

## Cooperative effects in the spontaneous emission from two non-identical atoms†

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**Abstract.** Spontaneous emission from two non-identical two-level atoms is examined from the viewpoint of quantum beats and super-radiant effects. Our analytical solution, illustrated graphically, shows that the total radiation rate exhibits temporal sinusoidal modulation (beats). The beats result from the fact that the atoms have different transition frequencies and different natural linewidths. For small interatomic separations, the total radiation rate is greater than its initial value. This 'super-radiant' property is absent in the case of non-interacting atoms, as well as for two identical atoms. In the case when the atoms are separated by distances much smaller than the resonant wavelength, the population trapping effect can take place in such a system.

### 1. Introduction

Collective spontaneous emission, or super-radiance from a large number of atoms has been studied in many theoretical and experimental papers [1–4]. The phenomenon itself is intrinsically interesting because it very subtly blends the quantum and classical aspects, as well as the spontaneous and stimulated aspects of atomic emission. To understand collective radiation effects from a macroscopic system of atoms it is necessary to have a microscopic formulation of the interaction between the atoms and the electromagnetic field. Therefore, it seems useful to study the radiation from a system comprising a few atoms. The spontaneous radiation from two and three atoms has been investigated by many authors [1, 5–8]. Coffey and Friedberg [9] and Richter [10] have shown that the atomic configuration for a three-atom system has a strong influence on the total radiation rate. In some special cases the emission can be greater than its initial value. The emission is then referred to as super-radiant. Recently, Blank *et al.* [11] have shown that this effect, for atoms located in an equidistant linear chain, appears for six or more atoms. Coffey and Friedberg [9] have also considered the effect of inhomogeneous broadening of the atomic transition on the total radiation rate for two and three atoms. They have shown that in this case the total radiation rate is always lower than its initial value, and exhibits no temporal sinusoidal modulation (beats).

Milonni and Knight [12, 13] considered the effect of the retardation times on various spontaneous decay probability amplitudes for the case of two identical, as well as non-identical, atoms. The spontaneous decay probability for a system of two non-identical atoms has also been calculated by Varfolomeev [14].

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In the present paper we investigate the collective spontaneous emission from two non-identical two-level atoms assuming them to have different transition frequencies and different natural line-widths. We start from equations of motion (quantum Langevin equations) describing the time-evolution of the atomic operators. These equations are equivalent to a generalization of Lehmborg's [8] master equation to the case of non-identical atoms. The time-evolution of the atomic variables is dependent on the parameters  $\Delta_{12}$  and  $u_{12}$  which describe the difference between the atomic energies and that between their natural linewidths, respectively. This time-evolution is dependent also on the collective parameters  $\gamma_{12}$  and  $\Omega_{12}$ , which describe collective damping and the collective shift in energy levels. These two collective parameters, which provide a measure of the magnitude of interatomic interaction, determine the collective properties of our system of two non-identical atoms. We apply the solution of these equations of motion to derive the total radiation rate and discuss the possibility of a population trapping effect in such a system.

## 2. The equations of motion and their solutions

In the present paper, we consider two non-identical atoms separated by  $\mathbf{r}_{12}$ , coupled to each other via retarded dipole-dipole interaction and to the electromagnetic field vacuum, leading to spontaneous emission. Each atom is approximated by a two-level system: the ground state  $|1\rangle_i$  ( $i = 1, 2$ ) and the excited state  $|2\rangle_i$ , connected by an electric dipole transition. The atoms are assumed to have the transition frequencies  $\omega_1$  and  $\omega_2$ , respectively, and the corresponding natural linewidths  $2\gamma_1$  and  $2\gamma_2$ .

The time-evolution of the atomic operators is described by the following equations [15]:

$$\left. \begin{aligned} \frac{d}{dt} S_1^-(t) &= -(\gamma_1 + i\Delta_{12})S_1^-(t) - (\gamma_{12} + i\Omega_{12})S_2^-(t) \\ &\quad + 2(\gamma_{12} + i\Omega_{12})S_1^+(t)S_1^-(t)S_2^-(t) + 2E_0^{(+)}(\mathbf{r}_1, t)[S_1^+(t)S_1^-(t) - \frac{1}{2}] \\ \frac{d}{dt} S_2^-(t) &= -(\gamma_2 - i\Delta_{12})S_2^-(t) - (\gamma_{12} + i\Omega_{12})S_1^-(t) \\ &\quad + 2(\gamma_{12} + i\Omega_{12})S_2^+(t)S_2^-(t)S_1^-(t) + 2E_0^{(+)}(\mathbf{r}_2, t)[S_2^+(t)S_2^-(t) - \frac{1}{2}]. \end{aligned} \right\} (1)$$

where  $S_i^+$  and  $S_i^- = [S_i^+]^+$  are the raising and lowering operators of atom  $i$  ( $i = 1, 2$ ). The equations for  $S_1^+(t)$  and  $S_2^+(t)$  are the Hermitian adjoints of equation (1). In equations (1),  $\Delta_{12} = \frac{1}{2}(\omega_2 - \omega_1)$ ;  $2\gamma_1$  and  $2\gamma_2$  are the Einstein  $A$  coefficients for the atoms 1 and 2, respectively. The collective damping  $\gamma_{12}$  and frequency shift  $\Omega_{12}$  that arise from the retarded dipole-dipole interaction between the atoms have the form:

