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Quantum beats in two-atom resonance fluorescence

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We study the resonance fluorescence emitted by two laser-driven coupled identical two-level atoms and allow for an arbitrary separation between the atoms and an arbitrary orientation of the interatomic axis in the laser field. Quantum beats arise in the resonance fluorescence intensity profile when an interatomic axis orientation is not perpendicular to the driving field propagation direction and are well resolved for an interatomic separation comparable to the resonant wavelength. These beats vanish for noninteracting atoms as well as for atoms separated by distances much smaller than the resonant wavelength. We also consider the time-dependent resonance fluorescence photon statistics that also exhibit quantum beats. Sub-Poissonian photon statistics occur for large interatomic separations and, for small separations, sub- and super-Poissonian statistics arise depending on the atomic orientation in the external driving field.

I. INTRODUCTION

The concept of the cooperative effects in the interaction of atoms and molecules with the vacuum field and with the laser field has been the subject of many theoretical papers since the pioneering article of Dicke.[1] Examples of such effects include superradiance,[2-5] coherent spectral line broadening,[6,7] collective quantum beats,[8-12] and collective quantum jumps.[13-16] These phenomena have also been extensively studied in experiments,[17-19] after the development of ultrashort pulse 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 studying these phenomena lies in their close connection with the quantum and classical (as well as with the spontaneous and stimulated) aspects of atomic emission.

An appropriate model by which to study these phenomena consists of N identical or nonidentical two-level atoms, coupled to a continuum of quantized electromagnetic field modes, and possibly to an external driving field. Some papers have been devoted to the study of these phenomena for the case of several atoms. Although a few-atom system is admittedly an elementary model, it offers some advantages over the multiatom problems. 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.

There have been a number of attempts to calculate the spectrum of fluorescence from cooperative atomic systems.[20-23] For example it has been shown that the multiatom resonance fluorescence spectrum of the atoms driven by a strong external field, in addition to the three peaks of the one-atom spectrum,[24,25] contains additional sidebands at the frequency $\pm 2\Omega$, where Ω is the Rabi frequency. However, their amplitude is very small and proportional to Ω^{-2}, and this causes difficulties in the experimental detection of these sidebands. Detection of these sidebands would be a new and interesting demonstration of collective atomic effects. Furthermore, for the resonance fluorescence spectrum, collective effects are manifested in the integral intensity of the N-atom fluorescence spectrum which is $N(N+2)/3$ times that for a single-atom fluorescence. However, this enhancement of the integrated fluorescence by N atoms, labeled in the literature as a scaling factor,[26] appears only in S^2-conserving systems, i.e., for systems with interatomic separations much less than the resonant wavelength. Otherwise, for interatomic separations comparable to the resonant wavelength (S^2-breaking system), the integral intensity of the N-atom fluorescence is N times that for a single-atom case,[27,28] and the intensity correlation function for the S^2-breaking system differs considerably from that for S^2-conserving systems.[23,29]

To study the collective behaviors of S^2-breaking systems authors have restricted their considerations to the case of two atoms only, and, moreover, they have assumed that both atoms, which are at different points, experience the same driving field intensity and phase.[27-30] In this model, atoms are located in such a way that the interatomic separation vector $\mathbf{r}_{12}$ is perpendicular to the direction of propagation of the external driving field. For this case the Rabi frequency Ω is identical for both atoms. Therefore, it is interesting to study in some detail how different locations of the atoms, with reference to the propagation direction of the external driving field, can change the behavior of the emitted fluorescence field. Thus the purpose of this paper is to consider resonance fluorescence from two identical two-level atoms which are separated by $\mathbf{r}_{12}$ and experience different driving fields.

We focus our attention on time-dependent effects in collective two-atom resonance fluorescence, such as quantum beats in the integrated resonance fluorescence intensity and in the time-dependent photon statistics. To describe the two-atom resonance fluorescence we adapt the Lehmberg master equation[31] to the case of coherent pumping and obtain a closed system of 15 coupled equations describing the time evolution of the atomic variables. We solve this set of equations numerically, assuming that the driving laser field is weak (i.e., the Rabi frequency is much less than the coefficient for spontaneous decay). There are two reasons for which we restrict ourselves to the case of a weak driving field. Firstly, the sinusoidal modulations of the time-dependent resonance fluorescence intensity for a weak driving field are not modified by the Rabi oscillations, whereas the modulations are present for a strong driving field.[25] Secondly, for a strong driving field, both atoms are highly excited and in this case quantum beats do not appear. This is easy to understand within the framework of collective states in a two-atom system.[1,31] In this representation, the two-atom system is equivalent to a single four-level system with one upper state $|2\rangle$, one ground state $|0\rangle$, and two intermediate states: a superradiant state $|+\rangle$ and a subradiant state $|-\rangle$. For highly excited atoms, the system starts from the common excited state $|2\rangle$ and decays to the states $|\pm\rangle$. It is well known that an atomic configuration with one common upper level and two or more closely spaced lower levels does not lead to quantum beats.[32] Only a configuration with two or more closely spaced upper levels and a single lower level predicts quantum beats. For a weak driving field, the probability that the state $|2\rangle$ is populated is very small and the population oscillates between the intermediate states $|\pm\rangle$ and the ground state $|0\rangle$.[16] In this case we have an approximate configuration with two upper levels: superradiant $|+\rangle$ and subradiant $|-\rangle$, and one ground level $|0\rangle$, which can give quantum beats. However, there is another restriction on the occurrence of quantum beats in such a system: both transitions $|+\rangle \rightarrow |0\rangle$ and $|-\rangle \rightarrow |0\rangle$ must be correlated. We shall show in this paper that such correlations appear in a two-atom system only if the atoms experience a different driving field. In this case we can observe quantum beats in the transient regime of the resonance fluorescence intensity.

