Quantum beats in intensity correlations of spontaneous emission from two non-identical atoms†

Z. FICEK, R. TANAŚ and S. KIELICH

Nonlinear Optics Division, Institute of Physics,
Adam Mickiewicz University, 60-780 Poznań, Poland

Abstract. The two-time second-order correlation function of the electric field spontaneously emitted by a system of two non-identical two-level atoms with different transition frequencies and different natural linewidths is calculated from the viewpoint of quantum beats and photon anticorrelation effects. Our analytical solution shows that the intensity correlation exhibits quantum beats. The quantum beats are shown to appear even if the two atoms are independent. For identical atoms, beats appear only if dipole–dipole interaction between the atoms is included, and they are then strongly dependent on the direction of observation with respect to the line connecting the two atoms. The anticorrelation effect between the photons emitted in different directions is also discussed. It is shown that for a single-time correlation function the condition for anticorrelation is independent of interatomic interactions. For a two-time correlation function, interatomic interactions reduce the anticorrelation effect.

1. Introduction

The subject of quantum beats, or quantum-interference effects, has already been discussed in a famous paper by Breit [1] and has recently become an important method of studying atomic spectra and the various quantum mechanical interactions that lead to different types of splitting and shifts of the energy levels. This phenomenon results from the interference between two transition amplitudes to a common lower level.

Another interesting problem is the interference between two independent beams with different frequencies. Light beating resulting from the superposition of beams from two independent sources has been demonstrated by Javan *et al.* [2] and Lipsett and Mandel [3]. Mandel [4] has shown that second-order correlation functions of light emitted by independent sources exhibit a sinusoidal dependence on the space and time variables. This has been recently verified in experiments by Vainshtein *et al.* [5].

The presence or absence of beats in the emission of atoms has attracted considerable attention [6–10]. The discussion has centred around two types of atoms: one in which the transitions occur between each of two or more closely spaced upper levels and a single lower level (type I), and another in which the transitions occur between a common upper level and each of two or more closely spaced lower

levels (type II). A qualitative difference in the theoretical prediction of beats in spontaneous emission from the two types of atoms was noted by Breit [1]: if the upper states of a type I atom are coherently excited, quantum beats in the fluorescence intensity are predicted; on the other hand, no beats are predicted from excitation and decay of type II atoms. This difference has been the subject of numerous experiments [11].

Recently, Zajonc [12] has pointed out that for single-atom emission, beats are present in the second-order correlation function of the light emitted by type II atoms. Chow *et al.* [6], Herman *et al.* [7], Senitzky [8], and Agarwal *et al.* [10] have discussed the presence or absence of beats in many-atom systems, showing that beats are present in the intensity emitted by type I as well as type II atoms. However, the atoms must radiate cooperatively in the case of type II atoms. These beats arise from the interference of the fields emitted by a spatially random N-atom gas, and has been observed by Vrehen *et al.* [13] in superfluorescence, i.e. in the cooperative emission of an initially inverted system. Gross *et al.* [14] have reported beats arising from interference between the light emitted by two groups of atoms with different velocities. One should point out that in many-atom systems beats are present for three- and more-level systems but are not present for identical two-level atoms [15]. Varfolomeev [16] and Milonni and Knight [17] considered the spontaneous decay probability for a system of two non-identical atoms and have shown that, in special cases, it exhibits beats.

Recently, we have studied spontaneous emission from two identical as well as non-identical atoms from the viewpoint of quantum beats and super-radiant effects [18]. We have shown that for two non-identical atoms the total radiation intensity in the case when initially only one atom was excited exhibits beats. These beats vanish for non-interacting atoms as well as for identical atoms. Moreover, beats are not present in the case of an initially fully inverted system (both atoms excited). This is easily understood within the framework of collective states of a two-atom system. The Hamiltonian for a two-atom system can be diagonalized [19–21] including the dipole–dipole interaction, giving the eigenstates

$$|0\rangle = |1\rangle_1 |1\rangle_2, \ |\pm\rangle = (1/\sqrt{2})(|2\rangle_1 |1\rangle_2 \pm |1\rangle_1 |2\rangle_2) \text{ and } |2\rangle = |2\rangle_1 |2\rangle_2$$