$$\left. \begin{aligned} \gamma_{12} &= -\frac{3}{2}\sqrt{(\gamma_1\gamma_2)} \operatorname{Im} f_{12}(kr_{12}), \\ \Omega_{12} &= \frac{3}{2}\sqrt{(\gamma_1\gamma_2)} \operatorname{Re} f_{12}(kr_{12}), \end{aligned} \right\} (2)$$

with

$$\begin{aligned} f_{12}(kr_{12}) &= - \left\{ [\hat{\mu}_1 \cdot \hat{\mu}_2 - (\hat{\mu}_1 \cdot \hat{r}_{12})(\hat{\mu}_2 \cdot \hat{r}_{12})] \frac{1}{kr_{12}} \right. \\ &\quad \left. + [\hat{\mu}_1 \cdot \hat{\mu}_2 - 3(\hat{\mu}_1 \cdot \hat{r}_{12})(\hat{\mu}_2 \cdot \hat{r}_{12})] \left[ \frac{i}{(kr_{12})^2} - \frac{1}{(kr_{12})^3} \right] \right\} \exp(ikr_{12}), \end{aligned} \quad (3)$$

where  $\hat{\mu}_i$  and  $\hat{r}_{12}$  are unit vectors along the transition dipole moment of atom  $i$  ( $i = 1, 2$ ) and the vector  $\mathbf{r}_{12} = \mathbf{r}_2 - \mathbf{r}_1$ , respectively ( $r_{12} = |\mathbf{r}_{12}|$ ).

When deriving equations (1), we have assumed that  $|\Delta_{12}| \ll \omega_0 = \frac{1}{2}(\omega_1 + \omega_2)$  and that the rotating-wave approximation (RWA) is valid. The atomic operators of equations (1) are slowly varying parts of the operators

$$\tilde{S}_{1,2}^{\pm}(t) = S_{1,2}^{\pm}(t) \exp(\pm i\omega_0 t), \quad (4)$$

where the mean frequency  $\omega_0 = \frac{1}{2}(\omega_1 + \omega_2)$  is used to eliminate the rapid oscillation of both atoms.

Equation (3) describing the retarded dipole-dipole interaction between the atoms is quite general. In our further calculations, we assume the two dipoles are parallel ( $\hat{\mu}_1 = \hat{\mu}_2$ ) and  $k = \omega_0/c$ . In particular, for identical atoms,  $\gamma_{12}$  and  $\Omega_{12}$  are the same as those obtained by Stephen [5] and others [1, 6-8].

Obviously, in a gas sample, there are pairs of atoms with various values and orientations of the interatomic distance  $\mathbf{r}_{12}$ , and  $f_{12}(kr_{12})$  should be averaged over all orientations and interatomic distances in order to be applicable to such a sample. To make our model as simple as possible and, at the same time, compatible with results reported already [12-14], we prefer, however, to deal with just two atoms separated by a well defined distance  $\mathbf{r}_{12}$ . In practical terms, this can mean two colour centres in a crystal lattice rather than a gas sample.

The field operator  $E_0^{(+)}(\mathbf{r}_i, t)$  in equations (1) denotes the positive-frequency part of the vacuum field and, in the transverse mode decomposition, is given by

$$E_0^{(+)}(\mathbf{r}_i, t) = \sum_q \left( \frac{2\pi\omega_q}{V} \right)^{1/2} \mathbf{e}_q a_q(0) \exp[i(\mathbf{q} \cdot \mathbf{r}_i - \omega_q t)], \quad (5)$$

where  $V$  is the normalization volume,  $\mathbf{e}_q$  the unit polarization vector and  $a_q$  the annihilation operator for the  $q$ th mode.

In the case when the radiation is initially in the vacuum state  $|\{0\}\rangle$ , we have  $E_0^{(+)}(\mathbf{r}_i, t)|\{0\}\rangle = 0$  and equations (1) lead to the vacuum expectation values for the atomic correlation functions.

For two non-identical atoms, equations (1) lead to a closed set of five equations of motion for the vacuum expectation values, which we can write in matrix form:

$$\frac{d}{dt} \mathbf{X} = \check{A} \mathbf{X}, \quad (6)$$

where  $\check{A}$  is a  $5 \times 5$  matrix:

$$\check{A} = \begin{bmatrix} -2\gamma_1 & 0 & -(\gamma_{12} + i\Omega_{12}) & -(\gamma_{12} - i\Omega_{12}) & 0 \\ 0 & -2\gamma_2 & -(\gamma_{12} - i\Omega_{12}) & -(\gamma_{12} + i\Omega_{12}) & 0 \\ -(\gamma_{12} + i\Omega_{12}) & -(\gamma_{12} - i\Omega_{12}) & -(\gamma_1 + \gamma_2 - 2i\Delta_{12}) & 0 & 4\gamma_{12} \\ -(\gamma_{12} - i\Omega_{12}) & -(\gamma_{12} + i\Omega_{12}) & 0 & -(\gamma_1 + \gamma_2 + 2i\Delta_{12}) & 4\gamma_{12} \\ 0 & 0 & 0 & 0 & -2(\gamma_1 + \gamma_2) \end{bmatrix} \quad (7)$$

The vector  $\mathbf{X}$  has the components

$$\left. \begin{aligned} X_1 &= \langle S_1^+ S_1^- \rangle, \\ X_2 &= \langle S_2^+ S_2^- \rangle, \\ X_3 &= \langle S_1^+ S_2^- \rangle, \\ X_4 &= \langle S_2^+ S_1^- \rangle, \\ X_5 &= \langle S_1^+ S_2^+ S_2^- S_1^- \rangle. \end{aligned} \right\} \quad (8)$$

As is obvious from equation (7), the fifth equation is decoupled from the remaining four. This allows for an exact solution of the above set of equations.