We also study time-dependent photon statistics in a two-atom system which is characterized by the quantity $Q(t,T)$ introduced by Mandel.[33] Negative values of $Q(t,T)$ imply sub-Poissonian photon statistics; a positive $Q(t,T)$ implies that the statistics are super-Poissonian. We discuss the effects of interatomic separation and interatomic orientation in the external driving field on the quantity $Q(t,T)$. In the transient regime of resonance fluorescence, the photon statistics vary from sub-Poissonian to super-Poissonian and vice versa depending on the interatomic separation and the atomic orientation in the external driving field.

II. MODEL AND METHOD

We consider two identical two-level atoms, separated by $\mathbf{r}_{12}$, in the field of a linearly polarized, coherent laser beam, with a frequency ω_L which is assumed to be exactly equal to the atomic transition frequency ω_0, i.e., detuning is zero. The atoms are coupled to each other via a retarded dipole-dipole near-field interaction and to all other modes of the electromagnetic field which are assumed as being initially in their vacuum state. Each atom is modeled as 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.

To describe the evolution of the two-atom system in an external driving laser field, we use the Lehmberg master equation[31] which, for an arbitrary combination W of atomic operators, reads as

$$\frac{dW}{dt}=i\omega_0\sum_{i=1}^{2}[S_i^+S_i^-,W]-\frac{i\boldsymbol{\mu}}{\hbar}\cdot\sum_{i=1}^{2}\{[S_i^+,W]\mathbf{E}_0^{(+)}(\mathbf{r}_i,t)-\mathbf{E}_0^{(-)}(\mathbf{r}_i,t)[W,S_i^-]\}+\sum_{\substack{i,j\\ i\neq j}}i\Omega_{ij}[S_i^+S_j^-,W]$$
$$+\sum_{i,j}\gamma_{ij}[S_i^+WS_j^- -\tfrac{1}{2}(S_i^+S_j^-W+WS_i^+S_j^-)]\ , \tag{1}$$

where all atomic operators are evaluated at time t, and $\boldsymbol{\mu}$ is the transition electric dipole moment vector. The operators S_i^+ and $S_i^-=(S_i^+)^\dagger$ raise and lower the energy of the ith atom and satisfy the angular momentum commutation relations

$$[S_i^+,S_j^-]=2S_i^z\delta_{ij},\quad [S_i^z,S_j^\pm]=\pm S_i^\pm\delta_{ij}\ . \tag{2}$$

In Eq. (1) the free field operators $\mathbf{E}_0^{(\pm)}(\mathbf{r}_i,t)$ describe an external field, which we assume to be a linearly polarized laser beam of frequency $\omega_L=\omega_0$ and which is in a coherent state with amplitudes $\mathcal{E}_0^{(\pm)}(\mathbf{r}_i,t)$. For simplicity, we choose the reference frame such that the atoms are at the positions $\mathbf{r}_1=(-\frac{1}{2}r_{12},0,0)$ and $\mathbf{r}_2=(\frac{1}{2}r_{12},0,0)$, and then the driving laser field amplitude $\mathcal{E}_0^{(+)}(\mathbf{r}_i,t)$ is $\mathcal{E}_0^{(+)}\exp[-i(\omega_0 t+\frac{1}{2}\mathbf{k}_L\cdot\mathbf{r}_{12})]$ at the point $\mathbf{r}_1$, and $\mathcal{E}_0^{(+)}\exp[-i(\omega_0 t-\frac{1}{2}\mathbf{k}_L\cdot\mathbf{r}_{12})]$ at the point $\mathbf{r}_2$, where $\mathbf{k}_L$ is the laser field wave vector. In this case the Rabi frequency $\Omega=2\boldsymbol{\mu}\cdot\mathcal{E}_0^{(+)}(\mathbf{r}_i)/\hbar$ is different for different atoms. The collective parameters Ω_{ij} and γ_{ij}, which appear in Eq. (1), arise from the retarded dipole-dipole and radiative interaction between the atoms. The parameters depend on the interatomic distance $r_{ij}=|\mathbf{r}_{ij}|$ for $\mathbf{r}_{ij}=\mathbf{r}_i-\mathbf{r}_j$ and are defined as[6,12,34]

$$\Omega_{ij}=\tfrac{3}{2}\gamma\left[-[(\hat{\boldsymbol{\mu}}_i\cdot\hat{\boldsymbol{\mu}}_j)-(\hat{\boldsymbol{\mu}}_i\cdot\hat{\mathbf{r}}_{ij})(\hat{\boldsymbol{\mu}}\cdot\hat{\mathbf{r}}_{ij})]\frac{\cos(kr_{ij})}{kr_{ij}}+[(\hat{\boldsymbol{\mu}}_i\cdot\hat{\boldsymbol{\mu}}_j)-3(\hat{\boldsymbol{\mu}}_i\cdot\hat{\mathbf{r}}_{ij})(\hat{\boldsymbol{\mu}}_j\cdot\hat{\mathbf{r}}_{ij})]\left[\frac{\sin(kr_{ij})}{(kr_{ij})^2}+\frac{\cos(kr_{ij})}{(kr_{ij})^3}\right]\right] \tag{3}$$

and

$$\gamma_{ij}=\tfrac{3}{2}\gamma\left[[(\hat{\boldsymbol{\mu}}_i\cdot\hat{\boldsymbol{\mu}}_j)-(\hat{\boldsymbol{\mu}}_i\cdot\hat{\mathbf{r}}_{ij})(\hat{\boldsymbol{\mu}}_j\cdot\hat{\mathbf{r}}_{ij})]\frac{\sin(kr_{ij})}{kr_{ij}}+[(\hat{\boldsymbol{\mu}}_i\cdot\hat{\boldsymbol{\mu}}_j)-3(\hat{\boldsymbol{\mu}}_i\cdot\hat{\mathbf{r}}_{ij})(\hat{\boldsymbol{\mu}}_j\cdot\hat{\mathbf{r}}_{ij})]\left[\frac{\cos(kr_{ij})}{(kr_{ij})^2}-\frac{\sin(kr_{ij})}{(kr_{ij})^3}\right]\right], \tag{4}$$

where $\hat{\boldsymbol{\mu}}_i$ and $\hat{\mathbf{r}}_{ij}$ are unit vectors along the transition electric dipole moment and the vector $\mathbf{r}_{ij}$, respectively. Moreover, $k=\omega_0/c=2\pi/\lambda_0$ with λ_0 the resonant wavelength. In Eqs. (3) and (4), $2\gamma=\gamma_{ii}=\gamma_{jj}=(4\mu^2k^3/3\hbar)$ is the Einstein A coefficient for spontaneous emission.

