Intensity-intensity correlations and quantum interference in a driven three-level atom

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(Received 1 October 1999; published 15 March 2000)

We investigate the two-time intensity correlation functions of the fluorescence field emitted from a V-type three-level atom. We are particularly interested in the manner in which the atom emits photons in the presence of quantum interference. We show that under strong-field excitation quantum interference leads to anticorrelations of photons emitted from the atomic excited levels which can exist for extremely long times. This indicates that the excited atomic levels are not the preferred radiative states. We find that the atom spends most of its time in a superposition of the excited atomic levels from which it emits strongly correlated photons. The strong correlations are present only for a nonzero splitting between the excited levels, and for degenerate levels the correlations reduce to that of a two-level atom. Moreover, we find that the transition from the ground level to the symmetric superposition of the excited levels does not saturate even for a strong driving field. We also calculate the correlation functions for a weak driving field, and find that in this case the photon correlations are not significantly affected by quantum interference, but the atom can emit a strongly correlated pair of photons produced by a three-wave mixing process. Under appropriate conditions, with near-maximal quantum interference, it is possible to make the maximum value of the correlation function extremely large, in marked contrast with the corresponding case with no quantum interference.

PACS number(s): 32.80.Bx, 42.50.Gy, 42.50.Lc

I. INTRODUCTION

One of the most interesting developments in the area of atomic and molecular spectroscopy is the possibility of modifying spontaneous emission through the mechanism of quantum interferences. The phenomenon, first predicted by Agarwal [1] in a degenerate three-level V-type system, results from vacuum-induced coherences between two atomic transitions: the spontaneous emission from one of the transitions modifies the spontaneous emission of the other transition. Various atomic and molecular schemes have been studied, and the results demonstrate that quantum interference can lead to many effects which could have useful applications in spectroscopy and laser physics. Examples include a quenching of spontaneous emission [2–4], electromagnetically induced transparency [5], and amplification without population inversion [6]. Recent studies have also shown that quantum interference can lead to phase-dependent population inversions and phase control of spontaneous emission [7]. Keitel [8] proposed a scheme to control the intensity of very narrow spectral lines in a V-type system driven from a single auxiliary level, which could have applications in high-precision spectroscopy.

Here we are concerned with the effects of quantum interference on the intensity-intensity correlations in a three-level V-type atom consisting of two excited levels coupled to a singlet ground level by electric dipole interactions. The atom is driven by a single-mode laser coupled to both atomic transitions, as shown in Fig. 1. These correlations were investigated by Hegerfeldt and Plenio [9] for an incoherently driven atom. The results show that the intensity correlation may exhibit quantum beats despite the incoherent pumping. The case of excitation by two coherent fields was considered by Manka *et al.* [10], who showed how the resonance fluorescence and intensity-intensity correlation spectra on one transition can be influenced by the intensity of the driving field on the other transition [11]. In particular, they demonstrated that the decay rate of the intensity-intensity correlation spectrum could be reduced in this way. This is the counterpart of the line narrowing observed in the fluorescence spectra [12]. Jagatap *et al.* [13] and Huang *et al.* [14] also calculated the intensity correlations in a three-level ladder system driven by two coherent fields, and showed that the correlations can have secondary oscillations, in addition to the Rabi oscillations.

In this paper we concentrate on the role of quantum interference in the correlation of photons emitted from a coherently driven V-type atom. We find that in the presence of quantum interference there are extended *simultaneous* periods of darkness in the fluorescence from the two atomic transitions, even for equal decay rates of the excited levels. This is in contrast to the dark periods predicted by Cook and Kimble [15] and Pegg *et al.* [16] for a V-type atom with uncorrelated transitions and significantly different decay rates γ_1 and γ_2. In their case the atom ''prefers'' to stay in the transition with the larger decay rate (strong transition), and there is a small probability of finding the system in the other (weak) transition. We show that in the presence of quantum interference and a strong driving field, the atom occupies superposition states rather than the bare atomic lev-

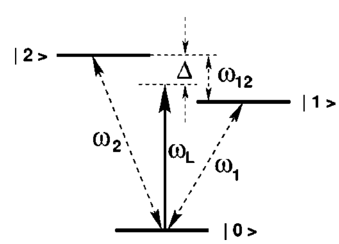

FIG. 1. Energy-level scheme of a three-level atom in the V configuration driven by a single laser field coupled to both atomic transitions.