In the case when initially ( $t = 0$ ) atom 1 was in its excited state  $|2\rangle_1$  and atom 2 was in its ground state  $|1\rangle_2$ , the solutions of equation (6) are

$$\begin{aligned} \langle S_1^+(t) S_1^-(t) \rangle &= \exp [-(\gamma_1 + \gamma_2)t] \left\{ \frac{1}{2} \left( 1 - \frac{\Delta_{12}^2 + u_{12}^2}{B} \right) \cos Et \right. \\ &+ \frac{1}{2} \left( 1 + \frac{\Delta_{12}^2 + u_{12}^2}{B} \right) \cosh Dt + \frac{u_{12}E^2 - 4\Delta_{12}(\Delta_{12}u_{12} - \gamma_{12}\Omega_{12})}{2BE} \sin Et \\ &\left. + \frac{u_{12}D^2 + 4\Delta_{12}(\Delta_{12}u_{12} - \gamma_{12}\Omega_{12})}{2BD} \sinh Dt \right\}, \end{aligned} \quad (9)$$

$$\langle S_2^+(t) S_2^-(t) \rangle = \frac{1}{2} \frac{\gamma_{12}^2 + \Omega_{12}^2}{B} [\cosh Dt - \cos Et] \exp [-(\gamma_1 + \gamma_2)t], \quad (10)$$

$$\begin{aligned} \langle S_1^+(t) S_2^-(t) \rangle &= \frac{(\gamma_{12} + i\Omega_{12}) \exp [-(\gamma_1 + \gamma_2)t]}{4B} \left\{ 2(u_{12} + i\Delta_{12}) [\cos Et - \cosh Dt] \right. \\ &\left. - \frac{D^2 + 4i(u_{12}\Delta_{12} - \gamma_{12}\Omega_{12})}{D} \sinh Dt - \frac{E^2 - 4i(\Delta_{12}u_{12} - \gamma_{12}\Omega_{12})}{E} \sin Et \right\}, \end{aligned} \quad (11)$$

$$\langle S_2^+(t) S_1^-(t) \rangle = \langle S_1^+(t) S_2^-(t) \rangle^* \quad (12)$$

and  $\langle S_1^+(t) S_2^+(t) S_2^-(t) S_1^-(t) \rangle = 0$ .

In equations (9)–(12) we have introduced the notation

$$\left. \begin{aligned} u_{12} &= \frac{1}{2}(\gamma_2 - \gamma_1), \\ B &= [(\Delta_{12}^2 + \Omega_{12}^2 + \gamma_{12}^2 + u_{12}^2)^2 - 4(\Delta_{12}\gamma_{12} + u_{12}\Omega_{12})^2]^{1/2}, \\ D &= [-2(\Delta_{12}^2 + \Omega_{12}^2 - \gamma_{12}^2 - u_{12}^2) + 2B]^{1/2}, \\ E &= [2(\Delta_{12}^2 + \Omega_{12}^2 - \gamma_{12}^2 - u_{12}^2) + 2B]^{1/2}. \end{aligned} \right\} \quad (13)$$

The parameters  $u_{12}$ ,  $B$ ,  $D$  and  $E$  define 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, and the parameter  $E$  describes the frequency of the modulation. 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 of each other.

If the two atoms are identical ( $u_{12} = \Delta_{12} = 0$ ) we have  $B = \Omega_{12}^2 + \gamma_{12}^2$ ,  $D = 2\gamma_{12}$  and  $E = 2\Omega_{12}$ . In this case, it is clear from (9)–(12) that the probability of finding

atom 1 (2) in its excited state  $\langle S_1^+(t)S_1^-(t) \rangle$  ( $\langle S_2^+(t)S_2^-(t) \rangle$ ) oscillates with frequency  $E = 2\Omega_{12}$  in such a way that the oscillations cancel out if we take the sum of the two probabilities (see [1]). The sum of the interference terms  $\langle S_1^+(t)S_2^-(t) \rangle + \langle S_2^+(t)S_1^-(t) \rangle$  has no oscillating terms either. So, the populations of the individual atoms oscillate, whereas the total population decays monotonically. Owing to the dipole-dipole interaction, the population is transferred back and forth from one atom to the other. Here, the dipole-dipole interaction parameter  $\Omega_{12}$  plays a role similar to that of the Rabi frequency in the atom-field interaction. 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  $E_2 = 2\hbar\omega_0$  (see figure 1 (a)). The difference in energy between the states  $|\pm\rangle$  is precisely  $2\hbar\Omega_{12}$ . So the oscillations that appear in (9) and (10) for the case of two identical atoms are of a dynamical nature and can easily be explained in terms of the states and energies of the system as a whole.