The parameters (3) and (4) describe the collective shift of energy levels and collective properties of the two-atom system. For large interatomic separations kr_{ij} goes to infinity, and then $\gamma_{ij}=\Omega_{ij}=0$, i.e., there is no coupling between the atoms. In this case, the master equation (1) produces the well-known optical Bloch equations commonly used to describe optical resonance phenomena.[35] Otherwise, for very small interatomic separations, kr_{ij} goes to zero, and from (3) and (4) we obtain (for parallel dipole orientations)

$$\Omega_{ij}\approx\frac{3\gamma}{2(kr_{ij})^3}[1-3(\hat{\boldsymbol{\mu}}_i\cdot\hat{\mathbf{r}}_{ij})^2], \tag{5a}$$

and

$$\gamma_{ij}=\gamma\ . \tag{5b}$$

For this case we see that γ_{ij} reduces to γ, and Ω_{ij} reduces to the static dipole-dipole interaction potential. Here, we assume that the atomic dipole moments $\boldsymbol{\mu}_i$ are parallel to each other, because both atoms are in the same linearly polarized external field. In general, however, atomic dipole moments are not parallel, which changes the strength of the interatomic interaction.

The validity of Eq. (1) hinges on the assumption that the system is Markovian, i.e., the retardation effects are ignored. As shown by Milonni and Knight,[36] the retardation effects play a significant role in the resonant interaction of two identical atoms only if $kr_{ij}>\pi$. This means that the atoms must be more than half of a transition wavelength apart. In this paper, we shall consider new phenomena like quantum beats, which strongly depend on the interatomic interactions and are significant for small interatomic separations[6,31] ($kr_{ij}<\pi$). The retardation does not play an important role for such small separations.

For a two-atom system, substituting the atomic operators $S_i^{\pm}$ $(i=1,2)$ for W in Eq. (1) leads to a closed system of 15 first-order differential equations describing the evolution of the atomic variables. This system can be written in matrix form as the inhomogeneous equation

$$\frac{d}{d\tau}\mathbf{Y}=\underline{M}\mathbf{Y}+\boldsymbol{\alpha}\ , \tag{6}$$

where $\underline{M}$ is a real invertible 15×15 matrix, $\mathbf{Y}$ is a column vector of Hermitian operators with the following components:

$$\begin{aligned}
&Y_1=S_1^++S_2^++S_1^-+S_2^-\ ,\\
&Y_2=i(S_1^-+S_2^--S_1^+-S_2^+)\ ,\\
&Y_3=S_1^+S_1^-+S_2^+S_2^-\ ,\quad Y_4=S_1^+S_2^-+S_2^+S_1^-\ ,\\
&Y_5=S_1^+S_2^++S_1^-S_2^-\ ,\quad Y_6=i(S_1^+S_2^+-S_1^-S_2^-)\ ,\\
&Y_7=S_1^+S_1^-S_2^-+S_2^+S_1^-S_2^-+S_1^+S_2^+S_1^-+S_1^+S_2^+S_2^-\ ,\\
&Y_8=i(S_1^+S_1^-S_2^-+S_2^+S_1^-S_2^--S_1^+S_2^+S_1^--S_1^+S_2^+S_2^-)\ ,\\
&Y_9=S_1^+S_2^+S_1^-S_2^-\ ,\quad Y_{10}=S_1^--S_2^-+S_1^+-S_2^+\ ,\\
&Y_{11}=i(S_1^--S_2^--S_1^++S_2^+),\quad Y_{12}=S_1^+S_1^--S_2^+S_2^-\ ,\\
&Y_{13}=S_1^+S_2^--S_2^+S_1^-\ ,\\
&Y_{14}=S_1^+S_1^-S_2^--S_2^+S_1^-S_2^-+S_1^+S_2^+S_1^--S_1^+S_2^+S_2^-\ ,\\
&Y_{15}=i(S_1^+S_1^-S_2^--S_2^+S_1^-S_2^--S_1^+S_2^+S_1^-+S_1^+S_2^+S_2^-)\ ,
\end{aligned} \tag{7}$$

and the vector $\boldsymbol{\alpha}$ is given by the components

$$\alpha_i=-4\beta[\delta_{1i}\cos(\tfrac{1}{2}\mathbf{k}_L\cdot\mathbf{r}_{12})+\delta_{11i}\sin(\tfrac{1}{2}\mathbf{k}_L\cdot\mathbf{r}_{12})]\ . \tag{8}$$

The matrix $\underline{M}$ is conveniently expressed in block form as

$$\underline{M}=\begin{bmatrix}A & D\\ C & B\end{bmatrix}, \tag{9}$$

for block A of dimension 9×9 and block B of dimension 6×6. The two off-diagonal blocks are C of dimension 6×9 and D of dimension 9×6.