with energies $E_0 = 0$, $E_{\pm} = \hbar(\omega_0 \pm \Omega_{12})$ and $E_2 = 2\hbar\omega_0$, where ω_0 is the transition frequency between the lower state $|1\rangle_i$ and the excited state $|2\rangle_i$ of the ith atom ($i = 1, 2$) and Ω_{12} is the dipole–dipole interaction potential [21]. In fact, the two-atom system is equivalent to a single four-level system with one upper state, one ground state and two intermediate states (figure 1 (*a*)). For identical atoms, the transition amplitude from the subradiant state $|-\rangle$ to the ground state $|0\rangle$ is very small, and only the super-radiant state $|+\rangle$ can radiate [20]: so, there are no quantum beats. For non-identical atoms, the states $|\pm\rangle$ are no longer eigenstates of the two-atom system. The Hamiltonian of the system can be rediagonalized [18] introducing eigenstates $|\phi_{\pm}\rangle$ which include the super-radiant state $|+\rangle$ and subradiant state $|-\rangle$ (figure 1 (*b*)). The transitions from the states $|\phi_{\pm}\rangle$ to the ground state $|0\rangle$ are allowed. For initially only one atom excited, the initial population distributes equally between the states $|+\rangle$ and $|-\rangle$. Since both channels $|\phi_{\pm}\rangle \rightarrow |0\rangle$ are open, the interference between the two amplitudes gives quantum beats. This system is equivalent to a type I system. If initially both atoms are excited we have the entire initial population in the upper state $|2\rangle$, and transitions $|2\rangle \rightarrow |\phi_{\pm}\rangle$ do not give quantum beats because we have only one upper state (type II).
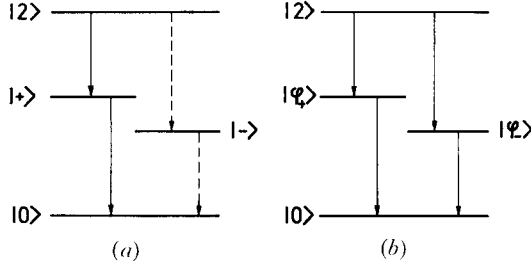
Figure 1.   Energy level diagrams for two-atom system showing the permitted transitions, in the case of (*a*) identical atoms and (*b*) non-identical atoms.

In the present paper we investigate the two-time second-order correlation function for spontaneous emission from a fully inverted two-atom system (type II). As we shall see later on, quantum beats are present in the second-order correlation function of light radiated by a system of two identical as well as a system of two non-identical atoms. We start from the equations of motion for the atomic operators assuming that the atoms have different transition frequencies and different natural linewidths. The time-evolution of the atomic variables is dependent on the parameters Δ_{12} and u_{12} which describe the difference between the atomic energies and that between their natural linewidths, respectively. This time-evolution is also dependent on the collective parameters γ_{12} and Ω_{12} describing 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 two-atom system. We apply the solution of these equations of motion to derive the second-order correlation function.

2.   Intensity correlations

The aim of this paper is to calculate the second-order two-time correlation function (intensity correlations):

$$G^{(2)}(\mathbf{R}_1, t_1; \mathbf{R}_2, t_2)$$

$$= \left(\frac{R^2 c}{2\pi\omega_0}\right)^2 \langle E^{(-)}(\mathbf{R}_1, t_1)E^{(-)}(\mathbf{R}_2, t_2)E^{(+)}(\mathbf{R}_2, t_2)E^{(+)}(\mathbf{R}_1, t_1)\rangle, \quad (1)$$

for the collective spontaneous emission from two non-identical atoms, separated by r_{12}, coupled to each other via retarded dipole–dipole near-field interaction and to all modes of the electromagnetic field, which are assumed to be initially in their vacuum state $|\{0\}\rangle$. 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 ω_1 and ω_2, respectively, and the corresponding natural linewidths $2\gamma_1$ and $2\gamma_2$.

In (1), we have introduced the factor $(R^2 c / 2\pi\omega_0)^2$ so that

$$G^{(2)}(\mathbf{R}_1, t_1; \mathbf{R}_2, t_2)\, \mathrm{d}\Omega_{\hat{R}_1}\, \mathrm{d}\Omega_{\hat{R}_2}\, \mathrm{d}t_1\, \mathrm{d}t_2$$

is the probability of finding one photon inside the solid angle element $\mathrm{d}\Omega_{\hat{R}_1}$ around the direction $\mathbf{R}_1$ in the time interval $\mathrm{d}t_1$ at the time t_1 and another photon inside the solid angle element $\mathrm{d}\Omega_{\hat{R}_2}$ around the direction $\mathbf{R}_2$ in the time interval $\mathrm{d}t_2$ at the moment of time t_2.

The positive frequency part of the field operator $E^{(+)}(\mathbf{R}, t)$ in the far-field zone

$$|\mathbf{R}_1| = |\mathbf{R}_2| = R \gg \lambda, r_{12},$$

where λ is the resonant wavelength, and for $t > R/c$, is given by the expression [18–20]

$$E^{(+)}(\mathbf{R}, t) = E_0^{(+)}(\mathbf{R}, t) - k^2 \sum_{i=1}^{2} \frac{\hat{R} \times (\hat{R} \times \boldsymbol{\mu}_i)}{R} S_i^- \left(t - \frac{R}{c} \right) \exp\left(-ik\hat{R} \cdot \mathbf{r}_i \right), \quad (2)$$

where $k = \omega_0/c$; $\omega_0 = \frac{1}{2}(\omega_1 + \omega_2)$; $\boldsymbol{\mu}_i$ is the atomic transition dipole moment; and S_i^+ and $S_i^- = (S_i^+)^+$ are operators that raise and lower the energy of atom i.