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els, and emits a stream of photons exhibiting strong correlations. For a weak driving field, quantum interference does not significantly affect the photon correlations, but the atom can still emit strongly correlated pairs of photons, resulting from a three-wave-mixing process. Under appropriate conditions, the maximum value of the normalized, second-order field correlation function can be made huge (values of the order of hundreds or thousands) under conditions of quantum interference, whereas the corresponding maximum in the absence of quantum interference is ''normal'' (values of the order of unity). We are not aware of such large values being previously reported in the literature for single atoms, but indefinitely large correlation functions for two two-level atoms were reported by Wiegand [17]. The origin of the effect in this case is different to that in our situation, because it arises from the fact that, in the three-dimensional problem with two atoms, there are positions where the field vanishes.

II. SECOND-ORDER CORRELATION FUNCTIONS

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

$$g^{(2)}(\vec{r},t;\vec{r},t+\tau)=\frac{G^{(2)}(\vec{r},t;\vec{r},t+\tau)}{G^{(1)}(\vec{r},t)G^{(1)}(\vec{r},t+\tau)} \tag{1}$$

for the fluorescent field emitted from a three-level V-type atom driven by a coherent laser field and observed by a single detector located at a point $\vec{r}=r\hat{r}$, where $\hat{r}$ is the unit vector in the direction of the observation. The energy-level scheme of the atom is shown in Fig. 1. The atom consists of two nondegenerate excited levels $|1\rangle$ and $|2\rangle$ separated from the ground level $|0\rangle$ by transition frequencies ω_1 and ω_2, and connected by the electric dipole moments $\vec{\mu}_1$ and $\vec{\mu}_2$, respectively. The transition between the excited levels is forbidden in the electric dipole approximation.

The first- and second-order correlation functions, appearing in Eq. (1), can be expressed in terms of the positive and negative frequency parts of the electric-field operator as

$$G^{(1)}(\vec{r},t)=\left(\frac{r^2c}{2\pi\omega_0}\right)\langle\vec{E}^{(+)}(\vec{r},t)\vec{E}^{(-)}(\vec{r},t)\rangle, \tag{2}$$

$$G^{(2)}(\vec{r},t;\vec{r},t+\tau)=\left(\frac{r^2c}{2\pi\omega_0}\right)^2\langle\vec{E}^{(+)}(\vec{r},t)\vec{E}^{(+)}(\vec{r},t+\tau)\times\vec{E}^{(-)}(\vec{r},t+\tau)\vec{E}^{(-)}(\vec{r},t)\rangle, \tag{3}$$

where $\omega_0=(\omega_1+\omega_2)/2$. In Eqs. (2) and (3), we have introduced a factor $(r^2c/2\pi\omega_0)$ such that $G^{(1)}(\vec{r},t)d\Omega_r dt$ is the probability of finding a photon inside the solid angle $d\Omega_r$, around the direction $\vec{r}$ in the time interval dt at the time t, and $G^{(2)}(\vec{r},t;\vec{r},t+\tau)d^2\Omega_r dt d\tau$ is the probability of finding one photon inside the solid angle $d\Omega_r$ in the time interval dt at the time t and another photon inside the same solid angle in the time interval $d\tau$ at the time $t+\tau$.