If the atoms are non-identical, we have to use the general expressions (9)–(12) to describe the time-evolution of the system. The oscillations in (9) and (10) no longer cancel out mutually, and the sum of the interference terms (11) and (12) also exhibits oscillatory behaviour. This time-evolution is quite complicated and not easy to interpret. However, if we assume  $\Omega_{12} \simeq \Delta_{12} \gg \gamma_{12}, u_{12}$  (strong dipole-dipole interaction and a large difference between the atomic transition frequencies) we have  $B \approx \Omega_{12}^2 + \Delta_{12}^2$ ,  $D \approx 4(\gamma_{12}\Omega_{12} - u_{12}\Delta_{12})/E$  and  $E = 2\sqrt{(\Delta_{12}^2 + \Omega_{12}^2)}$ . One can readily check that in this case the oscillating terms in (9) and (10) cancel out approximately (the remaining terms are of order  $B^{-1}$ ). We now have a situation similar to that of identical atoms, except that the frequency of oscillations is now  $E = 2\sqrt{(\Delta_{12}^2 + \Omega_{12}^2)}$ . What clearly distinguishes this case from the previous case is the existence of the strong oscillating term  $-\frac{1}{2}[\Omega_{12}\Delta_{12}/(\Delta_{12}^2 + \Omega_{12}^2)] \cos Et$  in the interference terms (11) and (12). Thus, one can expect quantum beats to appear in the spontaneous emission from such a system. This is the subject of the next section. Here, we would like to note that if  $\Delta_{12} \neq 0$ , the states  $|\pm\rangle$  (the super-radiant and sub-radiant state) are no longer eigenstates of the two-atom system. The Hamiltonian of the system can be re-diagonalized including  $\Delta_{12}$ , giving the new eigenstates  $|\phi_{\pm}\rangle = C_1|\pm\rangle \pm C_2|\mp\rangle$  with the energies  $E_{\pm} = \hbar(\omega_0 \pm \sqrt{(\Delta_{12}^2 + \Omega_{12}^2)})$  as shown in figure 1 (b). The coefficients  $C_1$  and  $C_2$  are  $C_1 = \alpha/\sqrt{(\alpha^2 + \Delta_{12}^2)}$  and  $C_2 = -\Delta_{12}/\sqrt{(\alpha^2 + \Delta_{12}^2)}$ , where  $\alpha = \Omega_{12} + \sqrt{(\Delta_{12}^2 + \Omega_{12}^2)}$ . For an  $S^2$ -conserving system of two identical atoms the

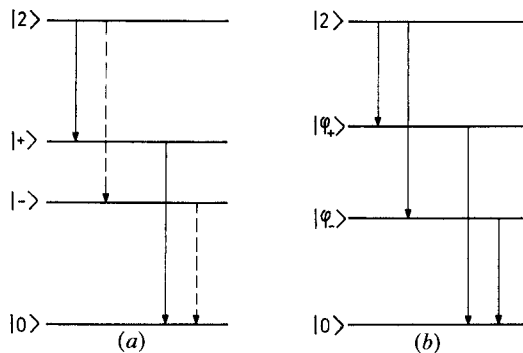


Figure 1. Energy levels and possible transitions for the two-atom system: (a) identical atoms; (b) non-identical atoms.

sub-radiant state  $|-\rangle$  is decoupled from the other states. Since both the states  $|\phi_{\pm}\rangle$  include the super-radiant state  $|+\rangle$ , the transitions from both states to the ground state are allowed. This is a typical configuration of levels for the production of quantum beats.

### 3. Total radiation rate

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

$$I(\mathbf{R}, t) = \frac{R^2 c}{2\pi\omega_0} \langle \mathbf{E}^{(-)}(\mathbf{R}, t) \cdot \mathbf{E}^{(+)}(\mathbf{R}, t) \rangle. \quad (14)$$

In equation (14) we have introduced the factor  $(R^2 c / 2\pi\omega_0)$  so that  $I(\mathbf{R}, t) d\Omega dt$  is the probability of finding one photon inside the solid-angle element  $d\Omega$  around the direction  $\mathbf{R}$  in the time interval  $dt$  at a time  $t$  in the far-field zone of the radiation emitted by the atomic system.

The positive frequency part of the field operator  $\mathbf{E}^{(+)}(\mathbf{R}, t)$  in the far zone  $R \gg r_{12}$ ,  $\lambda$  and  $t > R/c$  is given by the expression [1, 8, 15]

$$\mathbf{E}^{(+)}(\mathbf{R}, t) = \mathbf{E}_0^{(+)}(\mathbf{R}, t) - \left(\frac{\omega_0}{c}\right)^2 \sum_{i=1}^2 \frac{[\hat{R}_i \times (\hat{R}_i \times \boldsymbol{\mu}_i)]}{R_i} \exp(-ik\hat{R} \cdot \mathbf{r}_i) S_i^-(t). \quad (15)$$

Since the field is initially in the vacuum state, the vacuum part  $\mathbf{E}_0^{(+)}(\mathbf{R}, t)$  does not contribute to the expectation value of the normally ordered correlation operator in equation (14) and we obtain for  $I(\mathbf{R}, t)$  the expression

$$I(\mathbf{R}, t) = w(\mathbf{R}) \sum_{i,j=1}^2 2(\gamma_i \gamma_j)^{1/2} \langle S_i^+(t) S_j^-(t) \rangle \exp(ik\hat{R} \cdot \mathbf{r}_{ij}), \quad (16)$$

where

$$w(\mathbf{R}) = \frac{3 \sin^2 \Theta}{8\pi}, \quad (17)$$

with  $\Theta$  the angle between the observation direction  $\mathbf{R}$  and the atomic transition dipole moment  $\boldsymbol{\mu}$ .