Since the field is initially in the vacuum state, the vacuum part $E_0^{(+)}(\mathbf{R}, t)$ does not contribute to the expectation values of the normally ordered correlation operator in equation (1), and we obtain for $G^{(2)}(\mathbf{R}_1, t_1; \mathbf{R}_2, t_2)$ the expression

$$G^{(2)}(\mathbf{R}_1, t_1; \mathbf{R}_2, t_2) = u(\hat{R}_1) u(\hat{R}_2) \sum_{i,j,k,l=1}^{2} (\gamma_i \gamma_j \gamma_k \gamma_l)^{1/2}$$

$$\times \langle S_i^+(t_1) S_k^+(t_2) S_l^-(t_2) S_j^-(t_1) \rangle \exp[ik(\hat{R}_1 \cdot \mathbf{r}_{ij} + \hat{R}_2 \cdot \mathbf{r}_{kl})], \quad (3)$$

where

$$u(\hat{R}) = \frac{3}{8\pi} \sin^2 \Theta, \quad (4)$$

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

According to equation (3), in order to study the intensity correlation we have to find the atomic correlation functions. To this aim we have used the equations of motion for the atomic operators described by Lehmberg's [20] master equation generalized to the case of non-identical atoms [18]

$$\frac{\mathrm{d}}{\mathrm{d}t} S_i^\pm(t) = -(\gamma_i \mp i\Delta_i) S_i^\pm(t) - (\gamma_{ij} \mp i\Omega_{ij}) S_j^\pm(t)$$

$$+ 2(\gamma_{ij} \mp i\Omega_{ij}) S_i^+(t) S_i^\pm(t) S_i^+(t), \quad i \neq j \quad (5)$$

where $\Delta_1 = -\Delta_2 = \Delta_{12} = \frac{1}{2}(\omega_1 - \omega_2)$, and $2\gamma_1$ and $2\gamma_2$, are the Einstein A coefficients for the atoms 1 and 2, respectively. The collective parameters Ω_{ij} and γ_{ij} arising from the retarded dipole–dipole and radiative interaction between the atoms, depend on the interatomic distance r_{12}, and are defined as [18–21]

$$\Omega_{ij} = \frac{3}{2}\sqrt{(\gamma_i \gamma_j)} \left\{ -[(\hat{\mu}_i \cdot \hat{\mu}_j) - (\hat{\mu}_i \cdot \hat{r}_{ij})(\hat{\mu}_j \cdot \hat{r}_{ij})] \frac{\cos(kr_{ij})}{kr_{ij}} \right.$$

$$\left. + [(\hat{\mu}_i \cdot \hat{\mu}_j) - 3(\hat{\mu}_i \cdot \hat{r}_{ij})(\hat{\mu}_j \cdot \hat{r}_{ij})] \left[\frac{\sin(kr_{ij})}{(kr_{ij})^2} + \frac{\cos(kr_{ij})}{(kr_{ij})^3} \right] \right\}, \quad (6)$$

$$\gamma_{ij} = \frac{3}{2}\sqrt{(\gamma_i \gamma_j)} \left\{ [(\hat{\mu}_i \cdot \hat{\mu}_j) - (\hat{\mu}_i \cdot \hat{r}_{ij})(\hat{\mu}_j \cdot \hat{r}_{ij})] \frac{\sin(kr_{ij})}{kr_{ij}} \right.$$

$$\left. + [(\hat{\mu}_i \cdot \hat{\mu}_j) - 3(\hat{\mu}_i \cdot \hat{r}_{ij})(\hat{\mu}_j \cdot \hat{r}_{ij})] \left[\frac{\cos(kr_{ij})}{(kr_{ij})^2} - \frac{\sin(kr_{ij})}{(kr_{ij})^3} \right] \right\}, \quad (7)$$

where $\hat{\mu}$ and $\hat{r}_{12}$ are unit vectors along the transition electric dipole moment and the vector $\mathbf{r}_{12} = \mathbf{r}_2 - \mathbf{r}_1$, respectively. When deriving equations (5), we have assumed that

the system is Markovian. Moreover, we have assumed that $|\Delta| \ll \omega_0$ and that the rotating-wave approximation (RWA) is valid [22, 23]. The Markovian approximation used in deriving these equations has been discussed by Milonni and Knight [24].

For two non-identical atoms, equations (5) lead to a closed set of four equations of motion for the vacuum expectation values: $\langle S_1^+(t)S_1^-(t) \rangle$, $\langle S_2^+(t)S_2^-(t) \rangle$, $\langle S_1^+(t)S_2^-(t) \rangle$, $\langle S_2^+(t)S_1^-(t) \rangle$, and one equation, decoupled from the remaining four, for $\langle S_1^+(t)S_2^+(t)S_2^-(t)S_1^-(t) \rangle$.

