QUANTUM BEATS AND SUPERRADIANT EFFECTS IN THE SPONTANEOUS EMISSION FROM TWO NONIDENTICAL ATOMS*

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The problem considered is that of the spontaneous emission from two nonidentical two-level atoms coupled to a continuum of quantized electromagnetic modes. The atoms are separated by distances comparable to the resonant wavelength and have different transition frequencies and natural linewidths. Correlation functions and radiation rates are expressed in terms of expectation values of time-dependent atomic operators. The radiation pattern, total radiation rate and spectral distribution of radiation are obtained with the initial conditions that only one atom is excited and that the system is fully inverted. We find that the radiation pattern and total radiation rate show quantum beats when initially only one atom is excited. Moreover, the total radiation rate for strong interatomic interaction becomes greater than its initial value at the beginning of the emission process. This "superradiant" property is absent for two identical atoms. For initially fully inverted system, the radiation pattern and total radiation rate decay monotonically in time. Some weak beats can appear for drastically different atoms. The spectrum of radiation calculated for the case of strong interatomic interaction, i.e., for separations much smaller than the resonant wavelength shows two peaks, located at frequency $\pm \Omega_{12}$, contrary to the case of identical atoms, when the spectrum consists of only one peak located at the frequency $\pm \Omega_{12}$.

1. Introduction

The concept of collective spontaneous emission, or superradiance, has been the subject of a very great number of theoretical papers[†] since the pioneering article of Dicke¹), who showed that the influence on each atomic dipole of the electromagnetic field produced by the other atomic dipoles could, in certain circumstances, cause each atom to release its energy of excitation more rapidly than it would on its own. The shortening of the atomic lifetime resulting from the interaction of N atoms via the electromagnetic field can generally involve an enhancement of the intensity of radiation up to as much N^2I_1 , with I_1 : the

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[†] For a survey of publications, see the recent review article of Gross and Haroche²).

single atom intensity. This phenomenon has also been extensively studied in experiment³⁻⁷) during the last fifteen years, after the development of ultrashort laser systems made it easy to excite in a very short time a collection of atoms into a well defined electronic level.

The interest in superradiance studies lies in its close connection with the quantum and classical as well as with the spontaneous and stimulated aspects of atomic emission. Another intrinsic feature of superradiance is that the emitted field which propagates along an initially inverted medium exhibits strong nonlinear behaviour. The phenomenon of superradiance is, in general, characteristic for macroscopic systems with a large number of emitting atoms. Some papers have been devoted to the study of this phenomenon in the case of several atoms. Although the several (two or three) atom system is admittedly an elementary model, it offers some advantages over the multiatom problem. Because of its simplicity, one obtains detailed and almost exact dynamical solutions with a variety of initial conditions. Many of these results are analogous to phenomena that one would expect in multiatom systems. For example, the nonexponential decay law and simultaneous radiation at two frequencies are elementary examples of superradiant pulse formation and interaction broadening, respectively.

Coffey and Friedberg⁸) and Richter⁹) have shown that for some special configurations of a three atom system the total radiation intensity* can be greater than its initial value. This superradiant emission is due to the radiative as well as the dipole–dipole near-field interactions. Recently, Blank et al.¹⁰), ignoring dipole–dipole interaction, have shown that this effect for atoms located in an equidistant linear chain appears starting from six atoms.

Collective radiation from a system of three identical atoms for all possible geometrical configurations and for N atoms radiating in cascade process has been recently studied by Freedhoff¹¹). Some modifications in collective radiations arising from omission of the rotating wave approximation (RWA) have been discussed by Seke¹²).

Numerous papers have dealt with the spontaneous emission from a two atom system^{13–18}). In all the above discussed papers it is assumed that each atom cooperating in the emission process has the same transition frequency. In this case the total radiation rate is always lower than its initial value, meaning that superradiance is not present.

The formalisms developed ignore inhomogeneous spectral broadening, which is often very important, for example in the observation of superradiant effects in solids¹⁹⁻²¹) as well as for moving atoms^{22,23}). The influence of

^{*} In the literature, many authors use the term total radiation intensity instead of total radiation rate. Both terms are correct and will be used in our paper equivalently.

inhomogeneous broadening on radiation by macroscopic atomic systems has been discussed by Eberly²⁴), Agarwal²⁵) and Jodoin and Mandel²⁶). They have shown that the effect of inhomogeneous broadening generally reduces the amplitude of the superradiant pulse. Coffey and Friedberg⁸) considered the effect of inhomogeneous broadening on the total radiation intensity for spontaneous emission from two and three initially excited atoms and have shown that in this case the total radiation intensity exhibits no superradiance.

Varfolomeev²⁷) considered spontaneous decay probability amplitudes for two nonidentical atoms with one atom initially excited and has shown them to exhibit sinusoidal modulation (beats). This sinusoidal modulation of probability amplitudes also has been found by Milonni and Knight²⁸), who considered the effect of all the retardation times on various spontaneous decay probability amplitudes.

In this paper we show that superradiant effect and quantum beats can occur in the spontaneous emission from two nonidentical two-level atoms. We assume that the two two-level atoms have different transition frequencies and different natural linewidths. The methods and results presented here differ considerably from those of earlier treatments by Varfolomeev²⁷) and Milonni and Knight²⁸); in particular, the emphasis is on time-dependent decay rates and correlation functions rather than on quantities such as the probability amplitude for finding a particular set of photon states. Although these admittedly convey less information than probability amplitudes, they are usually the quantities of most direct physical interest, and are easier to calculate.

We start from the Hamiltonian for a system of N nonidentical atoms, interacting with the electromagnetic field via electric dipole interaction, and derive general equations of motion for the atomic operators. We then apply them to the problem of spontaneous emission from two nonidentical atoms for two initial conditions: (a) only one atom excited, (b) both atoms excited. We study in detail the directional properties of the radiation, the total radiation intensity, and the spectral distribution of the radiation field. As is well known^{13–18}), the total radiation intensity for two identical atoms does not differ from the single-atom exponential decay. We shall see later, however, that the total radiation intensity emitted from two nonidentical atoms differs from the exponential decay and exhibits sinusoidal modulation (quantum beats) as well as superradiant effect.

2. Derivation of the equations of motion

We consider a collection of N nonidentical nonoverlapping atoms, coupled to a quantized multimode electromagnetic field, on the assumption that the atoms

have different transition frequencies $\omega_1 \neq \omega_2 \neq \cdots \neq \omega_N$ and different transition dipole moments $\mu_1 \neq \mu_2 \neq \cdots \neq \mu_N$. Each atom is approximated by a two-level system with the ground state $|1\rangle_i$ $(i = 1, 2, \ldots, N)$ and the excited state $|2\rangle_i$, connected by an electric dipole transition μ_i .

In the electric dipole approximation the Hamiltonian of this system has the form

$$H = \sum_{i=1}^{N} \left\{ \hbar \omega_{i} R_{i}^{z}(t) - \boldsymbol{\mu}_{i} \cdot \boldsymbol{E}(\boldsymbol{r}_{i}, t) [R_{i}^{+}(t) + R_{i}^{-}(t)] \right\}$$

$$+ \sum_{k\lambda} \hbar \omega_{k} a_{k\lambda}^{+}(t) a_{k\lambda}(t) , \qquad (1)$$

where $R_i^+(t)$ and $R_i^-(t) = [R_i^+(t)]^+$ are operators raising and lowering the energy of atom i, and R_i^z describes its energy. These operators fulfil the well-known commutation relations

$$[R_i^+, R_j^-] = 2R_i^z \delta_{ij}, \quad [R_i^z, R_j^{\pm}] = \pm R_i^{\pm} \delta_{ij}.$$
 (2)

If the atom *i* is initially in its ground state $|1\rangle_i$ then $\langle R_i^z \rangle = -1/2$, whereas $\langle R_i^z \rangle = +1/2$ if the atom is in its excited state.

The field operator $E(r_i, t)$ in the transverse mode decomposition is given by

$$E(\mathbf{r}_i, t) = i \sum_{k\lambda} \left(\frac{2\pi\hbar \omega_k}{V} \right)^{1/2} e_{k\lambda} [a_{k\lambda}(t) e^{i\mathbf{k}\cdot\mathbf{r}_i} - \text{h.c.}], \qquad (3)$$

where V is the normalization volume, $e_{k\lambda}$ the unit polarisation vector, $a_{k\lambda}$ the annihilation operator for the kth mode with polarization λ , and r_i is the vector describing the position of the ith atom.

Inserting (3) into (1) we have

$$H = \sum_{i=1}^{N} \hbar \omega_{i} R_{i}^{z}(t) + \sum_{k\lambda} \hbar \omega_{k} a_{k\lambda}^{+}(t) a_{k\lambda}(t) - i \sum_{i=1}^{N} \sum_{k\lambda} \left[g_{k\lambda}(\mathbf{r}_{i}) a_{k\lambda}(t) - \text{h.c.} \right] \left[R_{i}^{+}(t) + R_{i}^{-}(t) \right],$$
(4)

with

$$g_{k\lambda}(\mathbf{r}_i) = \left(\frac{2\pi\hbar\omega_k}{V}\right)^{1/2}\boldsymbol{\mu}_i \cdot \boldsymbol{e}_{k\lambda} e^{i\mathbf{k}\cdot\mathbf{r}_i}.$$
 (4a)

The Heisenberg equation of motion for the atomic operator $R_i^-(t)$ now takes the form (a dot denotes d/dt)

$$\dot{R}_{i}^{-}(t) = -i\omega_{i}R_{i}^{-}(t) + 2\sum_{k\lambda} [g_{k\lambda}(r_{i})a_{k\lambda}(t) - \text{h.c.}]R_{i}^{z}(t), \qquad (5)$$

while the corresponding equation of motion for the radiation field operators has the form:

$$\dot{a}_{k\lambda}(t) = -i\omega_k a_{k\lambda}(t) + \sum_{i=1}^{N} g_{k\lambda}^*(r_i) [R_i^+(t) + R_i^-(t)].$$
 (6)

The equation of motion for the operator $R_i^+(t)$ is Hermitian conjugate to eq. (5). Moreover, the equation of motion for the operator $R_i^2(t)$ can be obtained from (5) using the relation

$$\dot{R}_{i}^{z}(t) = \frac{1}{2} \{ [\dot{R}_{i}^{+}(t), R_{i}^{-}(t)] + [R_{i}^{+}(t), \dot{R}_{i}^{-}(t)] \}.$$

Eq. (6) may be integrated formally and its solution is:

$$a_{k\lambda}(t) = a_{\text{free}}(t) + \sum_{i=1}^{N} g_{k\lambda}(\mathbf{r}_i) \int_{0}^{t} dt' \left[R_i^{+}(t') + R_i^{-}(t') \right] e^{-i\omega_k(t-t')},$$
 (7)

where $a_{\text{free}}(t) = a_{k\lambda}(0) \exp(-i\omega_k t)$ is the solution of the homogeneous or free-field equation that characterizes the field without sources (atoms).