In the far-field zone, $r\gg c/\omega_0$, and $t>r/c$, the positive frequency part of the electric field operator is given by

$$\vec{E}^{(+)}(\vec{r},t)=\vec{E}_0^{(+)}(\vec{r},t)-\frac{1}{c^2}\sum_{i=1}^{2}\frac{\hat{r}\times(\hat{r}\times\vec{\mu}_i)}{r}\omega_i^2A_{i0}(t-r/c), \tag{4}$$

where $A_{i0}=|i\rangle\langle 0|$ is the dipole operator of the transitions between the excited and ground levels.

Since the field is initially in the vacuum state, the vacuum part $\vec{E}_0^{(+)}(\vec{r},t)$ does not contribute to the expectation values of the normally ordered field operators, and then we obtain the following expression for the correlation functions:

$$G^{(1)}(t)\equiv G^{(1)}(\vec{r},t)=u(\vec{r})\sum_{i,j=1}^{2}\gamma_{ij}\langle A_{i0}(t)A_{0j}(t)\rangle \tag{5}$$

and

$$G^{(2)}(t,\tau)\equiv G^{(2)}(\vec{r},t;\vec{r},t+\tau)=u^2(\vec{r})\sum_{i,j,k,l=1}^{2}\gamma_{il}\gamma_{jk}\langle A_{i0}(t)A_{j0}(t+\tau)\times A_{0k}(t+\tau)A_{0l}(t)\rangle, \tag{6}$$

where $\gamma_{ii}=\gamma_i$ is the spontaneous decay constant of the excited sublevel $|i\rangle$ $(i=1,2)$ to the ground level $|0\rangle$, while

$$\gamma_{ij}=\frac{2\sqrt{\omega_i^3\omega_j^3}}{3\hbar c^3}\vec{\mu}_i\cdot\vec{\mu}_j=\beta\sqrt{\gamma_i\gamma_j}\quad(i\neq j) \tag{7}$$

arises from the cross-damping (quantum interference) between the transitions $|1\rangle\rightarrow|0\rangle$ and $|2\rangle\rightarrow|0\rangle$. The cross-damping term is sensitive to the mutual orientation of the atomic transition dipole moments, which is represented here by the parameter β. If the dipole moments are parallel, $\beta=1$, and the cross-damping term is maximal with $\gamma_{12}=\sqrt{\gamma_1\gamma_2}$, while $\gamma_{12}=0$ if the dipole moments are perpendicular $(\beta=0)$.

In Eqs. (5) and (6), $u(\vec{r})$ is a constant such that $u^2(\vec{r})=1$ for a random orientation of the atomic dipole moments with respect to the direction of observation $\vec{r}$, whereas

$$u^2(\vec{r})=\frac{3}{8\pi}\sin^2\Theta \tag{8}$$

for a fixed orientation, with Θ the angle between $\vec{r}$ and $\vec{\mu}$.

It is easily seen that the second-order correlation function [Eq. (6)] contains various two-time atomic correlation functions of the form $\langle A_{i0}(t)A_{jj}(t+\tau)A_{0i}(t)\rangle$ which are proportional to the probabilities of detecting two photons emitted from the same $(i=j)$ or different $(i\neq j)$ atomic transitions. For example, $\langle A_{20}(t)A_{11}(t+\tau)A_{02}(t)\rangle$ is proportional to the probability of detecting a photon at time $t+\tau$ emitted from the transition $|1\rangle \rightarrow |0\rangle$ if a photon emitted from the transition $|2\rangle \rightarrow |0\rangle$ was detected at time t.

Function (6) also depends, through the cross-damping term, on correlation functions of the form $\langle A_{i0}(t)A_{jk}(t+\tau)A_{0l}(t)\rangle$ ($i\neq l$ and/or $j\neq k$), which result from correlations of photons emitted from a superposition of the excited levels. Therefore, we introduce symmetric and antisymmetric superposition states

$$|s\rangle = \frac{1}{\sqrt{\gamma_1+\gamma_2}}(\sqrt{\gamma_1}|1\rangle + \sqrt{\gamma_2}|2\rangle), \tag{9}$$

$$|a\rangle = \frac{1}{\sqrt{\gamma_1+\gamma_2}}(\sqrt{\gamma_2}|1\rangle - \sqrt{\gamma_1}|2\rangle), \tag{10}$$