Introducing the Laplace transform

$$F(p) = \mathscr{L}(F(t)) = \int_0^\infty F(t) \exp(-pt)\, \mathrm{d}t, \tag{8}$$

this closed set of equations of motion transforms into an easily solvable system of algebraic equations in transformed variables. The solution can be written as:

$$\mathbf{X}(t) = \mathscr{L}^{-1}\left\{ \breve{A}^{-1}(p)\mathbf{X}(0) + \frac{4\gamma_{12}\rho(0)\mathbf{B}(p)}{(p + 2\gamma_1 + 2\gamma_2)\det \breve{A}(p)} \right\}, \tag{9}$$

and the remaining equation for $\langle S_1^+(t)S_2^+(t)S_2^-(t)S_1^-(t) \rangle$ has the simple solution:

$$\langle S_1^+(t)S_2^+(t)S_2^-(t)S_1^-(t) \rangle = \rho(0)\exp[-2(\gamma_1 + \gamma_2)t], \tag{10}$$

where $\rho(0) = \langle S_1^+(0)S_2^+(0)S_2^-(0)S_1^-(0) \rangle$ is dependent on the initial population of the excited states of the atoms. If both atoms are initially in their excited states, then $\rho(0) = 1$, whereas in other cases $\rho(0) = 0$.

In equation (9), $\mathbf{X}(t)$ and $\mathbf{B}(p)$ are column vectors, defined as

$$\mathbf{X} = \mathrm{col}\,(\langle S_1^+ S_1^- \rangle, \langle S_2^+ S_2^- \rangle, \langle S_1^+ S_2^- \rangle, \langle S_2^+ S_1^- \rangle), \tag{11}$$

$$\mathbf{B} = \mathrm{col}\,(B_1, B_2, B_3, B_3^*), \tag{12}$$

where

$$\left.\begin{aligned}
B_1 &= -[2\gamma_{12}z^2 + 4(\gamma_{12}u_{12} - \Delta_{12}\Omega_{12})z + 8\Omega_{12}(\gamma_{12}\Omega_{12} - \Delta_{12}u_{12})], \\
B_2 &= -[2\gamma_{12}z^2 - 4(\gamma_{12}u_{12} - \Delta_{12}\Omega_{12})z + 8\Omega_{12}(\gamma_{12}\Omega_{12} - \Delta_{12}u_{12})], \\
B_3 &= [z^3 + 2\mathrm{i}\Delta_{12}z^2 + 4(\Omega_{12}^2 - u_{12}^2)z + 8\mathrm{i}u_{12}(\gamma_{12}\Omega_{12} - \Delta_{12}u_{12})],
\end{aligned}\right\} \tag{12 a}$$

with $z = (p + \gamma_1 + \gamma_2)$ and $u_{12} = \frac{1}{2}(\gamma_2 - \gamma_1)$, and $\breve{A}(p)$ is a 4×4 matrix with the elements

$$\left.\begin{aligned}
A_{11} &= (z - 2u_{12}), & A_{12} &= A_{21} = A_{34} = A_{43} = 0, \\
A_{22} &= (z + 2u_{12}), & A_{33} &= A_{44}^* = (z - 2\mathrm{i}\Delta_{12}), \\
A_{13} &= A_{14}^* = A_{23}^* = A_{24} = A_{31} = A_{32}^* = A_{41}^* = A_{42} = (\gamma_{12} + \mathrm{i}\Omega_{12}).
\end{aligned}\right\} \tag{13}$$

For further calculations one needs information about the zeros of $\det \breve{A}(p)$, which is a biquadratic polynomial in p:

$$\det \breve{A}(p) = z^4 + 4(\Delta_{12}^2 + \Omega_{12}^2 - \gamma_{12}^2 - u_{12}^2)z^2 - 16(\gamma_{12}\Omega_{12} - \Delta_{12}u_{12}). \tag{14}$$

This biquadratic equation (14) has two real roots and two complex conjugate roots:

$$\left.\begin{aligned}
p_{1,2} &= -(\gamma_1 + \gamma_2) \pm D, \\
p_{3,4} &= -(\gamma_1 + \gamma_2) \pm \mathrm{i}E,
\end{aligned}\right\} \tag{15}$$

where we have introduced the notation

$$D = [-2(\Delta_{12}^2 + \Omega_{12}^2 - u_{12}^2 - \gamma_{12}^2) + 2B]^{1/2},$$

$$E = [2(\Delta_{12}^2 + \Omega_{12}^2 - u_{12}^2 - \gamma_{12}^2) + 2B]^{1/2}, \qquad\qquad (16)$$

$$B = [(\Delta_{12}^2 + \Omega_{12}^2 + u_{12}^2 + \gamma_{12}^2)^2 - 4(u_{12}\Omega_{12} + \Delta_{12}\gamma_{12})^2]^{1/2}.$$

To obtain the second-order correlation function, we have to find the correlation functions $\langle S_1^+(t)S_2^+(t+\tau)S_1^-(t+\tau)S_2^-(t)\rangle$, etc. To this end, we make use of the quantum regression theorem [25], which states that for $\tau > 0$ the two-time average $\langle S^+(t)S^+(t+\tau)S^-(t+\tau)S^-(t)\rangle$ satisfies the same equation of motion as the single-time average $\langle S^+(t)S^-(t)\rangle$.