On integrating over all solid angles  $d\Omega$ , equation (16) yields the total radiation rate  $I(t)$  given in photons per second:

$$I(t) = \sum_{i,j=1}^2 2\gamma_{ij} \langle S_i^+(t) S_j^-(t) \rangle, \quad (18)$$

where  $\gamma_{ij}$  is given by equation (2). Using (9)–(12), we find that for two non-identical atoms the total radiation rate has the form

$$\begin{aligned} I(t) = \exp[-(\gamma_1 + \gamma_2)t] & \left\{ \left[ \gamma_1 - \frac{\gamma_1(u_{12}^2 + \gamma_{12}^2 + \Delta_{12}^2) + 2\gamma_{12}\Delta_{12}\Omega_{12} + \gamma_2\Omega_{12}^2}{B} \right] \cos Et \right. \\ & + \left[ \gamma_1 + \frac{\gamma_1(u_{12}^2 + \gamma_{12}^2 + \Delta_{12}^2) + 2\gamma_{12}\Delta_{12}\Omega_{12} + \gamma_2\Omega_{12}^2}{B} \right] \cosh Dt \\ & + \frac{(\gamma_1 u_{12} - \gamma_{12}^2)E^2 + 4(\gamma_1 \Delta_{12} + \gamma_{12}\Omega_{12})(\gamma_{12}\Omega_{12} - u_{12}\Delta_{12})}{BE} \sin Et \\ & \left. + \frac{(\gamma_1 u_{12} - \gamma_{12}^2)D^2 - 4(\gamma_1 \Delta_{12} + \gamma_{12}\Omega_{12})(\gamma_{12}\Omega_{12} - u_{12}\Delta_{12})}{BD} \sinh Dt \right\}. \quad (19) \end{aligned}$$

First of all, we shall discuss the solution (19) for some limiting cases of interatomic interactions and differences between the atomic transition frequencies.

3.1. *The case  $\Omega_{12} \gg \Delta_{12}, \gamma_{12}, u_{12}$  of strong interatomic interaction*

In this case the interatomic separation is very small in comparison to the resonant wavelength and the differences between the transition frequencies and natural linewidths of the atoms are small, so that equations (13) give  $B \approx \Omega_{12}^2, D \approx 2\gamma_{12}, E \approx 2\Omega_{12}$ , and from equation (19) we have

$$I(t) = \exp [-(\gamma_1 + \gamma_2)t] \{ (\gamma_1 - \gamma_2) \cos 2\Omega_{12}t + (\gamma_1 + \gamma_2) \cosh 2\gamma_{12}t - 2\gamma_{12} \sinh 2\gamma_{12}t \}. \tag{20}$$

Equation (20) shows smooth exponential decay with the decay rates  $\gamma_1 + \gamma_2 \pm 2\gamma_{12}$  and an oscillating term with amplitude proportional to the difference  $\gamma_1 - \gamma_2$  of the natural linewidths of the two atoms. The oscillating term in (20) appears because the atoms lose their populations owing to spontaneous emission at different rates and the dynamical terms describing population transfer between the atoms due to the dipole-dipole interaction cannot cancel out completely, as was the case for identical atoms. For identical atoms we have  $\mu_1 = \mu_2$ , and the transition amplitude from the sub-radiant state  $|-\rangle$  to the ground state  $|0\rangle$  is equal to zero, and only the super-radiant state  $|+\rangle$  can radiate. We now have a single transition in the system, and there are no quantum beats. The situation changes if the two atoms are not identical ( $\mu_1 \neq \mu_2$ ). There is a non-zero transition amplitude  $|-\rangle \rightarrow |0\rangle$ , and both the super-radiant state  $|+\rangle$  and sub-radiant state  $|-\rangle$  decay to the common ground state  $|0\rangle$ . The interference between the two amplitudes gives the quantum beat term in (20). As expected, the beat frequency is  $2\Omega_{12}$ , reflecting the energy difference between the two states. For the initial conditions used by us in this paper (atom 1 excited at  $t = 0$ ), the initial population distributes equally between the states  $|+\rangle$  and  $|-\rangle$ . As the channel  $|-\rangle \rightarrow |0\rangle$  opens, one can expect an increase in the radiation rate. It is seen from (20) that, for  $\gamma_2 > \gamma_1, I(t)$  can actually become greater than its initial value  $I(0)$ . This 'super-radiant' behaviour is absent in the case of identical atoms. Since equation (20) holds also for  $\Delta_{12} = 0$ , the effect is due solely to the difference in atomic transition dipole moments leading to different atomic linewidths. The interatomic terms (11) and (12) do not contribute to the beats in this case.