Block A has the form

$$A=\begin{bmatrix} -\frac{1}{2}(1+a) & -\frac{1}{2}b & 4\beta_1 & 0 & 0 & 0 & a & b & 0 \\ \frac{1}{2}b & \frac{1}{2}(1+a) & 0 & 0 & 0 & 0 & -b & a & 0 \\ -\beta_1 & 0 & -1 & -a & 0 & 0 & 0 & 0 & 0 \\ -\beta_1 & 0 & -a & -1 & 0 & 0 & 2\beta_1 & 0 & 4a \\ -\beta_1 & 0 & 0 & 0 & -1 & 0 & 2\beta_1 & 0 & 0 \\ 0 & \beta_1 & 0 & 0 & 0 & -1 & 0 & -2\beta_1 & 0 \\ 0 & 0 & -2\beta_1 & -2\beta_1 & -2\beta_1 & 0 & -\frac{1}{2}(3+a) & \frac{1}{2}b & 8\beta_1 \\ 0 & 0 & 0 & 0 & 0 & 2\beta_1 & -\frac{1}{2}b & -\frac{1}{2}(3+a) & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -\beta_1 & 0 & -2 \end{bmatrix}, \tag{9a}$$

and B is given by

$$B=\begin{bmatrix} -\frac{1}{2}(1-a) & \frac{1}{2}b & 4\beta_1 & 0 & a & b \\ -\frac{1}{2}b & -\frac{1}{2}(1-a) & 0 & 0 & -b & a \\ -\beta_1 & 0 & -1 & -b & 0 & 0 \\ 0 & \beta_1 & b & -1 & 0 & 2\beta_1 \\ 0 & 0 & -2\beta_1 & 0 & -\frac{1}{2}(3-a) & -\frac{1}{2}b \\ 0 & 0 & 0 & -2\beta_1 & \frac{1}{2}b & -\frac{1}{2}(3-a) \end{bmatrix}. \tag{9b}$$

The off-diagonal blocks C and D possess the nonzero elements

$$\begin{aligned} C_{41} &= -C_{32}=\tfrac{1}{4}C_{23}=-\tfrac{1}{2}C_{47}=\tfrac{1}{2}C_{56}=\tfrac{1}{2}C_{63}=-\tfrac{1}{2}C_{64} \\ &=\tfrac{1}{2}C_{65}=-\tfrac{1}{8}C_{69}=\tfrac{1}{4}D_{23}=-D_{32}=D_{42}=\tfrac{1}{2}D_{46} \\ &=-D_{52}=-\tfrac{1}{2}D_{56}=-D_{61}=-\tfrac{1}{2}D_{65}=\tfrac{1}{2}D_{74} \\ &=\tfrac{1}{2}D_{83}=D_{96}=\beta_2 \, . \end{aligned} \tag{9c}$$

In Eqs. (6)–(9) we have introduced the notation

$$\begin{gathered} \tau=2\gamma t, \quad a=\frac{\gamma_{ij}}{\gamma}, \quad b=\frac{\Omega_{ij}}{\gamma}, \quad \beta=\frac{\Omega}{4\gamma}, \\ \beta_1=\beta\cos(\tfrac{1}{2}k_L r_{12}\cos\varphi), \quad \beta_2=\beta\sin(\tfrac{1}{2}k_L r_{12}\cos\varphi) \end{gathered} \tag{10}$$

with φ the angle between $\mathbf{k}_L$ and $\mathbf{r}_{12}$. It is obvious from (9c) and (10) that, for small interatomic separations ($k_L r_{12} \ll 1$), $\beta_2 \approx 0$, whence all elements of blocks C and D are zero. These elements (9c) are also zero for the case when the interatomic axis $\mathbf{r}_{12}$ is perpendicular to the direction of propagation of the laser field.[27–30] In these cases the Rabi frequency is the same for both atoms and the matrix $\underline{M}$ is block diagonal. However, in general, $\mathbf{r}_{12}$ cannot be perpendicular to the direction of propagation of the laser field, and we have to solve a set of 15 coupled equations.

In Eq. (6) the vector $\mathbf{Y}$ has been written in such a form that the first nine components are symmetric under interchange of the atoms, and the remaining six components are antisymmetric. In the framework of collective states,[1,31] a two-atom system is equivalent to a single four-level system (Fig. 1), with one upper state $|2\rangle=|2\rangle_1|2\rangle_2$, one ground state $|0\rangle=|1\rangle_1|1\rangle_2$, and two intermediate states $|\pm\rangle=(1/\sqrt{2})(|2\rangle_1|1\rangle_2\pm|1\rangle_1|2\rangle_2)$. The energies of these states are $E_2=2\hbar\omega_0$, $E_0=0$, and $E_\pm=\hbar(\omega_0\pm\Omega_{12})$. In this representation the symmetric combination of the atomic operators describes transitions

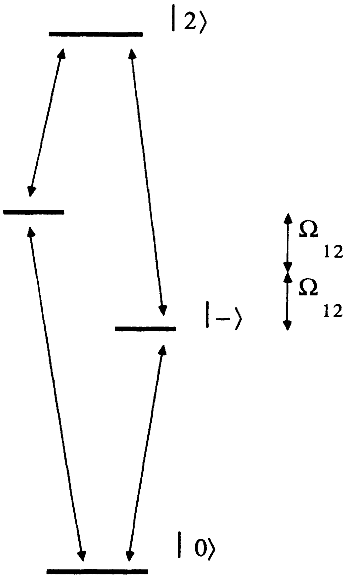

FIG. 1. Energy-level diagram of the two two-level atomic system represented as a single four-level atom. The excited and ground states are $|2\rangle$ and $|0\rangle$, respectively, and the intermediate levels are the symmetric state $|+\rangle$ and the antisymmetric state $|-\rangle$. The frequency shift Ω_{12} is due to the dipole-dipole interaction.

via the superradiant state, and the antisymmetrical combination describes transitions via the subradiant state. From (9) we see that for interatomic separations r_{12} much less than the resonant wavelength, and also for a perpendicular orientation of $\mathbf{r}_{12}$ to the direction of propagation of the laser field, the symmetrical and antisymmetrical combinations of the atomic operators are decoupled. In these cases the superradiant and subradiant states radiate independently. If $\mathbf{r}_{12}$ is not too small and is not perpendicular to the direction of propagation of the laser field, these states are coupled and radiate collectively.