Hence from equation (5) and equations (9)–(16) we find that for two non-identical atoms the second-order correlation function, for initially both atoms excited, has the form:

$$G^{(2)}(\mathbf{R}_1, t; \mathbf{R}_2, t+\tau) = 2\gamma_1\gamma_2 \frac{u(\hat{R}_1)u(\hat{R}_2)}{B} \exp[-2w(2t+\tau)]$$

$$\times \left\{ \tfrac{1}{4}X(D^2\cos E\tau + E^2\cosh D\tau) + X'(\cosh D\tau - \cos E\tau) \right.$$

$$\left. + \tfrac{1}{2}Y(D\sinh D\tau + E\sin E\tau) + 2m\,Y'\left(\frac{1}{D}\sinh D\tau - \frac{1}{E}\sin E\tau\right)\right\}, \qquad (17)$$

where

$$w = \tfrac{1}{2}(\gamma_1 + \gamma_2), \quad m = (\Delta_{12}u_{12} - \gamma_{12}\Omega_{12}),$$

$$X = 1 + \cos[k\mathbf{r}_{12}\cdot(\hat{R}_1 - \hat{R}_2)],$$

$$X' = \gamma_{12}^2 + \frac{u_{12}^2}{\gamma_1\gamma_2}(\gamma_1\gamma_2 + \gamma_{12}^2 + \Omega_{12}^2) + (\gamma_{12}^2 - \Delta_{12}^2)\cos(k\mathbf{r}_{12}\cdot\hat{R}_1)\cos(k\mathbf{r}_{12}\cdot\hat{R}_2)$$

$$- (\Delta_{12}^2 + \Omega_{12}^2)\sin(k\mathbf{r}_{12}\cdot\hat{R}_1)\sin(k\mathbf{r}_{12}\cdot\hat{R}_2)$$

$$- \frac{w(\Delta_{12}\gamma_{12} + u_{12}\Omega_{12})}{\sqrt{(\gamma_1\gamma_2)}}[\sin(k\mathbf{r}_{12}\cdot\hat{R}_1) - \sin(k\mathbf{r}_{12}\cdot\hat{R}_2)]$$

$$+ \frac{u_{12}(u_{12}\gamma_{12} - \Delta_{12}\Omega_{12})}{\sqrt{(\gamma_1\gamma_2)}}[\cos(k\mathbf{r}_{12}\cdot\hat{R}_1) - \cos(k\mathbf{r}_{12}\cdot\hat{R}_2)],$$

$$Y = \Delta_{12}\sin[k\mathbf{r}_{12}\cdot(\hat{R}_1 - \hat{R}_2)] - \frac{w\gamma_{12}}{\sqrt{(\gamma_1\gamma_2)}}[\cos(k\mathbf{r}_{12}\cdot\hat{R}_1) + \cos(k\mathbf{r}_{12}\cdot\hat{R}_2)]$$

$$+ \frac{u_{12}\Omega_{12}}{\sqrt{(\gamma_1\gamma_2)}}[\sin(k\mathbf{r}_{12}\cdot\hat{R}_1) + \sin(k\mathbf{r}_{12}\cdot\hat{R}_2)],$$

$$Y' = -u_{12}\sin[k\mathbf{r}_{12}\cdot(\hat{R}_1 - \hat{R}_2)] + \frac{w\Omega_{12}}{\sqrt{(\gamma_1\gamma_2)}}[\cos(k\mathbf{r}_{12}\cdot\hat{R}_1) + \cos(k\mathbf{r}_{12}\cdot\hat{R}_2)]$$

$$+ \frac{u_{12}\gamma_{12}}{\sqrt{(\gamma_1\gamma_2)}}[\sin(k\mathbf{r}_{12}\cdot\hat{R}_1) + \sin(k\mathbf{r}_{12}\cdot\hat{R}_2)]. \qquad (18)$$

Equation (17) is the exact analytical expression for the two-time second-order correlation function describing photon correlations in the spontaneous emission from two non-identical two-level atoms. The time-evolution of this second-order

correlation function is defined by the parameters u_{12}, B, D and E, and is dependent on the direction of observation, $\mathbf{R}_1$ and $\mathbf{R}_2$, with respect to the line connecting both atoms. The parameter D, together with the overall damping factor $\exp[-2w(2t+\tau)]$ describes the decay rates of the particular terms, and the parameter E describes a sinusoidal modulation superimposed on this smooth exponential decay. This sinusoidal modulation describes quantum beats resulting from the interference between transitions $|2\rangle\rightarrow|+\rangle\rightarrow|0\rangle$ and $|2\rangle\rightarrow|-\rangle\rightarrow|0\rangle$ (see figure 1 (*a*)) or $|2\rangle\rightarrow|\phi_+\rangle\rightarrow|0\rangle$ and $|2\rangle\rightarrow|\phi_-\rangle\rightarrow|0\rangle$ if the atoms are non-identical (see figure 1 (*b*)).

