COHERENCE AND QUANTUM OPTICS VI

Proceedings of the Sixth Rochester Conference on
Coherence and Quantum Optics held at the
University of Rochester, June 26–28, 1989

Edited by

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University of Rochester
Rochester, New York

PLENUM PRESS · NEW YORK AND LONDON

1990

PHOTON ANTIBUNCHING IN SPONTANEOUS EMISSION FROM TWO NONIDENTICAL ATOMS

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INTRODUCTION

A number of methods for the visualisation of quantum properties of the radiation field such as photon antibunching, sub-Poissonian photon statistics and squeezing have recently been proposed and in some cases implemented [1,2]. Photon antibunching is characterized by a field for which the normalized two-time second-order correlation function $g^{(2)}(\vec{R},t;\vec{R},t+\tau)$ for $\tau > 0$ is greater than its initial value for $\tau = 0$. This phenomenon describes a situation in which fewer photons appear close together than further apart, and is the opposite of photon bunching for which the photons tend to bunch in time and $g^{(2)}(\vec{R},t;\vec{R},t+\tau)$ for $\tau > 0$ is always below its initial value for $\tau = 0$. Photon antibunching is a quantum phenomenon with no classical analog in the sense that the corresponding state of the electromagnetic field cannot be given in the form of a positive diagonal coherent-state representation [3]. This nonclassical effect has been predicted theoretically for many optical processes [1,2] and observed experimentally in resonance fluorescence from single atoms [4,5]. However, antibunching vanishes if the field is radiated by many atoms, but is still preserved for two or three atoms [6,7]. This is due to the fact that antibunching is specific to fields with a small number of photons. Hence it is interesting to consider the possibility of obtaining photon antibunching in spontaneous emission from two two-level atoms, where we have two photons only.

INTENSITY CORRELATION FUNCTION

The aim of this paper is to calculate the spontaneous emission from two nonidentical atoms with different transition frequencies and/or different natural linewidths from the viewpoint of the photon antibunching effect. We concentrate on the normalized two-time second-order correlation function /intensity correlation/ for photons detected in the same direction $\vec{R}$ and at different times, defined as

Coherence and Quantum Optics VI
Edited by J. H. Eberly *et al.*
Plenum Press, New York, 1990

$$g^{(2)}(\vec{R},t;\vec{R},t+\tau)=\frac{\langle E^{(-)}(\vec{R},t)E^{(-)}(\vec{R},t+\tau)E^{(+)}(\vec{R},t+\tau)E^{(+)}(\vec{R},t)\rangle}{\langle E^{(-)}(\vec{R},t)E^{(+)}(\vec{R},t)\rangle\langle E^{(-)}(\vec{R},t+\tau)E^{(+)}(\vec{R},t+\tau)\rangle} \quad (1)$$

and particularized for the collective spontaneous emission from two nonidentical atoms, separated by r_{12} and coupled to each other via retarded dipole-dipole near-field interaction as well as to all the modes of the electromagnetic field, which are assumed to be initially in their vacuum state $|\{0_k\}\rangle$. The atoms are assumed to have the transition frequencies ω_1 and ω_2, respectively, the corresponding natural linewidths $2\gamma_1$ and $2\gamma_2$, and both to be initially in their respective excited state.

In the far-field limit the positive /negative/ frequency part of the field operator $E(\vec{R},t)$ can be expressed in terms of the atomic operators [8,9]

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

where $k=\omega_0/c$, with $\omega_0=(\omega_1+\omega_2)/2$ — the renormalized frequency of the two-atom system, $\vec{\mu}_i$ is the atomic transition dipole moment, $\hat{R}=\vec{R}/R$ — the unit vector in the direction $\vec{R}$, and S_i^+ and $S_i^-=(S_i^+)^+$ are operators that raise and lower the energy of the atom i and together with the operator S_i^z /describing the energy of the atom i / satisfy the well-known commutation relations $[S_i^+,S_j^-]=2S_i^z\delta_{ij}$ and $[S_i^z,S_j^\pm]=\pm S_i^\pm\delta_{ij}$. Since the field is initially in the vacuum state, the vacuum part $E_0^{(+)}(\vec{R},t)$ does not contribute to the expectation values of the normally ordered correlation operator in equation (1), and we obtain for $g^{(2)}(\vec{R},t;\vec{R},t+\tau)$ the expression

$$g^{(2)}(\vec{R},t;\vec{R},t+\tau)=\frac{\sum_{ijkl}(\gamma_i\gamma_j\gamma_k\gamma_l)^{\frac{1}{2}}\langle S_i^+(t)S_k^+(t+\tau)S_l^-(t+\tau)S_j^-(t)\rangle e^{ik\hat{R}\cdot(\vec{r}_{ij}+\vec{r}_{kl})}}{\left[\sum_{ij}(\gamma_i\gamma_j)^{\frac{1}{2}}\langle S_i^+(t)S_j^-(t)\rangle e^{ik\hat{R}\cdot\vec{r}_{ij}}\right]\left[\sum_{kl}(\gamma_k\gamma_l)^{\frac{1}{2}}\langle S_k^+(t+\tau)S_l^-(t+\tau)\rangle e^{ik\hat{R}\cdot\vec{r}_{kl}}\right]} \quad (3)$$