Eqs. (5) and (7) cannot be solved explicitly. However, as far as atoms are concerned, spontaneous radiative decay is a very slow process, requiring on the average many millions of cycles of dipole oscillations before it is completed. Thus, we assume that $R_i^{\pm}(t)$ may be written as $S_i^{\pm}(t) \exp(\pm i\omega_0 t)$, where $S_i^{\pm}(t)$ is an unknown operator whose time variation, compared with $\exp(\pm i\omega_0 t)$, is very slow, and $\omega_0 = (1/N) \Sigma_i \omega_i$.

Then the second right-hand term in (7), called the "source part" of the quantized field, may only be approximated replacing $S_i^{\pm}(t')$ by $S_i^{\pm}(t)$ and extracting it from the integral. The integral can then be obtained with the following two approximations: the Born approximation, assuming weak coupling between the atom and the field, and the Markoff approximative assumption that the time required for the light signal to traverse the system is small in comparison to the time required for appreciable changes in population of the atomic levels, i.e.

$$(r_{ij})_{\max} \ll c \, \Delta t \,. \tag{8}$$

With these two approximations and for sufficiently long time $t \ge (1/\omega_0)$, the right-hand term of (7) may be expressed by way of $S^{\pm}(t)$ and the function $\zeta(x)$, defined as²⁹)

$$i\zeta(x) = \lim_{t \to \infty} \int_{0}^{t} d\tau e^{ix\tau} = i \frac{P}{x} + \pi \delta(x) , \qquad (9)$$

where P indicates the principal value of the integral.

On these approximations eq. (7) takes the form

$$a_{k\lambda}(t) = a_{\text{free}}(t) + \sum_{i=1}^{N} g_{k\lambda}^{*}(\mathbf{r}_{i})[S_{i}^{+}(t)J_{+} + S_{i}^{-}(t)J_{-}], \qquad (10)$$

with

$$J_{\pm} = \pi \delta(\omega_k \pm \omega_0) - iP\left(\frac{1}{\omega_k \pm \omega_0}\right). \tag{10a}$$

Substituting these results into (5) and taking normal ordering of the radiation and atomic operators [thus, $a_{k\lambda}(t)$ to the right of $S_i^+(t)$], we have

$$\dot{S}_{i}^{-}(t) = -i\omega_{i}S_{i}^{-}(t) + 2\sum_{k\lambda} \left[g_{k\lambda}(\mathbf{r}_{i})S_{i}^{z}(t)a_{\text{free}}(t) - g_{k\lambda}^{*}(\mathbf{r}_{i})a_{\text{free}}^{+}(t)S_{i}^{z}(t) \right]$$

$$+ \sum_{k\lambda} |g_{k\lambda}(\mathbf{r}_{i})|^{2} \left[(J_{+} + J_{-}^{*})S_{i}^{-}(t) - \text{h.c.} \right]$$

$$+ 2\sum_{k\lambda} \sum_{i \neq j} \left\{ \left[g_{k\lambda}(\mathbf{r}_{i})g_{k\lambda}^{*}(\mathbf{r}_{j})J_{+} - g_{k\lambda}^{*}(\mathbf{r}_{i})g_{k\lambda}(\mathbf{r}_{i})J_{-}^{*} \right] S_{i}^{z}(t)S_{j}^{+}(t) - \text{h.c.} \right\}.$$

$$(11)$$

If we now consider all modes of the radiation field available for spontaneous emission and go over to the free-space continuum limit $V \rightarrow \infty$, so that

$$\sum_{k} \cdots \rightarrow \frac{V}{(2\pi)^3} \int dk \sum_{k=1}^e \cdots, \tag{12}$$

we obtain from eq. (11) (with the rotating wave approximation):

$$\dot{S}_{i}^{-}(t) = -(\gamma_{i} - i\Delta)S_{i}^{-}(t) + 2\sum_{k\lambda} g_{k\lambda}(\mathbf{r}_{i})S_{i}^{z}(t)a_{\text{free}}(t) + \sum_{i \neq j} 2(\gamma_{ij} + i\Omega_{ij})S_{i}^{z}(t)S_{j}^{-}(t).$$

$$(13)$$

Here, $\gamma_i = 2\mu_i^2 \omega_0^3 / 3\hbar c^3$ is one-half the Einstein A coefficient for the *i*th atom, $\Delta = \omega_0 - \omega_i$, and

$$\gamma_{ij} = \sqrt{\gamma_i \gamma_j} a_{ij} = \frac{3}{2} \sqrt{\gamma_i \gamma_j} \left\{ \left[(\hat{\mu}_i \cdot \hat{\mu}_j) - (\hat{\mu}_i \cdot \hat{r}_{ij}) (\hat{\mu}_j \cdot \hat{r}_{ij}) \right] \frac{\sin k r_{ij}}{k r_{ij}} + \left[(\hat{\mu}_i \cdot \hat{\mu}_j) - 3(\hat{\mu}_i \cdot \hat{r}_{ij}) (\hat{\mu}_j \cdot \hat{r}_{ij}) \right] \left[\frac{\cos k r_{ij}}{(k r_{ij})^2} - \frac{\sin k r_{ij}}{(k r_{ij})^3} \right] \right\},$$

$$(14)$$

$$\Omega_{ij} = \sqrt{\gamma_i \gamma_j} b_{ij} = \frac{3}{2} \sqrt{\gamma_i \gamma_j} \left\{ -\left[(\hat{\mu}_i \cdot \hat{\mu}_j) - (\hat{\mu}_i \cdot \hat{r}_{ij}) (\hat{\mu}_j \cdot \hat{r}_{ij}) \right] \frac{\cos k r_{ij}}{k r_{ij}} + \left[(\hat{\mu}_i \cdot \hat{\mu}_j) - 3(\hat{\mu}_i \cdot \hat{r}_{ij}) (\hat{\mu}_j \cdot \hat{r}_{ij}) \right] \left[\frac{\sin k r_{ij}}{(k r_{ij})^2} + \frac{\cos k r_{ij}}{(k r_{ij})^3} \right] \right\}, \tag{15}$$

where $\hat{\mu}_i$ and \hat{r}_{ij} are unit vectors along the transition electric dipole moment and the vector $\mathbf{r}_{ij} = \mathbf{r}_j - \mathbf{r}_i$ respectively. Moreover, $r_{ij} = |\mathbf{r}_{ij}|$ and $k = \omega_0/c = 2\pi/\lambda_0$. When deriving eq. (13) we neglected all single-atom radiative level shifts as irrelevant for our purposes. The parameters (14) and (15), which both depend on the interatomic distance r_{ij} , describe collective damping and collective shift of energy levels and determine the collective properties of our system of N nonidentical atoms. In fig. 1, b_{ij} and a_{ij} , defined in (14) and (15), are plotted for the cases when $\hat{\mu}$ is parallel and perpendicular to \hat{r}_{ij} . Both parameters b_{ij} and a_{ij} strongly depend on the interatomic distance r_{ij} and on the spatial orientation of the dipole moments.

For $kr_{ii} \ll 1$ we obtain

$$a_{ii} \approx 1 , \quad b_{ii} \approx [(\hat{\mu}_i \cdot \hat{\mu}_i) - 3(\hat{\mu}_i \cdot \hat{r}_{ii})(\hat{\mu}_i \cdot \hat{r}_{ii})]r_{ii}^{-3} .$$
 (16)

For this case we see that a_{ij} reduces to unity, and b_{ij} reduces to the static dipole-dipole interaction.

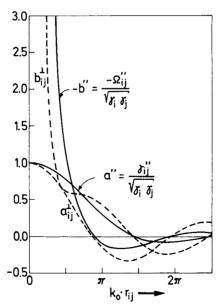


Fig. 1. The damping constants γ_{ij} and frequency shifts Ω_{ij} versus the interatomic separations \mathbf{r}_{ij} for dipole matrix elements $\hat{\mu}_i$ parallel and perpendicular to \mathbf{r}_{ij} .

Eq. (13) is fundamental for the theory of radiation of nonidentical two-level atoms. For identical atoms, it is the same as those obtained by Lehmberg¹³) and Agarwal¹⁴). Beside the parameters describing the nonidenticity of the atomic system it depends also on the collective parameters γ_{ij} and Ω_{ij} , which describe the collective properties of our system of N nonidentical atoms. We apply eq. (13) to derive the directional properties of the radiation, the total radiation intensity, and the spectral distribution of the radiation.

When deriving eq. (13) we omitted rapidly oscillating terms with the frequency $2\omega_0$ (so-called counter-rotating terms) using the rotating wave approximation (RWA). As shown by Seke¹²), the counter-rotating terms play a significant role for some processes with times shorter than spontaneous emission. In this paper, we shall consider new phenomena like quantum beats effects, which are slow processes not caused by the counter-rotating terms. The role of the rotating wave approximation in the spontaneous emission from two identical atoms has been discussed by Milonni and Knight¹⁸).

3. Calculation of the radiation intensity

We now calculate the radiation intensity $I(\mathbf{R}, t)$, which is proportional to the normally ordered one-time correlation function of the electromagnetic field at a point $\mathbf{R} = R\hat{R}$ in the wave zone:

$$I(\mathbf{R},t) = \frac{R^2 c}{2\pi\omega_0} \left\langle \mathbf{E}^{(-)}(\mathbf{R},t) \cdot \mathbf{E}^{(+)}(\mathbf{R},t) \right\rangle. \tag{17}$$

Here, we have introduced the factor $(R^2c/2\pi\omega_0)$ so that $I(\mathbf{R},t)\,\mathrm{d}\Omega\,\mathrm{d}t$ is now the probability of finding one photon inside the solid angle element $\mathrm{d}\Omega$ around the direction \hat{R} in the time interval $\mathrm{d}t$ at the moment of time t in the far field zone of the radiation emitted by the atomic system.

The field operator is given by the well known expression

$$E(R, t) = E^{(+)}(R, t) + E^{(-)}(R, t),$$
(18)

where $E^{(+)}$ ($E^{(-)}$) denotes the positive (negative) frequency part of the electromagnetic field and, in the transverse mode decomposition, is given by

$$\boldsymbol{E}^{(+)}(\boldsymbol{R}, t_R) = \mathrm{i} \sum_{k\lambda} \left(\frac{2\pi\hbar \omega_k}{V} \right)^{1/2} \boldsymbol{e}_{k\lambda} a_{k\lambda}(t) \, \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}} \,, \tag{19}$$

with $t_R = t + (R/c)$ and the same parameters as in eq. (3).