For $\tau=0$, equation (17) reduces to

$$G^{(2)}(\mathbf{R}_1, t; \mathbf{R}_2, t) = 2\gamma_1\gamma_2 u(\hat{R}_1)u(\hat{R}_2)\exp(-4wt)\{1+\cos[k\mathbf{r}_{12}\cdot(\hat{R}_1-\hat{R}_2)]\}. \qquad (19)$$

This implies that the probability of finding two photons simultaneously, does not exhibit quantum beats. Equation (19) reflects the correlation in emission and becomes zero for $1+\cos[k\mathbf{r}_{12}\cdot(\hat{R}_1-\hat{R}_2)]=0$ implying that for two directions $\hat{R}_1$ and $\hat{R}_2$, for which

$$k\mathbf{r}_{12}\cdot(\hat{R}_1-\hat{R}_2)=\pi(2n+1), \quad n=0, 1, 2, \ldots, \qquad (20)$$

simultaneous emission of photons is not possible. This photon anticorrelation effect, discussed recently by Mandel [26] and Paul [27] for independent atoms, has no classical analogue. It is due to spatial interference between different photons, and reflects the fact that one photon must have come from one source and one from the other, but we cannot tell which came from which. It should be noted that the single-time correlation function (19) is independent of interatomic interactions.

If the atoms are far apart from each other, we have $\gamma_{12}=0$ and $\Omega_{12}=0$, i.e. there is no coupling between the atoms, and equation (17) leads to the equation for the correlation function of independent atoms

$$G^{(2)}(\mathbf{R}_1, t; \mathbf{R}_2, t+\tau) = 2\gamma_1\gamma_2 u(\hat{R}_1)u(\hat{R}_2)\exp[-2w(2t+\tau)]$$
$$\times\{\cosh 2u_{12}\tau + \cos[k\mathbf{r}_{12}\cdot(\hat{R}_1-\hat{R}_2)-2\Delta_{12}\tau]\}. \qquad (21)$$

Thus, over a limited space-time region, the two-time second-order correlation function for non-interacting atoms shows a sinusoidal modulation in space and time, which can be interpreted both in terms of interference fringes and light beats. Equation (21) shows that interference effects are present even when the atoms (sources) are independent. The interference term in (21) when $u_{12}=0$ is the same as that obtained by Mandel [4], who considered the second-order correlation function for two beams emitted by independent lasers.

As already mentioned, the interference fringes in the single-time second-order correlation function are independent of interatomic interactions as well as of the differences between the atoms. For the two-time correlation function, however, these interference fringes show a different pattern. If $u_{12}\neq0$, i.e. if the atoms have different natural linewidths, the correlation function (21) always differs from zero.

If the atoms are identical, we have $u_{12}=0$, $\Delta_{12}=0$ and equation (17) takes the form

$$G^{(2)}(\mathbf{R}_1, t; \mathbf{R}_2, t+\tau) = 2\gamma^2 u(\hat{R}_1)u(\hat{R}_2)\exp[-2\gamma(2t+\tau)]$$
$$\times\{[1+\cos(k\mathbf{r}_{12}\cdot\hat{R}_1)\cos(k\mathbf{r}_{12}\cdot\hat{R}_2)]\cosh(2\gamma_{12}\tau)$$
$$-[\cos(k\mathbf{r}_{12}\cdot\hat{R}_1+\cos(k\mathbf{r}_{12}\cdot\hat{R}_2)]\sinh(2\gamma_{12}\tau)$$
$$+\sin(k\mathbf{r}_{12}\cdot\hat{R}_1)\sin(k\mathbf{r}_{12}\cdot\hat{R}_2)\cos(2\Omega_{12}\tau)\}. \qquad (22)$$

For $\tau = 0$, (22) reproduces equation (19) for the single-time correlation function meaning that the condition (20) for anticorrelation is the same for identical as well as for non-identical atoms. If the atoms are independent ($\gamma_{12} = 0, \Omega_{12} = 0$) then even for $\tau \neq 0$, (22) reproduces equation (19), signifying that for identical non-interacting atoms we obtain anticorrelations independently of τ.

Moreover, equation (22) shows that for identical atoms, the correlation function exhibits quantum beats with frequency $2\Omega_{12}$. These beats vanish if dipole–dipole interaction is ignored or if the atoms are confined in a region much smaller than the resonant wavelength ($kr_{12} \ll 1$). The amplitude of these beats depends on the direction of observation and vanishes for directions $\theta_1 = 90°$ or $\theta_2 = 90°$, where θ_1 (θ_2) is the angle between $\mathbf{r}_{12}$ and $\hat{R}_1$ ($\hat{R}_2$). This directional effect, which has its maximum for two photons detected in the direction $\theta_1 = \theta_2 = 0°$ is connected with the fact that the antisymmetrical state $|-\rangle$ does not radiate in the direction $\theta = 90°$ [20]. In the direction $\theta = 0°$ both states $|+\rangle$ and $|-\rangle$ radiate, and interference between them leads to the quantum beats.