where $2\gamma_i$ is the Einstein A coefficient for the atom i. According to eq. 3, in order to study the intensity correlation function we have to find the correlation functions for the atomic operators. To this aim we use Lehmberg's master equation 8, generalized to the case of nonidentical atoms 10, which enables us to obtain the equation of motion for the atomic correlation functions, and reads

$$\begin{aligned}\frac{\partial\varrho}{\partial t}=&-i\sum_{i=1}^{2}\Delta_i[S_i^z,\varrho]-i\sum_{i\neq j}\Omega_{ij}[S_i^+S_j^-,\varrho]\\&-\sum_{i\neq j}\gamma_{ij}(S_i^+S_j^-\varrho+\varrho S_i^+S_j^--2S_i^-\varrho S_j^+)\\&-\sum_{i}\gamma_i(S_i^+S_i^-\varrho+\varrho S_i^+S_i^--2S_i^-\varrho S_i^+),\end{aligned} \quad (4)$$

where $\Delta_1=-\Delta_2=\Delta=(\omega_2-\omega_1)/2$. The collective parameters Ω_{ij}

and γ_{ij} , both dependent on the interatomic distance r_{12} , arise from the retarded dipole-dipole and radiative interaction between the atoms, and are defined in [8-10].

For two nonidentical atoms, equation (4) leads to a closed set of equations of motion for the vacuum expectation values given in eq.(3). By the Laplace transform method, this set transforms into an easily solvable system of algebraic equations in transformed variables. The general solution for the intensity correlation function $g^{(2)}(\vec{R},t;\vec{R},t+\tau)$ is very complicated; thus, we restrict ourselves to discussing the solution for $g^{(2)}(\vec{R},t;\vec{R},t+\tau)$ in some limiting cases of interatomic interactions and directions of photon emission.

A/ The case $\Delta \neq 0$, $\gamma_1 = \gamma_2$ and $\Omega_{12} \gg \sqrt{\gamma_1 \gamma_2}$. Here, the interatomic separation is very small by comparison with the resonant wavelength, the difference between the transition frequencies of the atoms differs from zero, but the natural linewidths of the atoms are identical. For simplicity, we assume that the photons emitted are observed in the same direction $\theta = 90°$, where θ is the direction of observation of photon emission with respect to the line connecting the two atoms. On these assumptions, the solution for the correlation function (3) takes the form

$$g^{(2)}(90°,0;90°,\tau) = \frac{\left[2 - \frac{\Delta^2}{\Omega_{12}^2+\Delta^2}(1-\cos 2\Omega_{12}\tau)\right]}{2\left[1+2a(1+a)e^{-2\gamma\tau}(1-e^{-2\gamma\tau})\right]} \qquad (5)$$

where $a = \gamma_{12}/\sqrt{\gamma_1\gamma_2}$.
Equation (5) shows that for small τ the intensity correlation function decreases with τ , thus manifesting the tendency of the photons to bunch in time. This is illustrated in figure 1, where $g^{(2)}(90°,0;90°,\tau)$ is plotted for $r_{12} = \lambda/12$, with

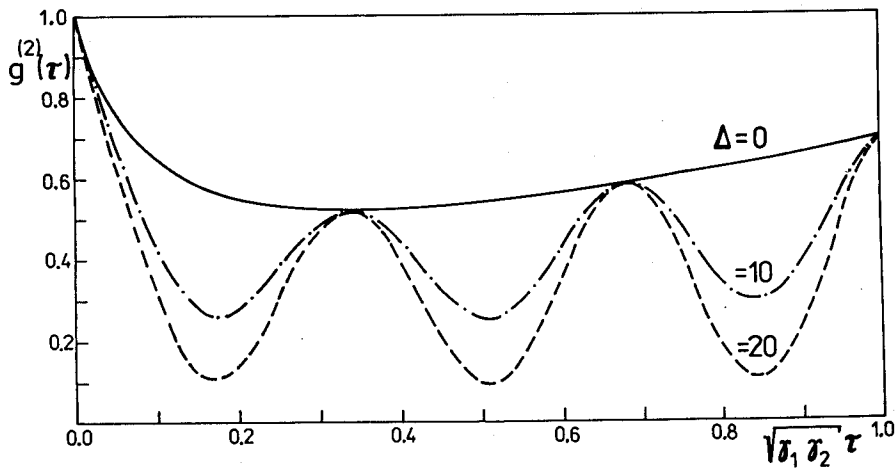

Fig.1

λ — the resonant wavelength, and various values of the parameter Δ . One photon is observed at the time $t=0$ and the other at the time τ in the same direction $\theta = 90^o$. Obviously, for all values of the parameter Δ , the intensity correlation function decreases with τ , and for $\Delta \neq 0$ it exhibits quantum beats with amplitude increasing with growing Δ . The explanation of these effects /photon bunching and quantum beats/ can be given with in the framework of collective states of a two-atom system [11].