In order to study the time-dependent effects in two-atom resonance fluorescence we have to solve Eq. (6). Direct integration of Eq. (6) leads to the following formal solution for $\mathbf{Y}$:

$$\mathbf{Y}(t)=\mathbf{Y}(0)e^{\underline{M}t}-\underline{M}^{-1}\boldsymbol{\alpha}(1-e^{\underline{M}t}) \ . \qquad (11)$$

There exists a complex invertible matrix $\underline{T}$ which diagonalizes $\underline{M}$ and $\underline{\lambda}=\underline{TMT}^{-1}$ is the diagonal matrix of complex eigenvalues. By introducing $\mathbf{X}=\underline{T}\mathbf{Y}$ and $\boldsymbol{\alpha}'=\underline{T}\boldsymbol{\alpha}$, Eq. (11) can be written as

$$\mathbf{X}(t)=\mathbf{X}(0)e^{\underline{\lambda}t}-\underline{\lambda}^{-1}\alpha'(1-e^{\underline{\lambda}t}) \ . \qquad (12)$$

To obtain solutions for $Y_i(t)$, we must determine the 15 eigenvalues λ_i and 15 eigenvectors $X_i(t)$; the eigenvalues and eigenvectors are solved numerically. The solution of Eq. (12) permits the calculation of the time-dependent characteristics of the two-atom system and is used for calculating the resonance fluorescence intensity and the photon statistics.

III. TIME-DEPENDENT RESONANCE FLUORESCENCE INTENSITY

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

$$I(\mathbf{R},t)=\frac{R^2c}{2\pi\omega_0}\langle \mathbf{E}^{(-)}(\mathbf{R},t)\cdot\mathbf{E}^{(+)}(\mathbf{R},t)\rangle \ . \qquad (13)$$

The vectors $\mathbf{E}^{(+)}$ ($\mathbf{E}^{(-)}$) denote the positive (negative) frequency components of the electromagnetic field, and we have introduced the factor $(R^2c/2\pi\omega_0)$ so that $I(\mathbf{R},t)d\Omega_{\hat{\mathbf{R}}}dt$ is now the probability of finding one photon inside the solid angle element $d\Omega_{\hat{\mathbf{R}}}$, around the direction $\hat{\mathbf{R}}$ in the time interval dt, at the moment of time in the far field zone of the radiation emitted by the atomic system. The positive frequency part $\mathbf{E}^{(+)}$ of the field operator in the far-field zone, $|\mathbf{R}| \gg \lambda_0, r_{12}$, where λ_0 is the resonant wavelength, and for $t > |\mathbf{R}|/c$, can be expressed in terms of the atomic operators[31,34]

$$\begin{aligned}\mathbf{E}^{(+)}(\mathbf{R},t)=\mathbf{E}_0^{(+)}(\mathbf{R},t)&\\ -k^2\sum_{i=1}^{2}\frac{\hat{\mathbf{R}}\times(\hat{\mathbf{R}}\times\boldsymbol{\mu})}{|\mathbf{R}|}S_i^-\left[t-\frac{|\mathbf{R}|}{c}\right]&\\ \times\exp(-ik\hat{\mathbf{R}}\cdot\mathbf{r}_i) \ ,&\qquad (14)\end{aligned}$$

where $k=\omega_0/c$, and $\mathbf{E}_0^{(+)}(\mathbf{R},t)$ denotes the operator of the free field, including the external driving field. The

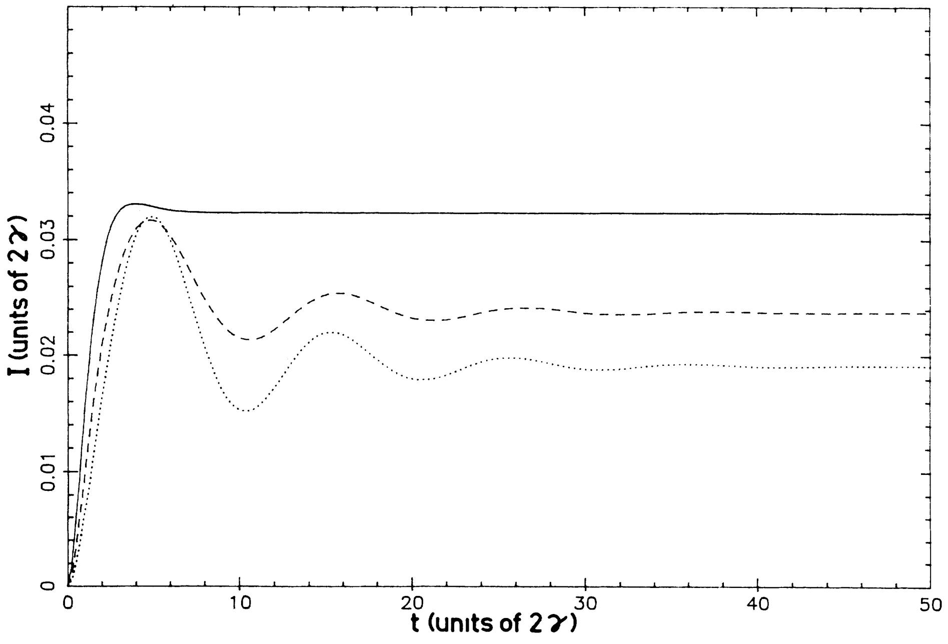

FIG. 2. Time-dependent resonance fluorescence intensity curves (in units of 2γ) as a function of $2\gamma t$ are presented for a driving field strength $\beta=0.1$ and an interatomic separation $r_{12}=\lambda_0/6$. The orientation angle φ assumes the values $\varphi=\pi/2$ (solid line), $\varphi=\pi/4$ (dashed line) and $\varphi=0$ (dotted line).

second part of Eq. (14) depends directly on the atomic operators and represents the radiation field emitted by the atoms. Substituting (14) into (13) and assuming that the observation point (detector) is outside of the external driving field [the free field part $\mathbf{E}_0^{(+)}(\mathbf{R},t)$ then does not contribute to the intensity of the radiated field observed at the point $\mathbf{R}$], we thus obtain the expression

$$I(\mathbf{R},t)=2\gamma U(\mathbf{R})\sum_{i,j=1}^{2}\langle S_i^+(t)S_j^-(t)\rangle \exp(ik\hat{\mathbf{R}}\cdot\mathbf{r}_{ij}) , \quad (15)$$

where $U(\mathbf{R})=(3/8\pi)\sin^2\Phi$, with Φ the angle between the observation direction $\mathbf{R}$ and the atomic transition dipole moment $\boldsymbol{\mu}$.