3.2. *The case  $\Omega_{12} \approx \Delta_{12} \gg \gamma_{12}, u_{12}$  of strong interatomic interaction and great difference between the atomic transition frequencies*

Here, according to equations (13),  $B \approx \Delta_{12}^2 + \Omega_{12}^2, D \approx 4(\gamma_{12}\Omega_{12} - u_{12}\Delta_{12})/E, E \approx 2\sqrt{(\Delta_{12}^2 + \Omega_{12}^2)}$ , so the total radiation rate takes the form

$$I(t) = \frac{\exp [-(\gamma_1 + \gamma_2)t]}{\Delta_{12}^2 + \Omega_{12}^2} \left\{ [(\gamma_1 - \gamma_2)\Omega_{12}^2 - 2\Delta_{12}\gamma_{12}\Omega_{12}] \cos 2\sqrt{(\Delta_{12}^2 + \Omega_{12}^2)}t + [(\gamma_1 + \gamma_2)\Omega_{12}^2 + 2\Delta_{12}(\gamma_1\Delta_{12} + \gamma_{12}\Omega_{12})] \cosh \frac{2(\gamma_{12}\Omega_{12} - u_{12}\Delta_{12})t}{\sqrt{(\Delta_{12}^2 + \Omega_{12}^2)}} - 2\sqrt{(\Delta_{12}^2 + \Omega_{12}^2)}(\gamma_{12}\Omega_{12} + \Delta_{12}\gamma_1) \sinh \frac{2(\gamma_{12}\Omega_{12} - u_{12}\Delta_{12})t}{\sqrt{(\Delta_{12}^2 + \Omega_{12}^2)}} \right\}. \tag{21}$$

In this case the total radiation rate exhibits sinusoidal modulation (beats) the amplitude of which consists of two terms: a term proportional to  $\gamma_1 - \gamma_2$  with exactly the same origin as that discussed previously, and a new term that stems from the interatomic correlation functions (11) and (12). It describes typical quantum beats; the transition amplitudes from the states  $|\phi_{\pm}\rangle$  to the ground state  $|0\rangle$  interfere. On putting  $\Delta_{12} = 0$ , equation (21) goes over into (20). Equation (21) indicates the two possible mechanisms for the appearance of quantum beats in the system comprising two non-identical atoms. Again, the cooperative behaviour leading to the increase in total radiation rate above its initial value can be observed in (21).

### 3.3. The case $\Delta_{12} \gg \Omega_{12}$ , $\gamma_{12}$ , $u_{12}$ of great difference between the atomic transition frequencies and weak interatomic interaction

In this case, equations (13) lead to  $B \approx \Delta_{12}^2$ ,  $D \approx 2u_{12}$ ,  $E \approx 2\Delta_{12}$ , and  $I(t)$  takes the form

$$I(t) = 2\gamma_1 \exp(-2\gamma_1 t). \quad (22)$$

Here, we have a simple exponential decay for the initially excited atom 1, without the influence of the other atom.

For the general case, we have plotted the total radiation rate  $I(t)$  given by equation (19) in figures 2–4 for several values of  $\gamma_2/\gamma_1$ ,  $\Delta_{12}$  and different interatomic separations. In the case of non-identical atoms we find a pronounced sinusoidal modulation superimposed on the smooth exponential decay. The frequency of this modulation is dependent on the interatomic interaction and its amplitude vanishes for large interatomic separations (figure 3). This oscillatory behaviour of the total radiation rate is in fact the quantum-beat effect that arises because of interference between the two possible transition amplitudes ( $|\pm\rangle$  or  $|\phi_{\pm}\rangle \rightarrow |0\rangle$ ) that contribute to the spontaneous emission. Two possible mechanisms leading to these quantum beats have been discussed earlier (§§ 3.1 and 3.2). The first mechanism, consisting in

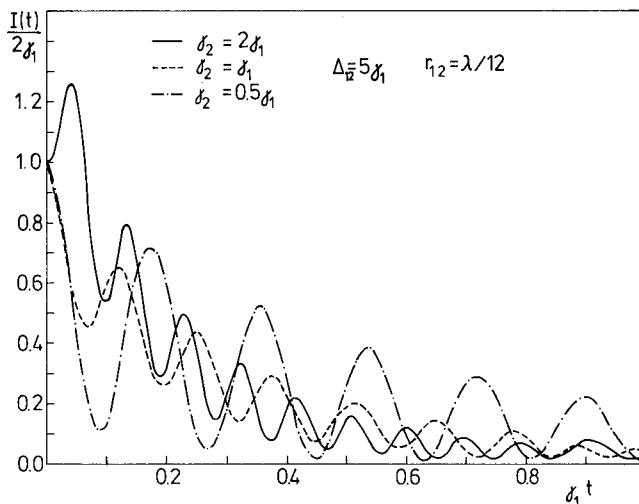


Figure 2. Normalized total radiation rate versus  $\gamma_1 t$  for interatomic separation  $r_{12} = \lambda/12$ ,  $\Delta_{12} = 5\gamma_1$ , and different natural linewidths.



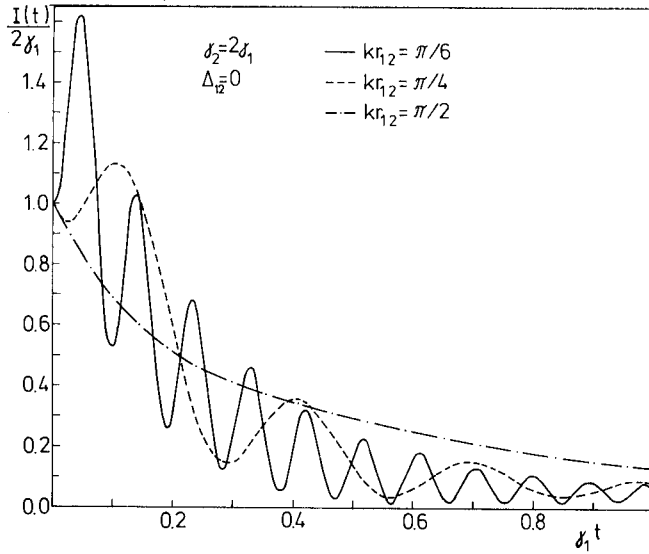


Figure 3. Normalized total radiation rate as a function of time  $\gamma_1 t$  for  $\Delta_{12} = 0$ ,  $\gamma_2 = 2\gamma_1$ , and different interatomic separations.