For non-identical atoms, however, the two states $|\phi_\pm\rangle$ include the super-radiant state $|+\rangle$ and subradiant state $|-\rangle$, so that emission from $|\phi_\pm\rangle$ in different directions is possible and quantum beats appear in all directions. To illustrate this behaviour we have plotted the general solution (17) in figures 2–4 for several values of γ_2/γ_1 and Δ_{12} as well as different directions of observation.

Figure 2 shows the temporal dependence of the second-order correlation function for interatomic separation $r_{12} = \lambda/12$, $\gamma_1 = \gamma_2$ and different Δ_{12}. The first photon has been detected in the direction $\theta_1 = 0°$ at the time $t = 0$ and the second photon in the direction $\theta_2 = 0°$ at the time τ. For identical atoms, the intensity correlations exhibit small oscillations which increase as the atoms become more and more distinct.

In figure 3 we illustrate the same situation as in figure 2 albeit now the second photon is detected in the direction $\theta_2 = 90°$. In this case, for identical atoms,
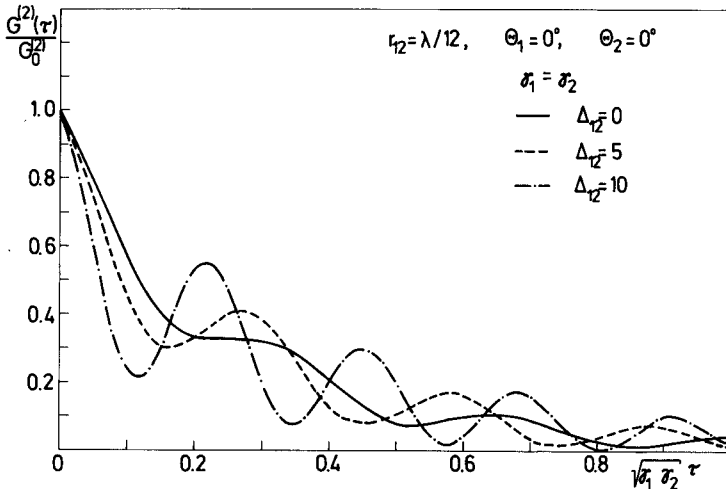

Figure 2.   Intensity correlations $G^{(2)}(\tau)$ plotted against the dimensionless time $\sqrt{(\gamma_1\gamma_2)}\tau$ for $r_{12} = \lambda/12, \gamma_1 = \gamma_2$ and different Δ_{12}. The first photon has been detected at the time t in the direction $\theta_1 = 0°$, where θ is the angle between $\mathbf{r}_{12}$ and $\mathbf{R}_1$, and the second photon in the direction $\theta_2 = 0°$ at the time τ. $G_0^{(2)} = 4\gamma_1\gamma_2 u(\hat{R}_1)u(\hat{R}_2)$.

$r_{12} = \lambda/12$, $\Theta_1 = 0°$, $\Theta_2 = 90°$

$\delta_1 = \delta_2$

——— $\Delta_{12} = 0$
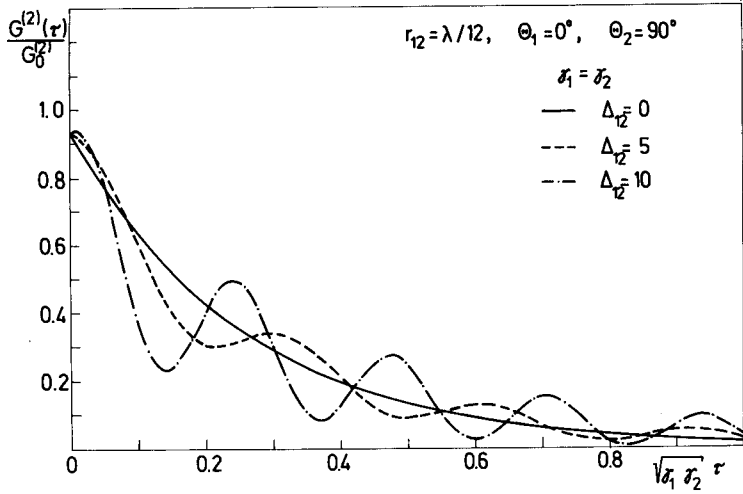
- - - $\Delta_{12} = 5$

—·— $\Delta_{12} = 10$

Figure 3. The same as in figure 2, except that the second photon has been detected in the direction $\theta_2 = 90°$.