On insertion of eq. (10) into (19) and applying the relation (12) we obtain

$$\boldsymbol{E}^{(+)}(\boldsymbol{R}, t_R) = \boldsymbol{E}_0^{(+)}(\boldsymbol{R}, t_R) + \frac{\mathrm{i}}{4\pi^2 c^3} \sum_{i=1}^N \int_0^\infty \omega_k^3 \, \mathrm{d}\omega_k \oint \mathrm{d}\Omega_k \left[\boldsymbol{\mu}_i - \hat{k}(\hat{k} \cdot \boldsymbol{\mu}_i) \right] \\ \times \exp\left(\mathrm{i} \frac{\omega_k}{c} \, \hat{k} \cdot \boldsymbol{R}_i\right) \left[S_i^+(t_R) J_+ + S_i^-(t_R) J_- \right], \tag{20}$$

where \hat{k} denotes the unit vector in the direction k and $E_0(R, t)$ denotes the positive frequency part of the vacuum field. The only important contributions come from those directions around $\hat{k} = \pm \hat{R}_i$ where the phase $\pm \omega_k R_i/c$ is stationary; hence, one can replace $[\mu_i - \hat{k}(\hat{k} \cdot \mu_i)]$ by $[\mu_i - \hat{R}_i(\hat{R}_i \cdot \mu_i)]$ and remove it from the integral over $d\Omega_k$. On integration over $d\Omega_k$ in (20) we obtain

$$E^{(+)}(\mathbf{R}, t_R) = E_0^{(+)}(\mathbf{R}, t_R) + \frac{1}{2\pi c^2} \sum_{i=1}^{N} \int_{0}^{\infty} \omega_k^2 d\omega_k \frac{[\boldsymbol{\mu}_i - \hat{R}_i(\hat{R}_i \cdot \boldsymbol{\mu}_i)]}{R_i}$$

$$\times \exp\left(i \frac{\omega_k}{c} R_i\right) [S_i^+(t_R) J_+ + S_i^-(t_R) J_-]. \tag{21}$$

Since the atomic operators $S_i^{\pm}(t)$ oscillate as $\exp(\pm i\omega_0 t)$ we write

$$S_i^-(t_R) \approx S_i^-(t) e^{-ikR}$$
 (22)

Thus, substituting (22) into (21) and then evaluating the integral, we obtain

$$E^{(+)}(\mathbf{R}, t_R) = E_0^{(+)}(\mathbf{R}, t_R) - \left(\frac{\omega_0}{c}\right)^2 \sum_{i=1}^N \frac{\left[\hat{R}_i \times (\hat{R}_i \times \boldsymbol{\mu}_i)\right]}{R_i} S_i^-(t) e^{ik(R_i - R)}.$$
 (23)

The term with $S_i^+(t)$ contributes nothing to $E^{(+)}(R, t)$ because integration in (21) extends over positive ω_k and J_- is the only contribution to the integral. If R lies in the far field zone of the radiation emitted by the atomic system, i.e. $R \gg (r_{ij})_{\max}$, λ_0 , so that $R_i \approx R - \hat{R} \cdot r_i$, then (23) becomes

$$E^{(+)}(\mathbf{R}, t_R) = E_0^{(+)}(\mathbf{R}, t_R) - \left(\frac{\omega_0}{c}\right)^2 \sum_{i=1}^N \frac{[\hat{R}_i \times (\hat{R}_i \times \boldsymbol{\mu}_i)]}{R_i} S_i^-(t) \exp(-ik\hat{R} \cdot \boldsymbol{r}_i).$$
 (24)

Hence, for the angular and time distribution of radiation intensity $I(\mathbf{R}, t)$ we

have the following expression:

$$I(\mathbf{R}, t) = I_0(\mathbf{R}, t) + u(\hat{R}) \sum_{i,j=1}^{N} 2(\gamma_i \gamma_j)^{1/2} \langle S_i^+(t) S_j^-(t) \rangle e^{ik\hat{R} \cdot \mathbf{r}_{ij}}, \qquad (25)$$

where the average is taken over the initial states of the entire system, $u(\hat{R}) = (3/8\pi) \sin^2 \vartheta$, with ϑ the angle between the observation direction R and the atomic transition dipole moment μ .

On integrating over all solid angles $d\Omega_{\hat{R}}$, eq. (25) yields the total radiation rate I(t) given in photons per second as:

$$I(t) = I_0(t) + \sum_{i,j=1}^{N} 2\gamma_{ij} \langle S_i^+(t) S_j^-(t) \rangle , \qquad (26)$$

where

$$I_0(t) = \frac{R^2 c}{2\pi\omega_0} \int d\Omega_{\hat{R}} \left\langle E_0^{(-)}(\mathbf{R}, t) \cdot E_0^{(+)}(\mathbf{R}, t) \right\rangle , \qquad (27)$$

and γ_{ii} is given by eq. (14).

From eqs. (25) and (26) it is obvious that, in order to calculate the angular distribution of radiation and the total radiation rate from a system of N nonidentical atoms, it is necessary to have available the correlation functions for the atomic operators. These atomic correlation functions can be found using the equations of motion for $S_i^{\pm}(t)$ given by (13).

4. Spontaneous emission from two nonidentical atoms

4.1. General solutions for the atomic correlation functions

In the case when the radiation field is initially in the vacuum state $|\{0\}\rangle$, we have $E_0^{(+)}(\mathbf{R}, t)|\{0\}\rangle = 0$, and eqs. (13) lead to the vacuum expectation values for the atomic correlation functions.

For two nonidentical atoms (i = 1, 2), eqs. (13) lead to the following, closed set of five equations of motion for the vacuum expectation values

$$\langle S_1^+ S_1^- \rangle_t = -2\gamma_1 \langle S_1^+ S_1^- \rangle_t - \kappa \langle S_2^+ S_1^- \rangle_t - \kappa^* \langle S_1^+ S_2^- \rangle_t, \qquad (28a)$$

$$\langle S_{2}^{+}, S_{2}^{-} \rangle_{t} = -2\gamma_{2} \langle S_{2}^{+} S_{2}^{-} \rangle_{t} - \kappa \langle S_{1}^{+} S_{2}^{-} \rangle_{t} - \kappa^{*} \langle S_{2}^{+} S_{1}^{-} \rangle_{t}, \qquad (28b)$$

$$\langle S_{1}^{+} \dot{S}_{2}^{-} \rangle_{t} = -(\gamma_{1} + \gamma_{2} - 2i\Delta) \langle S_{1}^{+} S_{2}^{-} \rangle_{t} - \kappa \langle S_{1}^{+} S_{1}^{-} \rangle_{t} - \kappa^{*} \langle S_{2}^{+} S_{2}^{-} \rangle_{t} + 4\gamma_{12} \langle S_{1}^{+} S_{2}^{+} S_{1}^{-} S_{2}^{-} \rangle_{t},$$
 (28c)

$$\langle S_0^+, S_1^- \rangle_t = -(\gamma_1 + \gamma_2 + 2i\Delta) \langle S_2^+, S_1^- \rangle_t - \kappa \langle S_2^+, S_2^- \rangle_t - \kappa^* \langle S_1^+, S_1^- \rangle_t + 4\gamma_{12} \langle S_1^+, S_2^+, S_1^-, S_2^- \rangle_t,$$
 (28d)

$$\langle S_1^+ S_2^+ S_1^- S_2^- \rangle_t = -2(\gamma_1 + \gamma_2) \langle S_1^+ S_2^+ S_1^- S_2^- \rangle_t, \qquad (28e)$$

where $\kappa = (\gamma_{12} + i\Omega_{12})$.

It is obvious from the above equations that eq. (28e) is decoupled from the other four equations and has the simple solution

$$\langle S_1^+ S_2^+ S_1^- S_2^- \rangle_t = \langle S_1^+(0) S_2^+(0) S_1^-(0) S_2^-(0) \rangle e^{-2(\gamma_1 + \gamma_2)t}.$$
 (29)

The remaining set of four equations can be solved strictly. The system of four equations (28a)–(28d) can be transformed using the Laplace transform method into a system of algebraic equations in transformed variables, which is easy to solve. To obtain the time dependence, however, we have to know the roots of the fourth-order biquadratic secular equation for the system (28a)–(28d),

$$(s + \gamma_1 + \gamma_2)^4 + 4(\Delta^2 + \Omega_{12}^2 - u_{12}^2 - \gamma_{12}^2)(s + \gamma_1 + \gamma_2)^2 - 16(\gamma_{12}\Omega_{12} - \Delta u_{12})^2 = 0,$$
(30)

where $u_{12} = (1/2)(\gamma_2 - \gamma_1)$.

Putting

$$z = (s + \gamma_1 + \gamma_2)^2 \,, \tag{31}$$

eq. (30) reduces to a quadratic equation with the roots

$$z_{1,2} = -2(\Delta^2 + \Omega_{12}^2 - \gamma_{12}^2 - u_{12}^2)$$

$$\pm 2[(\Delta^2 + \Omega_{12}^2 + u_{12}^2 + \gamma_{12}^2)^2 - 4(\Delta\gamma_{12} + u_{12}\Omega_{12})^2]^{1/2}.$$
(32)

Since the $z_{1,2}$ are real, $z_1 > 0$ and $z_2 < 0$, we find for the biquadratic equation (30) two purely real roots

$$s_1 = -(\gamma_1 + \gamma_2) + \sqrt{z_1} , \qquad (33)$$

$$s_2 = -(\gamma_1 + \gamma_2) - \sqrt{z_1}, \qquad (34)$$

and two complex conjugate roots

$$s_3 = -(\gamma_1 + \gamma_2) + i\sqrt{-z_2}$$
, (35)

$$s_4 = -(\gamma_1 + \gamma_2) - i\sqrt{-z_2}. \tag{36}$$

Tedious though straightforward calculations lead to the following general solution of eqs. (28a)–(28d) for arbitrary initial conditions:

$$\langle S_{1}^{+} S_{1}^{-} \rangle_{t} = \frac{32 \gamma_{12} G_{0} [\gamma_{1} \gamma_{12} w + \Omega_{12} (\gamma_{1} \Delta + \gamma_{12} \Omega_{12})]}{(D^{2} - 4w^{2})(E^{2} + 4w^{2})} e^{-4wt} + \sum_{\substack{i=1\\i \neq i \neq k \neq l}}^{4} \frac{A_{+}(s_{i})}{(s_{i} + 4w)(s_{i} - s_{j})(s_{i} - s_{k})(s_{i} - s_{l})} e^{s_{i}t},$$
(37)

$$\langle S_{2}^{+} S_{2}^{-} \rangle_{t} = \frac{32 \gamma_{12} G_{0} [\gamma_{2} \gamma_{12} w + \Omega_{12} (-\gamma_{2} \Delta + \gamma_{12} \Omega_{12})]}{(D^{2} - 4w^{2})(E^{2} + 4w^{2})} e^{-4wt} + \sum_{\substack{i=1\\i \neq i \neq k \neq l}}^{4} \frac{A_{-}(s_{i})}{(s_{i} + 4w)(s_{i} - s_{j})(s_{i} - s_{k})(s_{i} - s_{l})} e^{s_{i}t},$$
(38)