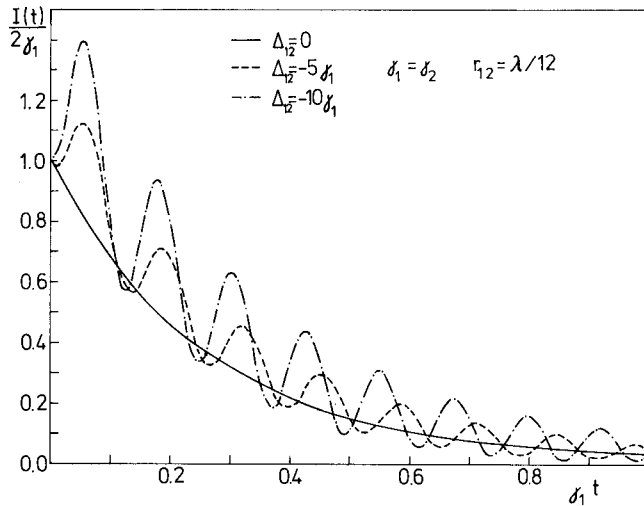


Figure 4. Normalized total radiation rate versus  $\gamma_1 t$  for two atoms with the same natural linewidths ( $\gamma_1 = \gamma_2$ ), the interatomic separation  $r_{12} = \lambda/12$ , and different transition frequencies.

the opening of the channel  $|-\rangle \rightarrow |0\rangle$  if the transition dipole moments (or linewidths) of the two atoms are different, is illustrated in figure 3. The second mechanism, consisting in a mixing of the states  $|+\rangle$  and  $|-\rangle$  for  $\Delta_{12} \neq 0$  leading to new states  $|\phi_+\rangle$  and  $|\phi_-\rangle$ , both of which are coupled to the ground state  $|0\rangle$ , is illustrated in figure 4. If the interatomic separation is small and  $\gamma_2 > \gamma_1$ , the total radiation rate becomes greater than its initial value for a definite period of time. This means that the emission is then super-radiant. This super-radiant behaviour, which is absent in the case of two identical atoms ( $\gamma_2 = \gamma_1$ ,  $\Delta_{12} = 0$ ), is quite apparent in the graphs. The

collective effects decrease with increasing distance between the atoms, as shown in figure 3. The emission can be super-radiant also in the case when the atoms have the same natural linewidths. As is shown in figure 4, this takes place for large differences between the atomic transition frequencies only (see §3.1).

The quantum beat effect has recently been considered with regard to numerous optical processes ([16–17] and references therein).

#### 4. Population trapping effect

The energy of each atom is described by the operator  $S_i^z$  ( $i = 1, 2$ ) which, with the spin operators  $S_i^\pm$ , fulfils the well-known commutation relations

$$[S_i^+, S_j^-] = 2S_i^z \delta_{ij}, \quad [S_i^+, S_j^-]_+ = \delta_{ij}. \quad (23)$$

Using the above commutation and anti-commutation relations for spin operators we can express  $\langle S_i^z \rangle$  by the correlation function  $\langle S_i^+ S_i^- \rangle$  as  $\langle S_i^z \rangle = \langle S_i^+ S_i^- \rangle - \frac{1}{2}$ .

For spontaneous emission in the absence of external fields in the steady state ( $t \rightarrow \infty$ ) both atoms should be in their ground states. This means that in the steady state  $\langle S_1^z(\infty) \rangle = \langle S_2^z(\infty) \rangle = -\frac{1}{2}$ , and we have zero solutions for  $\langle S_1^+ S_1^- \rangle$  and  $\langle S_2^+ S_2^- \rangle$ . The existence of zero solutions in the steady state requires, by equation (6), that the determinant of the matrix  $\check{A}$  shall differ from zero. If the determinant of the matrix  $\check{A}$  is zero we have non-zero steady-state solutions for  $\langle S_1^+ S_1^- \rangle$  and  $\langle S_2^+ S_2^- \rangle$ . This signifies that part of the energy has been trapped in the atomic system.

The determinant of the matrix  $\check{A}$  is zero if  $D = \gamma_1 + \gamma_2$ . For two identical atoms ( $\Delta_{12} = u_{12} = 0$ ) this condition is fulfilled if the atoms are separated by a distance much smaller than the resonance wavelength. This condition has been calculated by Agarwal [1] and recently has been extensively discussed by Dalton and Knight [18].

For two non-identical atoms the condition for population trapping  $D = \gamma_1 + \gamma_2$  is fulfilled if  $\Delta_{12}\gamma_{12} = -u_{12}\Omega_{12}$  and if the atoms are separated by a distance much smaller than the resonant wavelength.