oscillations are absent and we have a simple exponential decay of $G^{(2)}(\tau)$. If the atoms are non-identical, oscillations appear and increase with increasing Δ_{12}. According to equation (21) for two non-interacting atoms, beats appear if the atoms have different frequencies ($\Delta_{12} \neq 0$). In figure 4, we have plotted $G^{(2)}(\tau)$ for $\Delta_{12} = 0$ and different γ_2/γ_1. It is seen that for interacting atoms we have yet another mechanism leading to quantum beats. Namely, different atomic lifetimes of atoms cause the transition amplitudes $|2\rangle \rightarrow |-\rangle$ and $|-\rangle \rightarrow |0\rangle$ to differ from zero. This is due to the circumstance that the matrix elements $\langle 2|\mu_1 + \mu_2|-\rangle$ and $\langle -|\mu_1 + \mu_2|0\rangle$ are different from zero as the atoms have different transition dipole moments. For identical atoms
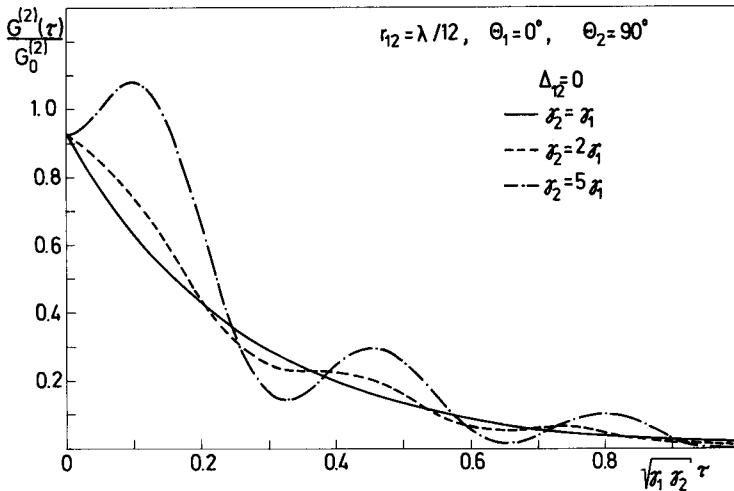
$r_{12} = \lambda/12$, $\Theta_1 = 0°$, $\Theta_2 = 90°$

$\Delta_{12} = 0$

——— $\delta_2 = \delta_1$

- - - $\delta_2 = 2\delta_1$

—·— $\delta_2 = 5\delta_1$

Figure 4. Intensity correlations $G^{(2)}(\tau)$ plotted against the dimensionless time $\sqrt{(\gamma_1\gamma_2)}\tau$ for $r_{12} = \lambda/12$, $\Delta_{12} = 0$ and different natural linewidths.

$(\boldsymbol{\mu}_1 = \boldsymbol{\mu}_2)$ these matrix elements are equal to zero; hence the transitions $|2\rangle \rightarrow |-\rangle$ and $|-\rangle \rightarrow |0\rangle$ are negligibly small.

However, in contrast to the case $\Delta_{12} \neq 0$, this mechanism vanishes if the atoms are mutually independent (see equation (21)).

3. Conclusions

We have studied the two-time second-order correlation function (intensity correlations) for spontaneous emission from two non-identical atoms. Particular attention has been paid to the appearance of quantum beats in such a system. The set of four equations describing the time-evolution of the system has been solved by the Laplace transform method. An exact analytical formula for the two-time second-order correlation function has been obtained using the quantum regression theorem.

We have shown that for interacting atoms beats appear for non-identical as well as for identical atoms. For non-identical atoms, two mechanisms lead to the quantum beats. The one, 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 $|0\rangle$, gives beats for interacting as well as for independent atoms. This latter situation is similar to the effect of interference between independent beams considered by Mandel [4] and observed experimentally by Javan *et al.* [2]. The other mechanism consists in the opening of the channels $|2\rangle \rightarrow |-\rangle$ and $|-\rangle \rightarrow |0\rangle$ as the linewidths of the atoms become different. It results from the circumstance that for different atoms $(\boldsymbol{\mu}_1 \neq \boldsymbol{\mu}_2)$ the matrix elements $\langle 2|\boldsymbol{\mu}_1 + \boldsymbol{\mu}_2|-\rangle$ and $\langle -|\boldsymbol{\mu}_1 + \boldsymbol{\mu}_2|0\rangle$ are different from zero; thus, these transitions are allowed. However, this mechanism is dependent on interatomic interactions, and vanishes for independent atoms.

For identical atoms, the quantum beats are strongly dependent on the direction of observation with respect to the line connecting both atoms. If the atoms are non-identical quantum beats are observed in all directions with respect to the line connecting them.

Finally, we have discussed the effect of photon anticorrelation due to spatial interference between the photons. Anticorrelation means that the joint probability of detecting two photons at two different points is zero. For two independent identical atoms this probability is zero if the distance between the two detectors is an odd multiple of half the wavelength. We find that this condition of anticorrelation for the single-time correlation function is independent of interatomic interactions and is the same for identical as well as for non-identical atoms. For the two-time correlation function, however, anticorrelation can appear only if the atoms are identical and mutually independent. If the atoms interact mutually or are non-identical, this effect is reduced.

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