin^2\left\{\frac{2}{\lambda}\left[\sin\left(\lambda\varphi+\frac{\psi}{2}\right)-\sin\frac{\psi}{2}\right]\right\}. \tag{2}$$

When $\lambda<1$, the Fourier series for $C_b(t)$ includes appreciable contributions due to harmonics with $n\leqslant 2/\lambda$, not only $n=0, \pm1$, as in the case of the monochromatic field.

From (2), we find that the mean population of the upper level, $w_b=\langle|C_b(t)|^2\rangle$, is given by

$$w_b=\frac{1}{2}\left[1-\cos\left(\frac{4}{\lambda}\sin\frac{\psi}{2}\right)J_0\left(\frac{4}{\lambda}\right)\right]. \tag{3}$$

Figure 1 shows w_b as a function of λ for $\psi=0$ and $\psi=\pi$. It is found that $w_b^{max}=0.70$ for $\psi=0$ and this occurs at $\lambda=1.05$. The corresponding figure for $\psi=\pi$ is 0.55 and occurs at $\lambda=2$. It is clear that in contrast to the

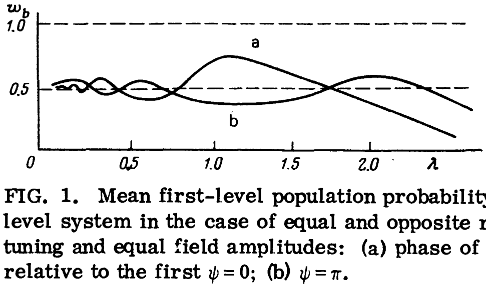

FIG. 1. Mean first-level population probability w_b for a two-level system in the case of equal and opposite resonant detuning and equal field amplitudes: (a) phase of second field relative to the first $\psi=0$; (b) $\psi=\pi$.

case of the single field, the upper-level population in the presence of two resonance fields may exceed 50%, and may even reach 70% when $\psi=0$. This result refers to the case of a fixed phase difference between the two acting fields. In the case of noncoherent fields, we can average over the phase ψ if we assume that all the phase values are equally probable. We then obtain $\langle w_b\rangle = \frac{1}{2}[1 - J_0^2(4/\lambda)]$ which is always less than $\frac{1}{2}$, just as in the case of a single field. It is, however, important to note that, both in the coherent and noncoherent cases, an increase in the fields is accompanied by a tendency of the population probability toward $\frac{1}{2}$ for each of the two levels and, in contrast to the single resonance field, this occurs nonmonotonically.

We note that the problem can be solved just as simply for any even number of resonance fields that are equal in pairs and have symmetric dispositions of the resonance detuning relative to the level separation (but not necessarily equal for different pairs). Instead of (3), the mean population w_b is then given by (to be specific, we take all phases $\psi_i=0$):

$$w_b=\frac{1}{2}-\frac{1}{2}\left\langle \cos\left\{\sum_i \frac{4V_i}{\omega_{2i}-\omega_{1i}}\sin\frac{(\omega_{2i}-\omega_{1i})t}{2}\right\}\right\rangle .$$

Since the number of parameters is large, it is not clear whether still higher population inversion can be achieved by a suitable choice of these parameters as compared with the case of the bichromatic field.

Returning now to the bichromatic field, we can introduce a real factor $f(t)$, representing the turning-on of both fields: $Vf(t)\cos\omega t$. This problem can also be solved: instead of (2), we obtain an analogous formula in which (for example, for $\psi=0$)

$$\frac{2}{\lambda}\sin\lambda\varphi\to 2\int_0^{\varphi}\cos\lambda\varphi f(\varphi)\,d\varphi.$$

We note that, for the bichromatic field with $x=\psi=0$, the above problem is equivalent to that of a resonance monochromatic field with slowly-varying complex amplitude $1+\exp(-2i\Delta t)$. Several workers (see, for example, Ref. 9) use the resonance approximation to consider the effect of a monochromatic field with amplitude varying slowly with time, $Vf(t)e^{i\Delta t}$, on a two-level system (Δ is the resonance detuning relative to the external field). The set of equations was solved exactly for $\Delta=0$.

§4. THE CASE OF SEPARATED RESONANCES ($\lambda \gg 1$)

When the frequency difference is large, only one of the fields is in resonance with the atomic transition and the curve representing w_b as a function of x is a superposition of two resonance Rabi curves. The approximate solution is

$$w_b=\frac{1}{2}\left[\frac{1}{1+(x+1)^2\lambda^2/4}+\frac{1}{1+(x-1)^2\lambda^2/4\alpha^2}\right]. \tag{4}$$

The resonance half-width for $x=-1$ is $2/\lambda \ll 1$, whereas the resonance half-width for $x=1$ is $2\alpha/\lambda\ll 1$. The phase ψ is unimportant in this case.