On integrating over all solid angles $d\Omega_{\hat{\mathbf{R}}}$, Eq. (15)

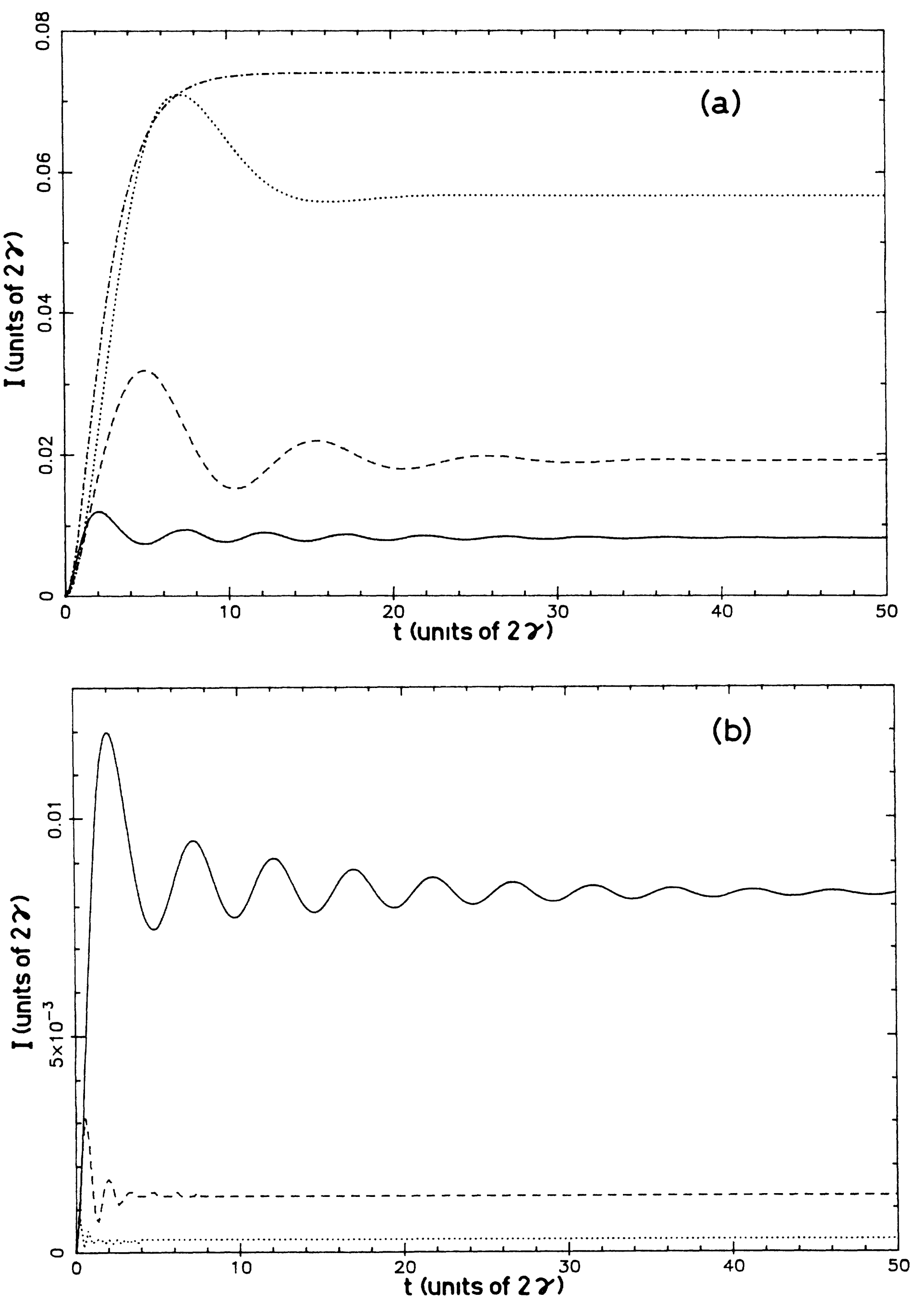

FIG. 3. Time-dependent resonance fluorescence intensity curves (in units of 2γ) are presented for a driving field strength $\beta=0.1$ and for the interatomic axis orientated parallel to the external driving field. The interatomic separation assumes the values (a) $r_{12}=\lambda_0/8$ (solid line), $r_{12}=\lambda_0/6$ (dashed line), $r_{12}=\lambda_0/4$ (dotted line), $r_{12}=50\lambda_0$ (dash-dotted line); and (b) $r_{12}=\lambda_0/8$ (solid line), $r_{12}=\lambda_0/12$ (dashed line), and $r_{12}=\lambda_0/16$ (dotted line).

yields the total radiation intensity $I(t)$ given in photons per second as

$$I(t)=\sum_{i,j=1}^{2} 2\gamma_{ij}\langle S_i^+(t)S_j^-(t)\rangle \ , \tag{16}$$

where γ_{ij} is given by Eq. (4). In the notation of the components of the vector **Y**, Eq. (16) takes the form

$$I(t)=2\gamma[Y_3(t)+aY_4(t)] \ . \tag{17}$$

The solution for the intensity of the fluorescent field [Eq. (17)] is illustrated graphically in Figs. 2 and 3 as a function of time for different interatomic separations and for different orientations of the interatomic axis $\mathbf{r}_{12}$ to the direction of propagation of the laser field. It is evident from Fig. 2 that the time evolution of the fluorescence intensity strongly depends on the orientation φ of the interatomic line $\mathbf{r}_{12}$ with reference to the direction of propagation of driving field, and shows an oscillatory modulation (quantum beats) for the atomic orientation, different from that perpendicular to $\mathbf{k}_L$. This oscillatory behavior of the resonance fluorescence intensity arises due to interference between the two possible transition amplitudes ($|\pm\rangle\rightarrow|0\rangle$). The amplitude of this modulation reaches a maximum for the interatomic line $\mathbf{r}_{12}$ located parallel to the direction of propagation of the external field ($\varphi=0$). Moreover, the amplitude as well as the frequency of these quantum beats is dependent on the interatomic interaction and vanishes for large interatomic separations [Fig. 3(a)] as well as for separations very small compared with the resonant wavelength [Fig. 3(b)]. This is easily explained in the framework of collective states of a two-atom system. For large interatomic separations, Ω_{12} is approximately zero, and transitions from the states $|\pm\rangle$ to the state $|0\rangle$ have the same frequency. Therefore there are no quantum beats in the emitted field. On the other hand, for very small interatomic separations, the subradiant state $|-\rangle$ is decoupled from the remaining states,[16,31,34] and in this case there are no quantum beats. This is also evident from the matrix (9). For very small interatomic separations, β_2 is approximately zero, and hence the superradiant and subradiant states are not coupled.