$$\langle S_{1}^{+} S_{2}^{-} \rangle_{t} = \frac{32 \gamma_{12} G_{0} [(\Omega_{12}^{2} + w^{2} - u_{12}^{2})w - i(\gamma_{1} \gamma_{2} \Delta + u_{12} \gamma_{12} \Omega_{12})]}{(D^{2} - 4w^{2})(E^{2} + 4w^{2})} e^{-4wt} + \sum_{\substack{i=1\\i \neq i \neq k \neq l}}^{4} \frac{G(s_{i})}{(s_{i} + 4w)(s_{i} - s_{j})(s_{i} - s_{k})(s_{i} - s_{l})} e^{s_{i}t},$$
(39)

$$\langle S_2^+ S_1^- \rangle_t = (\langle S_1^+ S_2^- \rangle_t)^* , \qquad (40)$$

where we have introduced the notation:

$$A_{\pm}(s) = A_{0}(s+4w)[(s+2w)^{3} \pm 2u_{12}(s+2w)^{2} + 2(2\Delta^{2} + \Omega_{12}^{2} - \gamma_{12}^{2})(s+2w) \pm 8\Delta(\Delta u_{12} - \gamma_{12}\Omega_{12})] + 2D_{0}(\gamma_{12}^{2} + \Omega_{12}^{2})(s+4w)(s+2w) - 4\gamma_{12}G_{0}[2\gamma_{12}(s+2w)^{2} \pm 4(\gamma_{12}u_{12} - \Delta\Omega_{12})(s+2w) + 8\Omega_{12}(\gamma_{12}\Omega_{12} - \Delta u_{12})],$$
(41)

$$G(s) = -A_0 \kappa (s + 4w) [(s + 2w + 2u_{12})(s + 2w + 2i\Delta) - 4i\gamma_{12}\Omega_{12}]$$

$$-D_0 \kappa^* (s + 4w) [(s + 2w - 2u_{12})(s + 2w + 2i\Delta) + 4i\gamma_{12}\Omega_{12}]$$

$$+ 4\gamma_{12}G_0 [(s + 2w)^3 + 2i\Delta(s + 2w)^2 + 4(\Omega_{12}^2 - u_{12}^2)(s + 2w)$$

$$+ 8iu_{12}(\gamma_{12}\Omega_{12} - \Delta u_{12})], \qquad (42)$$

$$u_{12} = (1/2)(\gamma_2 - \gamma_1), \quad w = (1/2)(\gamma_1 + \gamma_2),$$
 (43)

$$B = \left[\left(\Delta^2 + \Omega_{12}^2 + u_{12}^2 + \gamma_{12}^2 \right)^2 - 4(\Delta \gamma_{12} + u_{12}\Omega_{12})^2 \right]^{1/2}, \tag{44}$$

$$D = \left[-2(\Delta^2 + \Omega_{12}^2 - u_{12}^2 - \gamma_{12}^2) + 2B \right]^{1/2}, \tag{45}$$

$$E = \left[2(\Delta^2 + \Omega_{12}^2 - u_{12}^2 - \gamma_{12}^2) + 2B\right]^{1/2},\tag{46}$$

with $A_0 = \langle S_1^+ S_1^- \rangle_0$, $D_0 = \langle S_2^+ S_2^- \rangle_0$, $G_0 = \langle S_1^+ S_2^+ S_1^- S_2^- \rangle_0$, describing the initial population of the excited states of our nonidentical atoms. If initially atom 1 is in its excited state and atom 2 is initially in its ground state, then $A_0 = 1$ and $D_0 = G_0 = 0$, where $A_0 = D_0 = G_0 = 1$ if both atoms are initially in their excited states.

The parameters (43)—(46) define the shape of the time evolution of the system. The parameter D, together with the overall damping factor $\exp[-(\gamma_1 + \gamma_2)t]$ describes the decay rates of the particular terms whereas parameter E describes the frequency of modulation that is superimposed on the composite exponential decay (the existence of this modulation arises from the fact that eq. (30) has two complex roots). If the atoms are far apart $(\gamma_{12} = 0, \Omega_{12} = 0)$ we have $D = \gamma_2 - \gamma_1$ and $E = \omega_2 - \omega_1$ and the atoms evolve independently. In the opposite limit when the atoms are close to each other (very strong interatomic interaction $\Omega_{12} \gg \sqrt{\gamma_1 \gamma_2}$), $D \approx 2\gamma_{12}$ and $E = 2\Omega_{12}$. In this case the evolution of the atoms is strongly correlated.

In fact, the two-atom system Hamiltonian can be diagonalized including the dipole–dipole interaction, giving the eigenstates: $|0\rangle=|1\rangle_1|1\rangle_2$, $|\pm\rangle=(1/\sqrt{2})(|2\rangle_1|1\rangle_2\pm|1\rangle_1|2\rangle_2$) and $|2\rangle=|2\rangle_1|2\rangle_2$, with energies $E_0=0$, $E_\pm=\hbar(\omega_0\pm\Omega_{12})$ and $E_2=2\hbar\omega_0$, respectively (see fig. 2a).

For nonidentical atoms, the states $|\pm\rangle$ (the superradiant and subradiant state) are no longer eigenstates of the two-atom system. The Hamiltonian of the system can then be rediagonalized including Δ giving the new eigenstates $|\varphi_{\pm}\rangle = C_1 |\pm\rangle \pm C_2 |\mp\rangle$ with the energies $E_{\pm} = \hbar(\omega_0 \pm \sqrt{\Delta^2 + \Omega_{12}^2})$ as shown in fig. 2b. The coefficients C_1 and C_2 are: $C_1 = \alpha/\sqrt{\alpha^2 + \Delta^2}$, $C_2 = -\Delta/\sqrt{\alpha^2 + \Delta^2}$, where $\alpha = \Omega_{12} + \sqrt{\Delta^2 + \Omega_{12}^2}$. For small interatomic separations r_{12} the subradiant state $|-\rangle$ is decoupled from the other states. Since both the states $|\varphi_{\pm}\rangle$ include the superradiant state $|+\rangle$, the transitions from both states to the ground state are allowed. Thus, one can expect quantum beats to appear in the spontaneous emission from such a system. This is the subject of the next section.

It is interesting to note that the solutions (37) and (38) for $D = \gamma_1 + \gamma_2$ in the steady state $(t \to \infty)$ are nonzero. This signifies that part of the energy has been trapped in the atomic system. This effect of population trapping has been discussed recently by us³⁰).

(50)

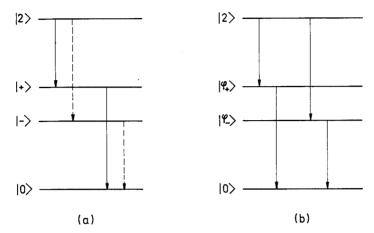


Fig. 2. Energy-levels diagrams for two-atom system with possible transitions, in the case of: (a) identical atoms, (b) nonidentical atoms.

4.2. Radiation pattern and total radiation intensity

4.2.1. Initially only one atom excited

 $\langle S_2^+ S_1^- \rangle_{\cdot} = (\langle S_1^+ S_2^- \rangle_{\cdot})^*$.

Assuming that initially only atom 1 is in its excited state and atom 2 is in the ground state, we have $A_0 = 1$, $D_0 = G_0 = 0$, and the general solutions (37)–(40) reduce to:

$$\langle S_{1}^{+}S_{1}^{-}\rangle_{t} = e^{-2wt} \left\{ \frac{1}{2} \left(1 - \frac{\Delta^{2} + u_{12}^{2}}{B} \right) \cos Et + \frac{1}{2} \left(1 + \frac{\Delta^{2} + u_{12}^{2}}{B} \right) \cosh Dt + \frac{u_{12}E^{2} - 4\Delta(\Delta u_{12} - \gamma_{12}\Omega_{12})}{2BE} \sin Et + \frac{u_{12}D^{2} + 4\Delta(\Delta u_{12} - \gamma_{12}\Omega_{12})}{2BD} \sinh Dt \right\},$$

$$\langle S_{2}^{+}S_{2}^{-}\rangle_{t} = \frac{\gamma_{12}^{2} + \Omega_{12}^{2}}{2B} \left[\cosh Dt - \cos Et \right] e^{-2wt},$$

$$\langle S_{1}^{+}S_{2}^{-}\rangle_{t} = \frac{\kappa e^{-2wt}}{4B} \left\{ 2(u_{12} + i\Delta) \left[\cos Et - \cosh Dt \right] - \frac{D^{2} + 4i(\Delta u_{12} - \gamma_{12}\Omega_{12})}{D} \sinh Dt - \frac{E^{2} - 4i(\Delta u_{12} - \gamma_{12}\Omega_{12})}{D} \sinh Dt \right\},$$

$$(49)$$

Eqs. (47) and (48) describe the probabilities of finding atom 1 and atom 2, respectively, in their excited states at time t. From the form of these equations it is obvious that the probabilities contain some oscillatory terms with the oscillation frequency E. These oscillations are superimposed on a smooth exponential decay and remain in the formulae even for identical atoms. For identical atoms, however, the sum of the two probabilities exhibits no oscillations¹⁴). This means that the excitation is transferred back and forth from one atom to the other with the frequency $E = 2\Omega_{12}$. It is also clear from eq. (48) that atom 2 can be excited due to its interaction with atom 1 only ($\gamma_{12} \neq 0$ and/or $\Omega_{12} \neq 0$). If the two atoms are not identical the situation is more complicated and we will discuss some consequences of the fact that the two atoms have different transition frequencies and/or different linewidths.

First, we consider the angular distribution of the radiation intensity from the system of two nonidentical atoms. Expression (25) for the intensity can be written in the form

$$I(\mathbf{R}, t) = I_t(\hat{R}, t)R(\hat{R}, t), \qquad (51)$$

where $I_{\rm f}(\hat{R},t)$ is the intensity radiated by free atoms, which in the case when initially only atom 1 was excited is given by

$$I_{\rm f}(\hat{R},t) = \gamma_1 u(\hat{R}) e^{-2\gamma_1 t}$$
 (52)

The radiation pattern introduced by eq. (51) describes the deviation of the radiation emitted by interacting atoms from that emitted by independent atoms at the same moment of time t. Thus, the radiation pattern $R(\hat{R}, t)$ provides a measure of the cooperative behaviour of the two-atom system. Of course, for independent atoms $R(\hat{R}, t) = 1$.