We note that for population trapping in spontaneous emission from two interacting atoms it is necessary to confine both atoms in a region much smaller than the resonant wavelength. In the case of population trapping for two identical atoms, if initially only one atom is inverted, equations (9) and (10) lead to

$$\langle S_1^z(\infty) \rangle = \langle S_2^z(\infty) \rangle = -\frac{1}{4}, \quad (24)$$

whereas for non-identical atoms

$$\langle S_1^z(\infty) \rangle = -\frac{1}{2} + \frac{\gamma_2^2}{(\gamma_1 + \gamma_2)^2}, \quad (25 a)$$

$$\langle S_2^z(\infty) \rangle = -\frac{1}{2} + \frac{\gamma_1\gamma_2}{(\gamma_1 + \gamma_2)^2}. \quad (25 b)$$

For identical atoms in the steady state, their energies are identical, whereas for non-identical atoms their energies are different, and in both cases are different from the energy of their ground states.

The total population that can be trapped in such a system is given by the steady-state solutions

$$\langle S_1^+ S_1^- \rangle + \langle S_2^+ S_2^- \rangle = \frac{\gamma_2}{\gamma_1 + \gamma_2}. \quad (26)$$

In the case of identical atoms the amount trapped is exactly one-half of the initial population. If the atoms are not identical, the amount of population that is trapped depends strongly on the rate  $\gamma_2/\gamma_1$  of the damping constants of the two atoms. It is close to zero for  $\gamma_2/\gamma_1 \ll 1$  and is close to unity for  $\gamma_2/\gamma_1 \gg 1$ . It should be emphasized, however, that the necessary condition for trapping is  $\gamma_{12} = \sqrt{\gamma_1\gamma_2}$ —the condition which is satisfied for small interatomic distances only. Otherwise there is no population trapping at all.

## 5. Conclusions

We have considered the problem of spontaneous emission from two non-identical atoms, when initially only one atom is inverted. Our calculations show that in this case, contrary to that of identical atoms, the total radiation rate exhibits oscillatory behaviour due to the quantum-beat effect. The two possible mechanisms leading to the beats are discussed in detail.

We have found that for small interatomic separation, i.e. for strong interatomic interaction, the total radiation rate can become greater than its initial value for a definite (short) interval of time. This super-radiant behaviour means that non-identical atoms may, in a sense, radiate more collectively than identical atoms. This may seem somewhat strange at a first glance, but can be understood if one keeps in mind that for the initial conditions used in this paper (only atom 1 excited at  $t = 0$ ) one-half of the initial population is stored in the sub-radiant state  $|-\rangle$ . This state does not radiate if the atoms are identical, and becomes active if the two atoms are not identical. The interference between the transition amplitudes from the super-radiant ( $|+\rangle$ ) and sub-radiant ( $|-\rangle$ ) states, giving the quantum-beat effect, can also lead to the increase in  $I(t)$  above its initial value. It takes place for certain values of the parameters  $\gamma_1$ ,  $\gamma_2$  and  $\Delta_{12}$ . One should emphasize, however, that this effect does not violate the energy conservation law. The total energy stored in the atoms, which for  $\Delta \ll \omega_0$  is equal to  $\hbar\omega_0(\langle S_1^+(t)S_1^-(t) \rangle + \langle S_2^+(t)S_2^-(t) \rangle)$  decays monotonically in time, as can easily be checked using equations (9) and (10) (see figure 5).

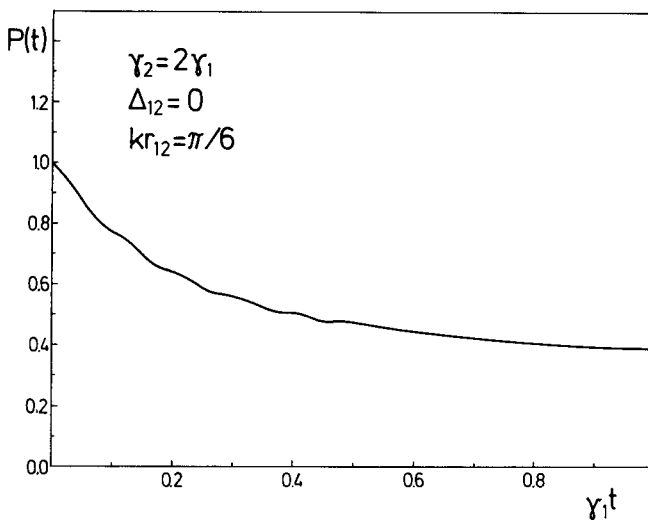


Figure 5. Total population  $P(t) = \langle S_1^+(t)S_1^-(t) \rangle + \langle S_2^+(t)S_2^-(t) \rangle$  versus  $\gamma_1 t$ , which, multiplied by  $\hbar\omega_0$ , expresses the total energy stored in the atoms.

The situation changes if both atoms are initially excited. For identical atoms the state  $|-\rangle$  will never be populated, and we have downward transitions via the  $|+\rangle$  state only. For non-identical atoms the sub-radiant state acquires some population from the state  $|2\rangle$  as time elapses. However, at the initial stage of evolution we have a system with the common upper level  $|2\rangle$  and transitions to the states  $|\pm\rangle$ . This configuration does not give quantum beats. Our calculations show that the total radiation rate decays monotonically for the same values of parameters used by us in this paper. Some weak beats can appear for very different atoms, but we never get an increase in  $I(t)$ . So, the cooperative behaviour of the system is manifestly weaker when both atoms are initially excited.

We have also shown that the effect of population trapping may occur in the system of two non-identical atoms, i.e. in the steady state the atoms have energies different from the energy of their ground states. This can take place, however, only if the distance between the atoms is much smaller than the resonant wavelength.

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