The role of the dipole-dipole interaction in the occurrence of quantum beats is shown in Fig. 4, where the real and imaginary parts of the 15 eigenvalues λ_i of the matrix $\underline{M}$, with and without dipole-dipole interaction Ω_{12}, are illustrated. These eigenvalues cluster in three groups corresponding to Re(λ_i) near 0, 1, and 2. Clustering in three groups occurs for values of the parameter a near unity [Eq. (10)] and the real parts of the eigenvalues are given by the diagonal elements of $\underline{M}$ (for dipole-dipole coupling *not* ignored). Contributions of eigenvalues with large real parts to the resonance fluorescence intensity are very small, because they rapidly damp the corresponding time-dependent eigenvector. Only eigenvalues with small real parts play a significant role in the time evolution of the resonance fluorescence intensity. In Fig. 4 we observe just three eigenvalues with small real parts: one is real and two are complex conjugates. For the case that the dipole-dipole interaction Ω_{12} is ignored, the imaginary parts of these Y eigenvalues are very small compared to the large values where Ω_{12} is included. These large imaginary parts of the eigenvalues determine the frequency of the quantum beats in the resonance fluorescence intensity and the real parts contribute to the decay of the transient beat phenomenon.

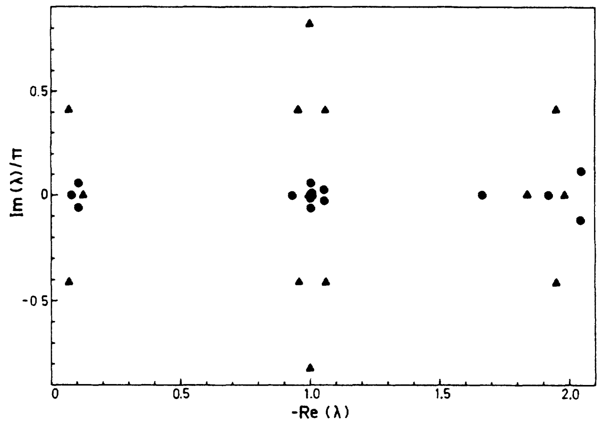

FIG. 4. Complex eigenvalues of the matrix $\underline{M}$ for $\beta=0.1$, $r_{12}=\lambda_0/8$, and $\varphi=0$, where the dipole-dipole interaction is included (triangles) and ignored (circles).

IV. PHOTON STATISTICS

Our time-dependent solution (12) for atomic correlation functions gives us a good starting point to also consider the problems of photon statistics of the fluorescent field radiated by a system of two interacting atoms. Photon statistics can be examined by the quantity $Q(t,T)$ introduced by Mandel[33,37] and which, for sufficiently short counting times T, has the form[37]

$$Q(t,T)=\eta T[g^{(2)}(t)-1] \ , \tag{18}$$

where η is the quantum efficiency of the detector, and

$$g^{(2)}(t)=g^{(2)}(\mathbf{R}_1,t;\mathbf{R}_2,t)=\frac{\langle E^{(-)}(\mathbf{R}_1,t)E^{(-)}(\mathbf{R}_2,t)E^{(+)}(\mathbf{R}_2,t)E^{(+)}(\mathbf{R}_1,t)\rangle}{\langle E^{(-)}(\mathbf{R}_1,t)E^{(+)}(\mathbf{R}_1,t)\rangle\langle E^{(-)}(\mathbf{R}_2,t)E^{(+)}(\mathbf{R}_2,t)\rangle} \tag{19}$$

is the normalized one-time second-order correlation function. Negative $Q(t,T)$ implies sub-Poissonian photon statistics; positive $Q(t,T)$ means that the photon statistics are super-Poissonian. It is evident from (18) that the statistics will be sub-Poissonian when $g^{(2)}(t)<1$ and super-Poissionian when $g^{(2)}(t)>1$.

The insertion of (14) into (19) and (19) into (18) (and assuming that the fluorescent photons are detected in the direction $\hat{\mathbf{R}}=\hat{\mathbf{R}}_1=\hat{\mathbf{R}}_2$ perpendicular to $\mathbf{r}_{12}$) produces

$$Q(t,T)=\eta T\frac{4Y_9(t)}{[Y_3(t)+Y_4(t)]^2}\ . \tag{20}$$

The time-dependent parameter (20), which describes the statistics of photons emitted from two atoms by resonance fluorescence, is shown in Fig. 5 as a function of time. Different interatomic separations are considered for parallel and perpendicular orientations of the atoms with reference to the external driving field. These graphs