If, at t = 0, only atom 1 was inverted, the radiation pattern $R(\hat{R}, t)$ takes the form

$$R(\hat{R}, t) = e^{-2u_{12}t} \left\{ \left[\frac{1}{2} \left(1 - \frac{u_{12}^2 + \Delta^2 + \left(\frac{\gamma_2}{\gamma_1} \right)^{1/2} (\gamma_{12}^2 + \Omega_{12}^2)}{B} \right) + \left(\frac{\gamma_2}{\gamma_1} \right)^{1/2} \frac{(\gamma_{12}u_{12} - \Delta\Omega_{12})}{B} \cos k\hat{R} \cdot r_{12} - \left(\frac{\gamma_2}{\gamma_1} \right)^{1/2} \frac{(\Delta\gamma_{12} + u_{12}\Omega_{12})}{B} \sin k\hat{R} \cdot r_{12} \right] \cos Et + \frac{1}{2BE} \left[u_{12}E^2 - 4\Delta(\Delta u_{12} - \gamma_{12}\Omega_{12}) \right]$$

$$-\left(\frac{\gamma_{2}}{\gamma_{1}}\right)^{1/2} \left[4\gamma_{12}(\Delta u_{12} - \gamma_{12}\Omega_{12}) - \Omega_{12}E^{2}\right] \sin k\hat{R} \cdot \mathbf{r}_{12}$$

$$-\left(\frac{\gamma_{2}}{\gamma_{1}}\right)^{1/2} \left[\gamma_{12}E^{2} + 4\Omega_{12}(\Delta u_{12} - \gamma_{12}\Omega_{12})\right] \cos k\hat{R} \cdot \mathbf{r}_{12} \right] \sin Et$$

$$+\left[\frac{1}{2}(B + u_{12}^{2} + \Delta^{2}) + \frac{1}{2}\left(\frac{\gamma_{2}}{\gamma_{1}}\right)^{1/2}(\gamma_{12}^{2} + \Omega_{12}^{2})\right]$$

$$+\left(\frac{\gamma_{2}}{\gamma_{1}}\right)^{1/2}(\Delta\gamma_{12} + u_{12}\Omega_{12}) \sin k\hat{R} \cdot \mathbf{r}_{12}$$

$$-\left(\frac{\gamma_{2}}{\gamma_{1}}\right)^{1/2}(\gamma_{12}u_{12} - \Delta\Omega_{12}) \cos k\hat{R} \cdot \mathbf{r}_{12} \right] \frac{\cosh Dt}{B}$$

$$+\left[u_{12}D^{2} + 4\Delta(\Delta u_{12} - \gamma_{12}\Omega_{12})\right]$$

$$+\left(\frac{\gamma_{2}}{\gamma_{1}}\right)^{1/2} \left[4\gamma_{12}(\Delta u_{12} - \gamma_{12}\Omega_{12}) + \Omega_{12}D^{2}\right] \sin k\hat{R} \cdot \mathbf{r}_{12}$$

$$+\left(\frac{\gamma_{2}}{\gamma_{1}}\right)^{1/2} \left[4\Omega_{12}(\Delta u_{12} - \gamma_{12}\Omega_{12}) - \gamma_{12}D^{2}\right] \cos k\hat{R} \cdot \mathbf{r}_{12} \right] \frac{\sinh Dt}{2BD}.$$

$$(53)$$

To visualize the behaviour of the radiation pattern given by eq. (53), we have plotted $R(\theta, t)$ in figs. 3-5, with θ : the angle between the observation direction and the vector \mathbf{r}_{12} , for $\mathbf{r}_{12} = \lambda_0/12$ and various γ_1 , γ_2 , Δ and t.

Fig. 3 shows that, for $\mu \perp r_{12}$, the radiation pattern is spherically symmetric at t=0 and becomes nonspherical for t>0. During the emission process this pattern oscillates in time as is shown in fig. 4; however, the amplitude of these oscillations is different in different directions. From fig. 4 it is seen that for identical atoms ($\gamma_2 = \gamma_1$ and $\Delta = 0$) the radiation pattern decays monotonically in time. It is also evident from figs. 3 and 4 that for nonidentical atoms the radiation pattern (53) can become considerably greater than unity meaning that during this interval of time the atoms radiate at a higher rate than independent atoms. This gives clear evidence of the cooperative behaviour of the atoms. It may seem puzzling at a first glance that nonidentical atoms can radiate more collectively than identical atoms, but this is a consequence of activating the transition $|-\rangle \rightarrow |0\rangle$ which, interfering with the transition $|+\rangle \rightarrow |0\rangle$, gives quantum beats and an enhanced radiation rate at time t $(R(\theta, t) > 1)$. If the transition dipole moment μ is parallel to the vector r_{12} connecting the two atoms, the radiation pattern $R(\theta, t)$ becomes asymmetric as

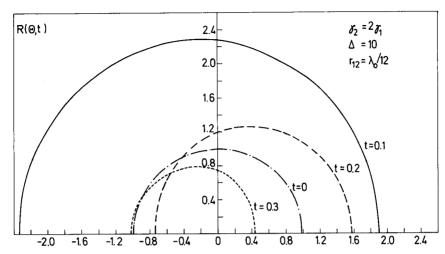


Fig. 3. Angular distribution of the radiation pattern for only one atom initially excited, with atomic separation $r_{12} = \lambda_0/12$, $\Delta = 10$, $\gamma_2 = 2\gamma_1$ and different times.

time elapses, as is shown in fig. 5; however, it should be kept in mind that in this case there is no radiation at all for $\theta = 0$, π because $\theta = \vartheta$ and $u(\hat{R}) = 0$. So, in this case, the concept of radiation pattern becomes rather artificial.

We now proceed to illustrate the influence of differences between the atoms on the total radiation intensity I(t) defined by eq. (26). Using eqs. (47)–(50), we find that for two nonidentical atoms the total radiation intensity takes the form

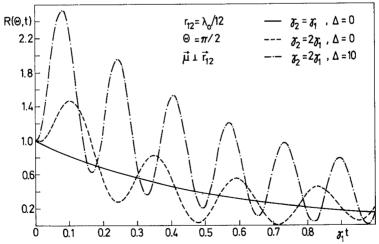


Fig. 4. Radiation pattern as a function of time $\gamma_1 t$ for only one atom initially excited, with $r_{12} = \lambda_0/12$, $\Theta = \pi/2$ and different transition frequencies and natural linewidths. Here, Θ is the angle between r_{12} and the observation direction R.

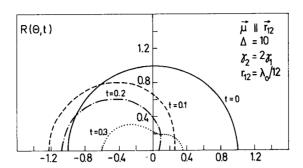


Fig. 5. As in fig. 3, albeit for $\hat{\mu}_1$ and $\hat{\mu}_2$ parallel to r_{12} .

$$I(t) = e^{-2wt} \left\{ \left[\gamma_{1} - \frac{\gamma_{1}(u_{12}^{2} + \gamma_{12}^{2} + \Delta^{2}) + 2\gamma_{12}\Delta\Omega_{12} + \gamma_{2}\Omega_{12}^{2}}{B} \right] \cos Et + \left[\gamma_{1} + \frac{\gamma_{1}(u_{12}^{2} + \gamma_{12}^{2} + \Delta^{2}) + 2\gamma_{12}\Delta\Omega_{12} + \gamma_{2}\Omega_{12}^{2}}{B} \right] \cosh Dt + \frac{\left[(\gamma_{1}u_{12} - \gamma_{12}^{2})E^{2} + 4(\gamma_{1}\Delta + \gamma_{12}\Omega_{12})(\gamma_{12}\Omega_{12} - \Delta u_{12}) \right]}{BE} \sin Et + \frac{\left[(\gamma_{1}u_{12} - \gamma_{12}^{2})D^{2} - 4(\gamma_{1}\Delta + \gamma_{12}\Omega_{12})(\gamma_{12}\Omega_{12} - \Delta u_{12}) \right]}{BD} \sinh Dt \right\}.$$
(54)

In figs. 6–9 we have plotted the total radiation intensity I(t) given by eq. (54) for several values of γ_2/γ_1 , Δ and different interatomic separations. In the case of nonidentical atoms, we find a pronounced deviation from the exponential decay law. The pronounced sinusoidal modulation (quantum beats effect) is clearly visible in the graphs. The frequency of this modulation is dependent on the interatomic interaction and vanishes for large interatomic separations (see fig. 9). This oscillatory behaviour of the total radiation intensity arises due to interference between the two possible transition amplitudes $(|\pm\rangle)$ or $|\varphi_+\rangle \rightarrow |0\rangle$) (see fig. 2) that contribute to the spontaneous emission. There are two possible mechanisms leading to quantum beats in such a system. One, consisting in opening the channel $|-\rangle \rightarrow |0\rangle$ when the two atoms have the same transition frequencies ($\Delta = 0$) and their transition dipole moments (or linewidths) are different, is illustrated in fig. 6. The other, consisting in a mixing of the states $|+\rangle$ and $|-\rangle$ (when $\Delta \neq 0$) leading to the new states $|\varphi_+\rangle$ and $|\varphi_-\rangle$ both of which are coupled to the ground state $|0\rangle$, is illustrated in fig. 7. It is seen that when the interatomic separations are small and the atom which was initially in its ground state has a natural linewidth larger than the initially excited atom i.e. when the total radiation intensity for some periods of time

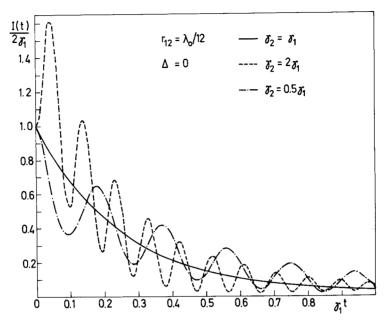


Fig. 6. Normalized total radiation rate as a function of time $\gamma_1 t$ for only one atom initially excited, $\Delta = 0$, $r_{12} = \lambda_0/12$ and different natural linewidths.

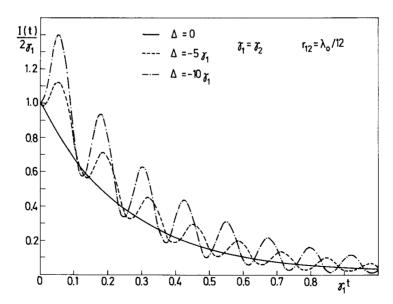


Fig. 7. Normalized total radiation rate versus $\gamma_1 t$ for initially one atom excited, $\gamma_1 = \gamma_2$, $r_{12} = \lambda_0/12$ and different transition frequencies.