When $\lambda\gg 1$, the presence of the two different frequencies ω_1 and ω_2 leads to resonance not only at these two frequencies but also at the combination frequencies of the form[6] $n\omega_1-(n-1)\omega_2$ or $n\omega_2-(n-1)\omega_1$. These are generated by multiphoton matrix elements. For example, the resonance at the combination frequency $2\omega_1-\omega_2$ is generated by the three-photon matrix element shown graphically[3] in Fig. 2. This leads to narrow satellite lines on the graph of w_b as a function of x, in addition to the peaks given by (4). The shape of the peak near $x=-3$ is described by the Breit-Wigner curve with $w_b^{\max}=\frac{1}{2}$, corresponding to the resonance at frequency $\omega_{ba}=2\omega_1-\omega_2$. If we elevate the matrix element (see Fig. 2), we find that the half-width of the three-photon Raman resonance is $\alpha/2\lambda^3\ll 1$, i.e., it is small in comparison with the half-widths of the single-photon resonances since, by assumption, $\lambda\gg 1$.

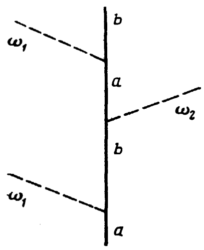

FIG. 2. Graph for the three-photon matrix element associated with resonance at the combination frequency $2\omega_1-\omega_2$.

Similarly, for the peak near $x=3$, which corresponds to resonance at the frequency $\omega_{ba}=2\omega_2-\omega_1$, we find that the half-width is $\alpha^2/2\lambda^3\ll 1$. The half-widths for the higher, i.e., five-photon, Raman-type resonances, are as follows: for $\omega_{ba}=3\omega_1-2\omega_2(x=-5)$, the half-width is $\alpha^2/(2\lambda)^5$ whereas, for $\omega_{ba}=3\omega_2-2\omega_1(x=5)$, the half-width is $\alpha^3/(2\lambda)^5$. The shape of these resonances is again described by the Breit-Wigner curve similar to that indicated above. The width of the five-photon resonances is small in comparison with the width of the three-photon resonances, and even more so in comparison with the width of the single-photon resonances.

Let us now determine the resonance Stark shifts of the resonance peaks. In the case of two fields, the frequencies ω_1 and ω_2 in the approximation which we are using ($\lambda\gg 1$) are so different that the fields V (frequency ω_1) and αV (frequency ω_2) produce line shifts that can be separated:

$$\Delta\omega_{ba}=\frac{V^2}{2(\omega_{ba}-\omega_1)}+\frac{\alpha^2V^2}{2(\omega_{ba}-\omega_2)}. \tag{5}$$

Thus, firstly, the positions of the single-photon resonances are shifted because of the interaction between them. These shifts are described by the first term in (5) for $x=1$ and by the second term for $x=-1$. The true positions of the peaks are, therefore, $x=-1+\alpha^2/\lambda^2$ and $x=1-1/\lambda^2$. As λ is reduced, the two single-photon resonances approach one another.

Secondly, the three-photon resonance is not exactly at $\omega_{ba}=2\omega_1-\omega_2$, i.e., for $x=-3$, but is shifted toward the single-photon resonance: $x=-3+(2+\alpha^2)/2\lambda^2$. This shift increases as the magnitude of λ is reduced. Similarly, for resonance at $\omega_{ba}=2\omega_2-\omega_1$, we have $x=3-(1+2\alpha^2)/2\lambda^2$. This is also shifted toward the $x=1$

single photon resonance as λ is reduced.

Finally, the shifts of the five-photon resonances are $x=-5+(3+2\alpha^2)/6\lambda^2$ and $x=5-(2+3\alpha^2)/6\lambda^2$. We note that, in contrast to the widths, the one-, three-, and five-photon resonance shifts are of the same order.

The quantity $|C_b(t)|^2$ exhibits characteristic beats at frequencies corresponding to the Raman resonances. For example, for the resonance at frequency $\omega_{ba}=2\omega_2-\omega_1$, we have the following combination of Rabi solutions for $x=3$, which corresponds to exact resonance (for simplicity, we again take $\psi=0$)

$$|C_b(t)|^2=\sin^2(\alpha\varphi/4\lambda^2)+(\alpha^2/\lambda^2)\sin^2\lambda\varphi+(1/4\lambda^2)\sin^2 2\lambda\varphi.$$

Since $\lambda\gg 1$, the graph of this expression takes the form of rapid oscillations of small amplitude (second and third terms) on the background of the slowly-varying mean value (first term). The beat frequencies and the fundamentals are in the ratio of, roughly, λ^3/α^2.