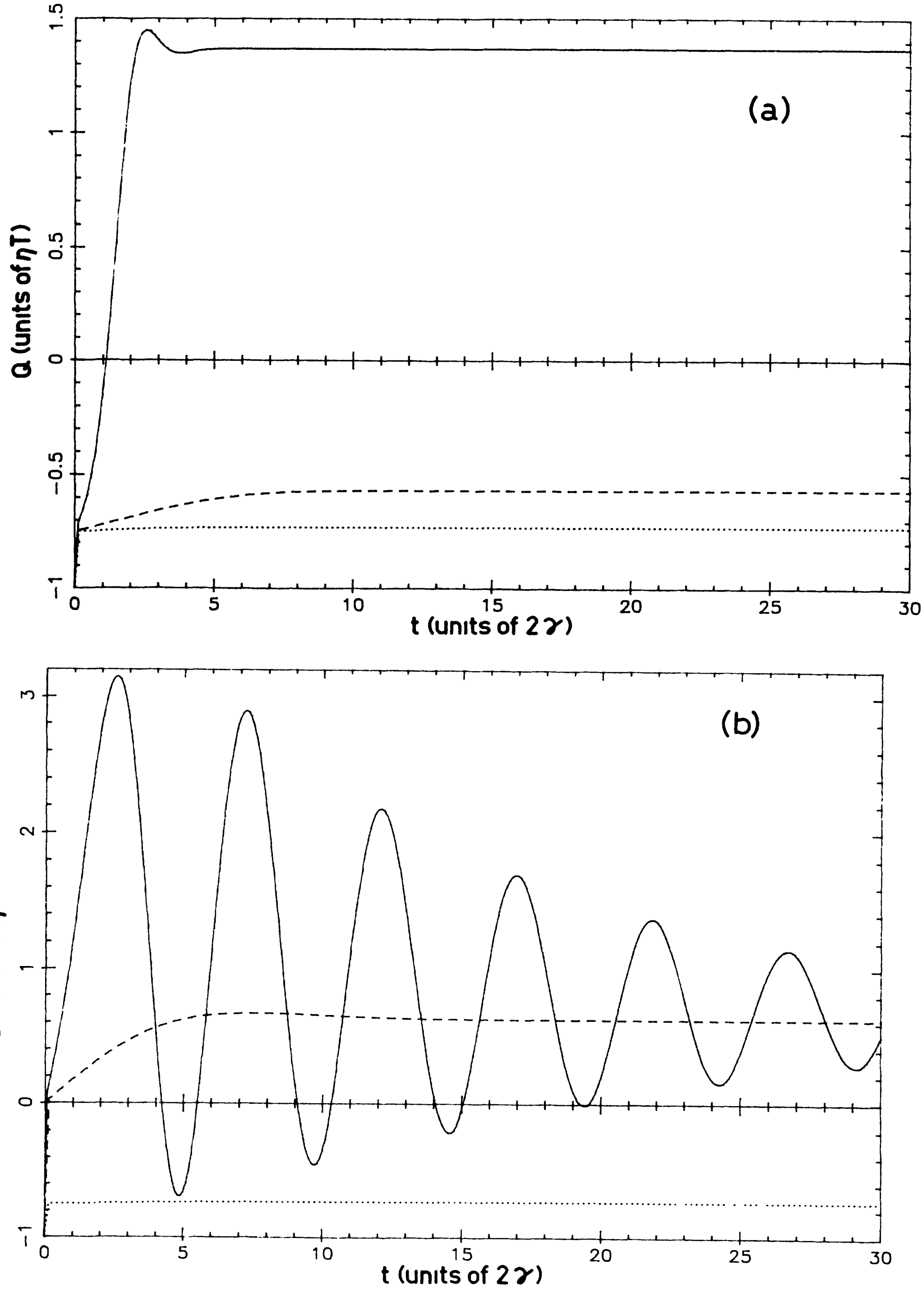

FIG. 5. Time-dependent photon statistics parameter $Q(t,T)$ (in units of ηT) as a function of $2\gamma t$ for $\beta=0.1$ and for the interatomic axis (a) perpendicular and (b) parallel to the driving field. The three interatomic separations presented are for $r_{12}=\lambda_0/8$ (solid line), $r_{12}=3\lambda_0/8$ (dashed line), and $r_{12}=50\lambda_0$ (dotted line).

show that the photon statistics strongly depend on the interatomic separation and the atomic orientation in the external driving field. For noninteracting atoms ($kr_{12} \gg 1$), the photon statistics are sub-Poissonian, independent of the atomic orientation. As the interatomic separation decreases, the photon statistics change from sub-Poissonian to super-Poissonian, in the case that the atoms are oriented perpendicular to the driving field [Fig. 5(a)]. However, small interatomic separations do not prevent sub-Poissonian statistics from arising as a transient phenomenon, when the atoms are oriented parallel to the driving field, as shown in Fig. 5(b) for $r_{12} = \lambda_0/8$. For the case $r_{12} = \lambda_0/8$, we observe the signature of quantum beats in the photon statistics for a parallel orientation.

V. CONCLUSIONS

In this paper we have been especially concerned with time-dependent resonance fluorescence from two atoms, separated by r_{12} comparable to the resonant wavelength. Particular attention has been paid to the effects of the atomic line orientation in the external driving field on the time-dependent resonance fluorescence intensity and photon statistics.

We have shown that by not assuming that the atoms are perpendicular to the driving field, which is a common assumption,[27–30] quantum beats appear in the time-dependent resonance fluorescence intensity for a weak driving field. These beats vanish for noninteracting atoms as well as for atoms separated by r_{12} much smaller than the resonant wavelength. For the interatomic separations comparable to the resonant wavelength, quantum beats are well resolved. Further analysis of the photon statistics of the fluorescent radiation demonstrates the presence of quantum beats for the case of the external driving field not propagating perpendicular to the interatomic axis. The photon statistics depend on the interatomic separation and on the interatomic axis orientation in the external driving field. For noninteracting atoms the photon statistics are sub-Poissonian, but super-Poissonian statistics arise for small interatomic separations. However, a parallel orientation of the atoms in the driving field produces transient sub-Poissonian statistics for small separations due to quantum beats.

We have studied the transient features of the resonance fluorescence intensity and photon statistics as they depend on the interatomic separation and on the atomic orientation in the driving field. Experimental detection of these interference effects is desirable, but it is an open question as to whether these effects are accessible to observation. Possible realizations of the dynamics discussed here include experiments with two trapped ions[19,38] or with two atoms fixed either in organic layers[39] or on the surface of a plane dielectric plate.[40,41] We hope that this research will contribute towards the clarification of collective effects in multiatom systems and will prove useful in designing future experiments.

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