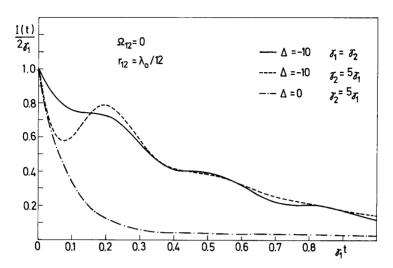


Fig. 8. Normalized total radiation rate versus $\gamma_1 t$ for initially only one atom excited, with dipole-dipole interaction ignored.

becomes greater than its initial value. This means that the emission is superradiant. Superradiant behaviour can also appear if the atoms have the same natural linewidth, but this can occur only if the atoms differ considerably in their transition frequencies (see fig. 7). This collective effect is absent in the case of two identical atoms ($\gamma_2 = \gamma_1$ and $\Delta = 0$) as well as in the case when the dipole-dipole interaction Ω_{12} is ignored, as is convincingly shown in fig. 8. For identical atoms the subradiant state $|-\rangle$ is disconnected from the other states of the system (although it stores one half of the initial population) and we have

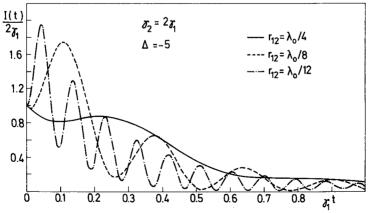


Fig. 9. Normalized total radiation rate as a function of time $\gamma_1 t$ for initially one atom excited, $\Delta = -5$, $\gamma_2 = 2\gamma_1$, and different interatomic separations.

the transition $|+\rangle \rightarrow |0\rangle$ only and, of course, there are no beats in the total intensity¹⁴).

As we have shown, in the case when initially one atom was excited, both the radiation pattern $R(\theta, t)$ can become larger than unity and the total radiation intensity I(t) can become larger than its initial value I(0). The radiation pattern $R(\theta, t)$ may be said to describe the instantaneous collective behaviour of the system while the total radiation intensity I(t) can be said to describe its global behaviour; thus, for initially one atom inverted, the system can radiate superradiantly in both the instantaneous and global sense.

The quantum beats effect has recently been the subject of theoretical as well as experimental investigations with regard to numerous optical processes^{3,31–33}).

4.2.2. Initially both atoms excited

When initially both atoms were excited, we have $A_0 = D_0 = G_0 = 1$, and from the general solutions (37)–(40) we arrive at the following formula for the radiation pattern:

$$\begin{split} R(\hat{R},t) &= [I_{\rm f}(\hat{R},t)]^{-1} \bigg\{ \frac{64\gamma_{12} \, \mathrm{e}^{-4wt}}{(D^2 - 4w^2)(E^2 + 4w^2)} \, \bigg[\, w[\gamma_{12}(w + u_{12}^2 + \Omega_{12}^2) \\ &- 2u_{12}\Delta\Omega_{12}] + \bigg(\frac{\gamma_2}{\gamma_1} \bigg)^{1/2} \, w(w^2 + \Omega_{12}^2 - u_{12}^2) \cos k\hat{R} \cdot r_{12} \\ &+ \bigg(\frac{\gamma_2}{\gamma_1} \bigg)^{1/2} \big[(w^2 - u_{12}^2)\Delta + u_{12}\gamma_{12}\Omega_{12} \big] \sin k\hat{R} \cdot r_{12} \bigg] \\ &+ \frac{2\mathrm{e}^{-2wt}}{B(D^2 - 4w^2)} \bigg[\, wX - 2\bigg(\frac{\gamma_2}{\gamma_1} \bigg)^{1/2} \, w\gamma_{12} [D^2 + 4(\Omega_{12}^2 - u_{12}^2)] \\ &\times \cos k\hat{R} \cdot r_{12} + Y\bigg(\frac{\gamma_2}{\gamma_1} \bigg)^{1/2} \sin k\hat{R} \cdot r_{12} \bigg] \cosh Dt \\ &+ \frac{\mathrm{e}^{-2wt}}{BD(D^2 - 4w^2)} \bigg[4M + \bigg(\frac{\gamma_2}{\gamma_1} \bigg)^{1/2} N \cos k\hat{R} \cdot r_{12} \\ &+ 8w\gamma_{12} \bigg(\frac{\gamma_2}{\gamma_1} \bigg)^{1/2} [\Delta D^2 - 4u_{12}(\Delta u_{12} - \gamma_{12}\Omega_{12})] \sin k\hat{R} \cdot r_{12} \bigg] \sinh Dt \\ &+ \frac{2\mathrm{e}^{-2wt}}{B(E^2 + 4w^2)} \, \bigg[\, wX' + 2w\gamma_{12} \bigg(\frac{\gamma_2}{\gamma_1} \bigg)^{1/2} [E^2 - 4(\Omega_{12}^2 - u_{12}^2)] \\ &\times \cos k\hat{R} \cdot r_{12} + Y'\bigg(\frac{\gamma_2}{\gamma_1} \bigg)^{1/2} \sin k\hat{R} \cdot r_{12} \bigg] \cos Et \end{split}$$

$$+ \frac{e^{-2wt}}{BE(E^{2} + 4w^{2})} \left[-4M' + N' \left(\frac{\gamma_{2}}{\gamma_{1}} \right)^{1/2} \cos k\hat{R} \cdot \boldsymbol{r}_{12} \right.$$

$$- 8w\gamma_{12} \left(\frac{\gamma_{2}}{\gamma_{1}} \right)^{1/2} \left[\Delta E^{2} + 4u_{12} (\Delta u_{12} - \gamma_{12} \Omega_{12}) \right] \sin k\hat{R} \cdot \boldsymbol{r}_{12} \right] \sin Et \right\},$$
(55)

where

$$X = D^{2}(\frac{1}{4}E^{2} - \gamma_{12}^{2}) - (w^{2} - u_{12}^{2})(E^{2} + 4u_{12}^{2}) + 16\Delta\gamma_{12}\Omega_{12}u_{12}$$

$$- 4u_{12}^{2}(\Delta^{2} + \Omega_{12}^{2}) - 4\gamma_{12}^{2}(w^{2} + u_{12}^{2} + 2\Omega_{12}^{2}),$$

$$Y = (D^{2} - 4w^{2})(u_{12}\Omega_{12} - \Delta\gamma_{12}) - 8\gamma_{12}\Delta(w^{2} - u_{12}^{2}) - 8\gamma_{12}^{2}u_{12}\Omega_{12},$$

$$M = D^{2}[-\frac{1}{4}u_{12}^{2}E^{2} + u_{12}^{2}(w^{2} - u_{12}^{2} + \gamma_{12}^{2} + \Omega_{12}^{2}) + 2w^{2}\gamma_{12}^{2} - \Delta u_{12}\gamma_{12}\Omega_{12}]$$

$$- 4w^{2}(\Delta u_{12} - \gamma_{12}\Omega_{12})(2\gamma_{12}\Omega_{12} - \Delta u_{12}),$$

$$N = \gamma_{12}D^{4} + 4D^{2}[2\gamma_{12}(w^{2} - u_{12}^{2}) + \gamma_{12}(\Omega_{12}^{2} - w^{2}) + \Delta u_{12}\Omega_{12}]$$

$$- 16w^{2}\Omega_{12}(\Delta u_{12} - \gamma_{12}\Omega_{12}),$$

and the primed quantities are obtained by the mutual interchange $E^2 \rightleftharpoons D^2$ in the above formulae. The intensity radiated by free atoms when both atoms were initially excited is obviously equal to

$$I_{f}(\hat{R}, t) = u(\hat{R})(\gamma_{1} e^{-2\gamma_{1}t} + \gamma_{2} e^{-2\gamma_{2}t}).$$
(56)

The formula for the total radiation intensity I(t) is in this case the following:

$$I(t) = \frac{128w\gamma_{12}[\gamma_{12}w^{2} - \Omega_{12}(\Delta u_{12} - \gamma_{12}\Omega_{12})]}{(D^{2} - 4w^{2})(E^{2} + 4w^{2})} e^{-4wt}$$

$$+ e^{-2wt} \left[\frac{wQ}{B(D^{2} - 4w^{2})} \cosh Dt + \frac{W}{BD(D^{2} - 4w^{2})} \sinh Dt + \frac{wQ'}{B(E^{2} + 4w^{2})} \cos Et + \frac{W'}{BE(E^{2} + 4w^{2})} \sin Et \right],$$
(57)

with

$$Q = D^{2}(\frac{1}{2}E^{2} - 6\gamma_{12}^{2} - 2\gamma_{1}\gamma_{2}) - 8w^{2}(\Omega_{12}^{2} + \Delta^{2}) + 32\gamma_{12}\Omega_{12}(\Delta u_{12} - \gamma_{12}\Omega_{12}),$$

$$W = D^{4}(\gamma_{12}^{2} - u_{12}^{2}) + 4D^{2}[\gamma_{12}^{2}(\Omega_{12}^{2} + 3w^{2}) + u_{12}^{2}(w^{2} - \Delta^{2})] + 16w^{2}(\Delta u_{12} - \gamma_{12}\Omega_{12})(\Delta u_{12} - 3\gamma_{12}\Omega_{12}),$$
(58)

and, again, the primed quantities are obtained on performing the interchange $E^2 \rightleftharpoons D^2$

The above formulae are very extensive and complicated but are explicit analytical formulae describing the radiation from two initially excited, nonidentical atoms. The radiation pattern given by eq. (55) is illustrated graphically in figs. 10 and 11. Fig. 10 shows that even for nonidentical atoms the radiation pattern starts out spherically symmetric, but later becomes nonspherical, although the asymmetry is not very pronounced. It is seen, however, that for nonidentical atoms the radiation in the directions with $\Theta = 0$ and π is different in contrast to the case of identical atoms. It is also evident from fig. 11 that, contrary to the case when only one atom was initially excited, the radiation pattern for short times t is greater than unity even for identical atoms. Its value can be greater or smaller if the atoms are not identical, depending on the values of the parameters. For large Δ , slight oscillations can occur as it is seen from fig. 11.

The total radiation intensity I(t) given by eq. (57) is plotted in figs. 12 and 13 for $r_{12} = \lambda_0/12$ and various γ_2/γ_1 and Δ . One notes that the total radiation intensity is always lower than its initial value and decreases monotonically in time. Some weak beats can appear for strongly different atoms, but one never gets an increase in I(t). The explanation for this behaviour may be given in terms of the eigenstates of the system as a whole (fig. 2). Initially the system is in the state $|2\rangle$, and as time elapses it decays through the states $|\pm\rangle$ to the ground state $|0\rangle$. For identical atoms the state $|-\rangle$ will never be populated, and we have downward transitions via the $|+\rangle$ state only. For nonidentical atoms the subradiant state $|-\rangle$ acquires some population from the state $|2\rangle$ as time elapses. However, at the initial stage of the evolution we have a system

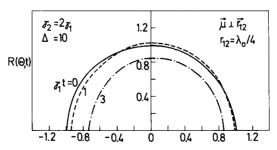


Fig. 10. Angular distribution of the radiation pattern $R(\Theta, t)$ for initially fully inverted system, $r_{12} = \lambda_0/4$, $\gamma_2 = 2\gamma_1$, $\Delta = 10$ and different times $\gamma_1 t$.