On this model, our two-atom system is equivalent to a single four-level system with one upper state $|2\rangle$, one ground state $|1\rangle$ and two intermediate superradiant $|+\rangle$ and subradiant $|-\rangle$ states. For identical ($\Delta = 0$) strongly interacting atoms, the transition probability to and from the subradiant state $|-\rangle$ is very small, and only the superradiant state $|+\rangle$ can radiate. We thus have simple exponential decay of the photon correlations, and quantum beats do not appear because we have only one channel for emission. For nonidentical atoms with $\omega_1 \neq \omega_2$ ($\Delta \neq 0$) , the states $|\pm\rangle$ are no longer eigenstates of the two-atom system. The Hamiltonian of the system with $\Delta \neq 0$ can be rediagonalized [10] introducing certain eigenstates with two new states $|\varphi_\pm\rangle$ which already include the superradiant $|+\rangle$ and subradiant $|-\rangle$ states. In this case we have two channels $|\varphi_+\rangle \to |1\rangle$ and $|\varphi_-\rangle \to |1\rangle$ which can radiate simultaneously, since at the same moment we have radiation of correlated pairs of photons leading to photon bunching. Moreover, interference between these two transitions gives quantum beats.

B/ The case $\gamma_1 \neq \gamma_2$, $\Delta = 0$ and $\Omega_{12} \gg \sqrt{\gamma_1 \gamma_2}$. Here, we have two atoms with identical transition frequencies $\omega_1 = \omega_2$ but different natural linewidths $\gamma_1 \neq \gamma_2$. For simplicity, as in the case A, we assume that dipole-dipole interaction between the atoms is very strong and the photons are observed in

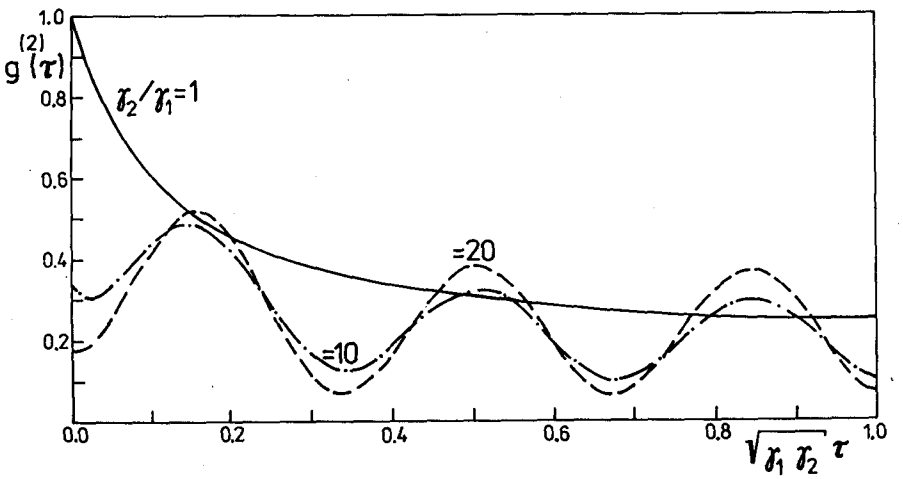

Fig.2

the direction $\vartheta = 90^\circ$. With these assumptions, the correlation function (3) takes the form:

$$g^{(2)}(90^\circ, 0; 90^\circ, \tau) = \frac{[2 + u^2(1 - \cos 2\Omega_{12}\tau)]}{2w^2\left\{1 + \frac{2\gamma_{12}(1 - e^{-(\gamma_1+\gamma_2)\tau})}{w^2\Omega_{12}^2}[\gamma_{12}(u^2 + \Omega_{12}^2) + \gamma_2(\Omega_{12}^2 - u^2)]\right\}} \quad ,(6)$$

with $u = (\gamma_2 - \gamma_1)/2\sqrt{\gamma_1\gamma_2}$ and $w = (\gamma_1 + \gamma_2)/2\sqrt{\gamma_1\gamma_2}$. Equation (6) shows sinusoidal modulation of the intensity correlation function with amplitude proportional to the difference of the natural linewidths of the two atoms. Moreover, for small τ the intensity correlation function (6) increases with τ and is greater than its initial value for $\tau = 0$. This means that in this case the photons have a tendency to anticorrelate in time. This is shown in figure 2, where $g^{(2)}(90^\circ, 0; 90^\circ, \tau)$ is plotted for $r_{12} = \lambda/12$, $\Delta = 0$ and different natural linewidths of the atoms.

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Work supported by the Research Program CPBP 01.06.