Similarly, for the five-photon resonance at $\omega_{ba}=3\omega_2-2\omega_1$, we have for $x=5$, which corresponds to exact resonance:

$$|C_b(t)|^2=\sin^2(\alpha^3\varphi/64\lambda^4)+(\alpha^2/4\lambda^2)\sin^2 2\lambda\varphi+(1/9\lambda^2)\sin^2 3\lambda\varphi.$$

The contribution of three-photon terms to this expression can be neglected because they are of the order of $1/\lambda^6$. The beat and fundamental frequencies are now in still greater ratio, namely, λ^5/α^3.

§5. THE CASE OF CLOSE FREQUENCIES ($\lambda \ll 1$)

Consider the situation opposite to that investigated in the last section, i.e., the case when $\lambda\ll 1$. The amplitudes C_a, C_b can now be obtained in the quasiclassical approximation. To show this, let $y\equiv\lambda\varphi$ and let us write (1) in the following form, assuming $\psi=0$, to be specific:

$$idC_a/dy=(1/\lambda)fC_b,\quad idC_b/dy=(1/\lambda)f^*C_a. \qquad (6)$$

In this expression:

$$f=\exp[-i(x+1)y]+\alpha\exp[-i(x-1)y].$$

Eliminating C_b from (6) and substituting $C_a=B_af^{1/2}$, we obtain the following expression for B_b which is valid in the quasiclassical approximation:

$$d^2B_a/dy^2+(1/\lambda^2)|f|^2B_a=0,$$

where $|f|^2=1+2\alpha\cos 2y+\alpha^2$. The solution of this equation for C_a and C_b is [subject to the initial condition $C_a(0)=1$]

$$C_a=\cos\left[\frac{1}{\lambda}\int_0^y |f|dy\right],$$

$$C_b=i\sin\left[\frac{1}{\lambda}\int_0^y |f|dy\right].$$

We see that there is no dependence on x for this particular range of variation of $x\lesssim 1$. Next, when $\alpha=1$, this yields the results of Sec. 3, as expected. Similarly, when $\alpha=0$, we obtain the Rabi solution for the strong field.

The mean populations of the upper and lower levels are close to $\frac{1}{2}$, as expected, because of the condition $\lambda\ll 1$. Thus, in this case, the two fields may be looked upon as one, with the corresponding amplitude (at least from the population point of view).

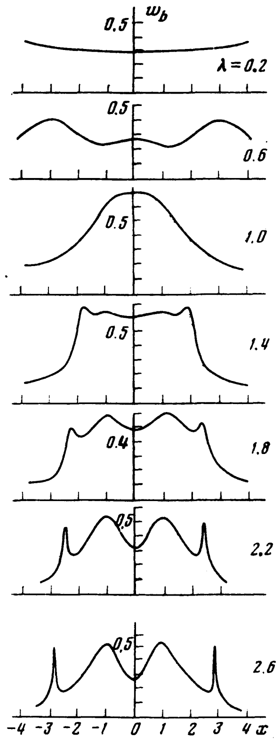

FIG. 3. Mean upper-level population probability w_b as a function of the dimensionless detuning from resonance, x, for the intermediate case $\lambda\sim 1$. In all cases, $\alpha=1$ and $\psi=0$.

§6. THE CASE $\lambda\sim 1$

The solution of (1) for intermediate $\lambda\sim 1$ can only be found numerically. Figure 3 shows the results of a numerical calculation of the mean upper-level population w_b as a function of x for $\alpha=1$, $\psi=0$, and different values of λ. It is clear that the oscillating values of w_b for $x=0$ (the origin of coordinates in all the graphs of Fig. 3) are in agreement with those shown in Fig. 1. Since we have taken $\alpha=1$, the graphs are symmetric relative to the origin. The graphs show an overall tendency toward the "collapse" of one or more photon resonances as λ decreases. For $\lambda\lesssim 1$, we have confirmation of the result given in Sec. 5, namely, that the population w_b is a slowly-varying function of x for $|x|\lesssim 1$.

Numerical calculations of w_b for $\lambda\sim 1$ have also been reported by Guccione-Gush and Gush.[6] However, because of the errors mentioned above, these authors have concluded that $w_b\le\frac{1}{2}$ always, whereas it is clear from Figs. 1 and 3 that $w_b>\frac{1}{2}$ for certain values of the parameters.

We may thus conclude that, in contrast to the monochromatic field, the bichromatic resonance field can produce population inversion in the two-level system if the parameters are suitably chosen. We note in this connection that there is no population inversion when two resonance fields act on two different transitions in the three-level system.[10]

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Superradiance in Raman scattering of light

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

Moscow State University
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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.

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1. INTRODUCTION

The effect of collective spontaneous emission of a sum of two-level atoms (the Dicke superradiance[1]) was by now investigated quite fully both theoretically[1–6] 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 class[12] 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-