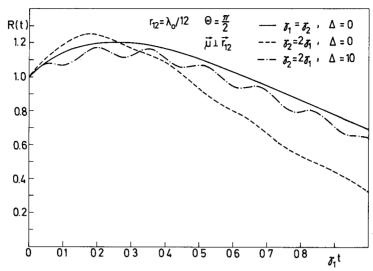


Fig. 11. Radiation pattern in the direction $\Theta = \pi/2$ as a function of time $\gamma_1 t$ for fully inverted atomic system, $r_{12} = \lambda_0/12$, $\hat{\mu} \perp r_{12}$ and different transition frequencies and different natural linewidths.

with the common upper level $|2\rangle$ which does not give quantum beats. As the states $|\pm\rangle$ become populated in the course of evolution, the possibility of interference leading to quantum beats appears. This explains the existence of small beats in I(t).

Thus, it can be said that for both atoms excited the system can radiate in a superrradiant way in the instantaneous sense $(R(\Theta, t) > 1)$ but not in the global

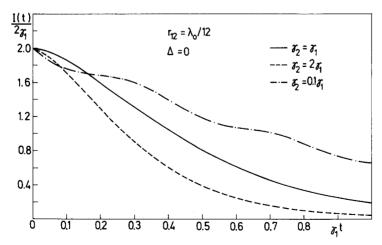


Fig. 12. Normalized total radiation rate versus $\gamma_1 t$ for initially fully inverted system, $r_{12} = \lambda_0/12$, $\Delta = 0$ and different natural linewidths.

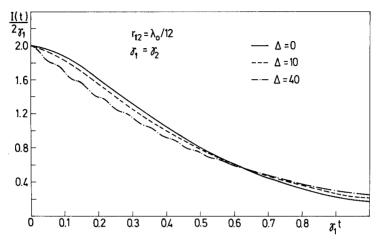


Fig. 13. Normalized total radiation rate versus $\gamma_1 t$ for initially fully inverted system, $r_{12} = \lambda_0/12$, $\gamma_2 = \gamma_1$, and different Δ .

sense (I(t)) is always less than I(0)). Thus, the radiation from a system of two nonidentical atoms depends in an essential manner on the initial excitation of the system.

5. Spectral properties of the radiation

In this section we investigate the spectral distribution of the radiation defined as ¹³)

$$\Phi(\omega) = \operatorname{Re} \left\{ \int_{0}^{\infty} dt \int_{0}^{\infty} dt' \int d\Omega_{R} \left\langle \boldsymbol{E}^{(-)}(\boldsymbol{R}, t) \cdot \boldsymbol{E}^{(+)}(\boldsymbol{R}, t') \right\rangle e^{-i\omega(t-t')} \right\}, \quad (59)$$

where integration $d\Omega_R$ is over all directions, $E^{(\pm)}(R,t)$ are given by eq. (24), and $t'=t+\tau$ ($\tau>0$).

In the case when the radiation is initially in the vacuum state $|\{0\}\rangle$, we have from eq. (24)

$$\Phi(\omega) = \operatorname{Re}\left\{\int_{0}^{\infty} dt \int_{0}^{\infty} dt' \sum_{i,j=1}^{2} \gamma_{ij} \langle S_{i}^{+}(t) S_{j}^{-}(t') \rangle e^{-i\omega(t-t')}\right\}.$$
(60)

In order to obtain the spectral distribution of the radiation it is necessary to know the time correlation function for the atomic dipole moments. We obtain this correlation function from the equations of motion (13) which lead to a

(64)

closed set of four equations of motion for the correlation functions. Solution of this set of equations by the Laplace transform method leads to the following results:

$$\begin{cases}
S_{1}^{+}(t)S_{1}^{-}(t+\tau) \rangle = \\
\sum_{\substack{i=1\\ i\neq j\neq k\neq i}}^{4} \left\{ \frac{\left[A_{0}(s_{i}+\gamma_{2}-i\Delta) - B_{0}\kappa\right]\left[(s_{i}+2\gamma_{i}+\gamma_{2}-i\Delta)(s_{i}+\gamma_{i}+2\gamma_{2}+i\Delta) - \kappa^{*2}\right]}{(s_{i}-s_{j})(s_{i}-s_{k})(s_{i}-s_{i})} \\
- \frac{2H_{0}\kappa\left[2\gamma_{12}(s_{i}+\gamma_{2}-i\Delta) + 2\kappa\right]}{(s_{i}-s_{j})(s_{i}-s_{k})(s_{i}-s_{i})} \right\} e^{t_{i}\tau},$$
(61)
$$\begin{cases}
S_{2}^{+}(t)S_{2}^{-}(t+\tau) \rangle = \\
\sum_{\substack{i=1\\ i\neq j\neq k\neq i}}^{4} \left\{ -\frac{2H_{0}\kappa\left[2\gamma_{12}(s_{i}+\gamma_{i}+i\Delta) + 2\kappa\right]}{(s_{i}-s_{j})(s_{i}-s_{k})(s_{i}-s_{i})} \\
+ \frac{\left[D_{0}(s_{j}+\gamma_{i}+i\Delta) - G_{0}\kappa\right]\left[(s_{i}+\gamma_{i}+2\gamma_{2}+i\Delta)(s_{i}+2\gamma_{i}+\gamma_{2}-i\Delta) - \kappa^{*2}\right]}{(s_{i}-s_{j})(s_{i}-s_{k})(s_{i}-s_{i})} \right\} e^{t_{j}\tau},$$
(62)
$$\langle S_{1}^{+}(t)S_{2}^{-}(t+\tau) \rangle = \\
\sum_{\substack{i=1\\ i\neq j\neq k\neq i}} \left\{ \frac{2H_{0}\kappa\left[(s_{j}+2\gamma_{i}+\gamma_{2}-i\Delta)(s_{i}+\gamma_{i}+i\Delta) + |\kappa|^{2}\right]}{(s_{i}-s_{j})(s_{i}-s_{k})(s_{i}-s_{i})} \\
+ \frac{\left[B_{0}(s_{i}+\gamma_{i}+i\Delta) - A_{0}\kappa\right]\left[(s_{i}+2\gamma_{i}+\gamma_{2}-i\Delta)(s_{i}+\gamma_{i}+2\gamma_{2}+i\Delta) - \kappa^{*2}\right]}{(s_{i}-s_{j})(s_{i}-s_{k})(s_{i}-s_{i})} \right\} e^{t_{i}\tau},$$
(63)
$$\langle S_{2}^{+}(t)S_{1}^{-}(t+\tau) \rangle = \\
\sum_{\substack{i=1\\ i\neq j\neq k\neq i}}^{4} \left\{ \frac{2\kappa H_{0}\left[(s_{i}+\gamma_{i}+2\gamma_{2}+i\Delta)(s_{i}+\gamma_{2}-i\Delta) + |\kappa|^{2}\right]}{(s_{i}-s_{j})(s_{i}-s_{k})(s_{i}-s_{i})} \\
+ \frac{\left[G_{0}(s_{j}+\gamma_{2}-i\Delta) - D_{0}\kappa\right]\left[(s_{i}+\gamma_{i}+2\gamma_{2}+i\Delta)(s_{i}+2\gamma_{i}+\gamma_{2}-i\Delta) - \kappa^{*2}\right]}{(s_{i}-s_{j})(s_{i}-s_{k})(s_{i}-s_{i})} \right\} e^{t_{i}\tau},$$

where $A_0 = \langle S_1^+(t)S_1^-(t) \rangle$, $B_0 = \langle S_1^+(t)S_2^-(t) \rangle$, $D_0 = \langle S_2^+(t)S_2^-(t) \rangle$, $G_0 \langle S_2^+(t)S_1^-(t) \rangle$ and $H_0 = \langle S_1^+(t)S_2^+(t)S_1^-(t)S_2^-(t) \rangle$ are given by eqs. (29),

(37)–(40) and their values depend on the initial conditions. s_1 , s_2 , s_3 , s_4 are the four roots of the equation

$$[(s + \gamma_1 + i\Delta)(s + \gamma_2 - i\Delta) - \kappa^2]$$

$$\times [(s + 2\gamma_1 + \gamma_2 - i\Delta)(s + \gamma_1 + 2\gamma_2 + i\Delta) - \kappa^{*2}] = 0.$$
(65)

The roots can be expressed in terms of the parameters w, Δ , u_{12} , γ_{12} and Ω_{12} and are given by:

$$s_{1,2} = -w \pm \left[(u_{12} - i\Delta)^2 + (\gamma_{12} + i\Omega_{12})^2 \right]^{1/2},$$

$$s_{3,4} = -3w \pm \left[(u_{12} + i\Delta)^2 + (\gamma_{12} - i\Omega_{12})^2 \right]^{1/2}.$$
(66)

A. When initially only one atom was excited and the other was in its ground state, and assuming strong interatomic interaction $(b_{12} \ge 1)$, we obtain from eq. (60) and eqs. (61)-(64) the following formula for the spectral distribution of the radiation field:

$$\Phi(\nu) = \frac{1}{(n^2 + 1)} \left\{ \frac{w + n(\gamma_1 n + \gamma_{12}) + (\gamma_1 n + \gamma_{12})\sqrt{n^2 + 1}}{(w + \frac{1}{2}D)^2 + (\nu - \frac{1}{2}E)^2} + \frac{w + n(\gamma_1 n + \gamma_{12}) - (\gamma_1 n + \gamma_{12})\sqrt{n^2 + 1}}{(w - \frac{1}{2}D)^2 + (\nu + \frac{1}{2}E)^2} - 2(u_{12} + n\gamma_{12}) \times \frac{(w^2 - \frac{1}{4}D^2) + (\nu^2 - \frac{1}{4}E^2)}{[(w + \frac{1}{2}D)^2 + (\nu - \frac{1}{2}E)^2][(w - \frac{1}{2}D)^2 + (\nu + \frac{1}{2}E)^2]} \right\},$$
(67)

where $\nu = \omega_0 - \omega$, and $n = \Delta/\Omega_{12}$ provides a measure of the nonidenticity of the atoms. The spectral distribution of radiation (67) is illustrated in fig. 14. If Ω_{12} exceeds Δ appreciably $(n \le 1)$, the atoms evolve as identical, and our spectrum is identical with that obtained by Lehmberg¹³).

In this case the spectrum consists of two peaks located at $\nu=\pm\Omega_{12}$. The broader peak is due to the transition $|+\rangle \rightarrow |0\rangle$ and the narrower peak to the transition $|-\rangle \rightarrow |0\rangle$. This narrow peak has a width practically equal to zero confirming that the probability of transition $|-\rangle \rightarrow |0\rangle$ is practically equal to zero for two identical atoms. A different situation occurs if the atoms are nonidentical. For n=1 the peaks have widths $w\pm \frac{1}{2}D$ and are located at $\nu=\pm 2\sqrt{2}\Omega_{12}$. In this case the probabilities of transition $|+\rangle \rightarrow |0\rangle$ and $|-\rangle \rightarrow |0\rangle$ are practically identical and the spectrum becomes symmetrical, as is evident from fig. 14.

For $n \ge 1$, $\Phi(\nu)$ takes the form

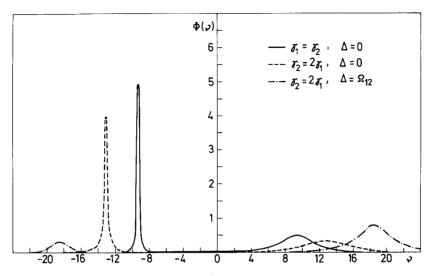


Fig. 14. Spectral distribution of radiation for initially only one atom excited, $r_{12} = \lambda_0/12$ and different natural linewidths and different transition frequencies.

$$\Phi(\nu) = \frac{2\gamma_1}{[\gamma_1^2 + (\omega - \omega_1)^2]} \ . \tag{68}$$

Here, we have a single Lorentzian peak, as the atoms radiate independently.

B. If both atoms are initially excited into their upper states, the spectral distribution of the radiation takes the form:

$$\Phi(\nu) = \frac{1}{2(n^{2}+1)} \left\{ \frac{W_{-}}{(2w+D)} \frac{1}{[(w-\frac{1}{2}D)^{2}+(\nu+\frac{1}{2}E)^{2}]} + \frac{W_{+}}{(2w-D)} \frac{1}{[(w+\frac{1}{2}D)^{2}+(\nu-\frac{1}{2}E)^{2}]} + \frac{8\gamma_{12}(w+\gamma_{1})\sqrt{n^{2}+1}}{(2w+D)} \right. \\
\times \frac{(3w+\frac{1}{2}D)(w-\frac{1}{2}D)+(\nu+\frac{1}{2}E)^{2}}{[(3w+\frac{1}{2}D)^{2}+(\nu+\frac{1}{2}E)^{2}][(w-\frac{1}{2}D)^{2}+(\nu+\frac{1}{2}E)^{2}]} \\
- \frac{8\gamma_{12}(w+\gamma_{1})\sqrt{n^{2}+1}}{(2w-D)} \\
\times \frac{(3w-\frac{1}{2}D)(w+\frac{1}{2}D)+(\nu-\frac{1}{2}E)^{2}}{[(3w-\frac{1}{2}D)^{2}+(\nu-\frac{1}{2}E)^{2}]} \right\}, \quad (69)$$

where

$$W_{\pm} = \sqrt{n^2 + 1} (2w \pm D) [(2w \mp D)\sqrt{n^2 + 1} \pm 4\gamma_{12}]. \tag{70}$$

The spectrum now contains two double peaks, and some nonresonant terms appear. These peaks are easily understood on referring to the energy-level diagram of the system, fig. 2. Transitions from level $|2\rangle$ to level $|\varphi_{-}\rangle$ and from level $|\varphi_{+}\rangle$ to level $|0\rangle$ occur at the same frequency $\omega_{0} + \frac{1}{2}E$. Simultaneously, transitions from level $|2\rangle$ to level $|\varphi_{+}\rangle$ and from level $|\varphi_{-}\rangle$ to level $|0\rangle$ occur at the same frequency $\omega_{0} - \frac{1}{2}E$. However, centred at each of these frequencies there are two lines having different widths due to the different lifetimes of the two states involved. For $n \ll 1$ the spectrum is the same as for identical atoms³⁴). For n = 1 it consists of two practically identical peaks signifying that both transitions $|2\rangle \rightarrow |\varphi_{\pm}\rangle \rightarrow |0\rangle$ are equally probable. If n is much greater than unity $(n \gg 1)$, the spectrum consists of two identical Lorentzian peaks with the widths γ_{1} and γ_{2} located at the frequencies ω_{1} and ω_{2} , respectively, as the atoms radiate independently.

The changes in spectral distributions confirm that for nonidentical atoms the transition $|-\rangle \rightarrow |0\rangle$ becomes important and equally probable as the transition $|+\rangle \rightarrow |0\rangle$ leading to quantum beats in the radiation intensity.

6. Conclusions

We have solved the problem of spontaneous emission from nonidentical atoms assuming that the atoms have different transition frequencies and different natural linewidths. Starting from the Hamiltonian for N nonidentical atoms coupled to the continuum of quantized electromagnetic modes we have derived, within the Born and Markoff approximations, the equations of motion for the atomic operators. We have used these equations in our considerations of the spontaneous emission from a pair of nonidentical atoms the separation r_{12} of which is comparable to the resonant wavelength λ_0 .

The temporal evolution of the atomic populations, the angular distribution of the radiation intensity, the total radiation intensity (rate) and the spectral distributions are found with the following initial conditions, a) only one atom excited and b) both atoms excited. We have found that in the case a) both the radiation pattern $R(\Theta, t)$ and the total radiation intensity I(t) exhibit oscillatory behaviour due to the quantum beats effect. A distinguishing feature of a system of nonidentical atoms is the presence of superradiant behaviour both, as we say, in the instantaneous (local in time) sense $(R(\Theta, t) > 1)$ and in the global sense (I(t) > I(0)). This means that nonidentical atoms are apt to radiate more collectively than identical atoms. This may seem somewhat strange at a first glance, but becomes understandable if one keeps in mind that for initial

conditions a) one half of the initial population is stored in the subradiant state $|-\rangle$ (see fig. 2), which state practically does not radiate if the atoms are identical and becomes active if the atoms are not identical. The interference between the transition amplitudes from the superradiant state $|+\rangle$ and the subradiant state $|-\rangle$, giving the quantum beats effect, can also lead to the increase of the total radiation intensity above its initial value for certain values of the parameters.

In the case b) the situation is different. The radiation pattern $R(\Theta,t)$ can become greater than unity, meaning that in the local instantaneous sense the radiation is superradiant. In other words, the system radiates at a higher rate at the time t than it would radiate at the same time if the atoms were independent. This is true even for identical atoms and differs from the case a) where it could happen only for nonidentical atoms. In the case b), however, there is no superradiant behaviour in the global sense i.e. the total radiation intensity I(t) can never be greater than its initial value I(0). In the representation of collective atomic states, the system starts from the common state $|2\rangle$ and decays via the states $|\varphi_{\pm}\rangle$ to its ground state $|0\rangle$. The configuration with a common upper level does not lead to quantum beats. After some time, when the states $|\varphi_{\pm}\rangle$ have acquired some population, the possibility of interference of the two transition amplitudes to the common ground state $|0\rangle$ arises. This explains the existence of weak oscillations in figs. 11–13.

We have also shown that the spectral distribution of the radiation is similar in both cases and consists of two peaks located at $\omega_0 \pm \frac{1}{2}E$ with the widths $w \pm \frac{1}{2}D$, respectively. When the interatomic distance is much smaller than the resonant wavelength, the spectrum for identical atoms has one Lorentzian peak located at $+\Omega_{12}$ with linewidth twice greater than that for isolated atoms. For nonidentical atoms the spectrum consists of two Lorentzian peaks the widths and intensities of which are practically identical. This confirms that for nonidentical atoms we have two possible transition frequencies between the collective states of the atomic system.

References

- 1) R.H. Dicke, Phys. Rev. 93 (1954) 99.
- 2) M. Gross and S. Haroche, Phys. Rep. 93 (1982) 301.
- 3) Q.H.F. Vrehen, H.M.J. Hikspoors and H.M. Gibbs, Phys. Rev. Lett. 38 (1977) 764.
- 4) M. Gross, J.M. Raimond and S. Haroche, Phys. Rev. Lett. 40 (1978) 1711.
- 5) J. Okada, K. Ikeda and M. Matsuoka, Opt. Commun. 27 (1978) 321.
- 6) Q.H.F. Vrehen and M.F.H. Schuurmans, Phys. Rev. Lett. 42 (1979) 224.
- 7) A.T. Rosenberger and T.A. De Temple, Phys. Rev. A 24 (1981) 868.
- 8) B. Coffey and R. Friedberg, Phys. Rev. A 17 (1978) 1033.
- 9) Th. Richter, Ann. der Phys. 38 (1981) 106.

- 10) H. Blank, M. Blank, K. Blum and A. Faridani, Phys. Lett. A 105 (1984) 39.
- 11) H.S. Freedhoff, J. Phys. B 19 (1986) 3035, 20 (1987) 285.
- 12) J. Seke, Lett. Nuovo Cimento 41 (1984) 321, 7D (1986) 447.
- 13) R.H. Lehmberg, Phys. Rev. A 2 (1970) 883.
- 14) G.S. Agarwal, Quantum Optics, G. Höhler, ed., Springer Tracts in Modern Physics, vol. 70 (Springer, Berlin, Heidelberg, New York, 1974).
- 15) M.J. Stephen, J. Chem. Phys. 40 (1964) 669.
- 16) A. Hutchinson and H.F. Hameka, J. Chem. Phys. 41 (1964) 2006.
- 17) E.A. Power, J. Chem. Phys. 46 (1967) 4297.
- 18) P.W. Milonni and P.L. Knight, Phys. Rev. A 10 (1974) 1096.
- 19) Th. Förster, Ann. der Phys. 2 (1948) 55.
- 20) D.L. Dexter, J. Chem. Phys. 21 (1952) 836.
- 21) J.R. Ríos Leite and C.B. De Araujo, Chem. Phys. Lett. 73 (1980) 71.
- 22) J. Cooper and D.N. Stacey, J. Phys. B 7 (1974) 2143.
- 23) E.A. Power, J. Phys. B 7 (1974) 2149.
- 24) J.H. Eberly, Acta Phys. Pol. A 39 (1971) 633.
- 25) G.S. Agarwal, Phys. Rev. A 4 (1971) 1778.
- 26) R. Jodoin and L. Mandel, Phys. Rev. A 9 (1974) 873, 10 (1974) 1898.
- 27) A.A. Varfolomeev, Zh. Eksp. Teor. Fiz. 59 (1970) 1702, (Sov. Phys.-JETP 32 (1971) 926.
- 28) P.W. Milonni and P.L. Knight, Phys. Rev. A 11 (1975) 1090.
- W. Heitler, The Quantum Theory of Radiation (Oxford Univ. Press, London, 1954) 3rd ed.,
 p. 354.
- 30) Z. Ficek, R. Tanaś and S. Kielich, Opt. Acta 33 (1986) 1149.
- 31) C. Mavroyannis, Physica 102A (1980) 193.
- 32) E. Abraham and R.K. Bullough, Opt. Commun. 34 (1980) 345.
- 33) A.G. Zajonc, Phys. Lett. A 96 (1983) 61.
- 34) H.S. Freedhoff, Can. J. Phys. 63 (1985) 114.