in terms of which the correlation functions (5) and (6) can be written as

$$\begin{aligned} G^{(1)}(t) = \frac{u(\vec{r})}{(\gamma_1+\gamma_2)} \{ & (\gamma_1^2+\gamma_2^2+2\beta\gamma_1\gamma_2)\langle A_{s0}(t)A_{0s}(t)\rangle \\ & +2(1-\beta)\gamma_1\gamma_2\langle A_{a0}(t)A_{0a}(t)\rangle \\ & +(1-\beta)\sqrt{\gamma_1\gamma_2}(\gamma_1-\gamma_2)\langle A_{s0}(t)A_{0a}(t) \\ & +A_{a0}(t)A_{0s}(t)\rangle\} \end{aligned} \tag{11}$$

and

$$\begin{aligned} G^{(2)}(t,\tau) = \frac{u^2(\vec{r})}{(\gamma_1+\gamma_2)^2} \{ & (\gamma_1^2+\gamma_2^2+2\beta\gamma_1\gamma_2)\langle A_{s0}(t) \\ & \times U(t+\tau)A_{0s}(t)\rangle + (1-\beta)\sqrt{\gamma_1\gamma_2} \\ & \times[2\sqrt{\gamma_1\gamma_2}\langle A_{a0}(t)U(t+\tau)A_{0a}(t)\rangle + (\gamma_1-\gamma_2) \\ & \times\langle A_{s0}(t)U(t+\tau)A_{0a}(t) \\ & +A_{a0}(t)U(t+\tau)A_{0s}(t)\rangle]\}, \end{aligned} \tag{12}$$

where

$$\begin{aligned} U(t+\tau) = & (\gamma_1^2+\gamma_2^2+2\beta\gamma_1\gamma_2)A_{s0}(t+\tau)A_{0s}(t+\tau) \\ & +(1-\beta)\sqrt{\gamma_1\gamma_2}\{2\sqrt{\gamma_1\gamma_2}A_{a0}(t+\tau)A_{0a}(t+\tau) \\ & +(\gamma_1-\gamma_2)[A_{s0}(t+\tau)A_{0a}(t+\tau) \\ & +A_{a0}(t+\tau)A_{0s}(t+\tau)]\}. \end{aligned} \tag{13}$$

Using the bases of the symmetric and antisymmetric states, there are three terms contributing to the first- and second-order correlation functions. In the expression for $G^{(1)}(t)$, the first term arises from the transition $|s\rangle \rightarrow |0\rangle$, the second from the transition $|a\rangle \rightarrow |0\rangle$, and the third from the coupling between them. When the decay rates are equal, $\gamma_1 = \gamma_2$, then the transitions are independent regardless of the mutual orientation of the atomic transition dipole moments. In the expression for $G^{(2)}(t)$, the first term arises from processes in which the first transition is $|s\rangle \rightarrow |0\rangle$, the second term arises from processes in which the first transition is $|a\rangle \rightarrow |0\rangle$, and the third term is due to the coupling between them. Moreover, for parallel dipole moments ($\beta=1$) only the transition $|s\rangle \rightarrow |0\rangle$ contributes to the fluorescence intensity and the second-order correlation function, indicating that in this case the system reduces to a two-level system. However, correlations between the emitted photons can be significantly different from those one would expect for a two-level system.

To show this, we consider the two-time normalized second-order correlation function of the fluorescence field emitted by the atom. According to Eqs. (5) and (6), the two-time correlation function is proportional to the two-time correlation functions of the atomic operators, which we can find from the master equation of the system and the quantum regression theorem [18]. In the frame rotating with the laser frequency ω_L the master equation is of the form

$$\dot{\rho} = -i[\rho,H] + \mathcal{L}\rho, \tag{14}$$

where the Hamiltonian is

$$H = (\Delta-\omega_{12})A_{11} + \Delta A_{22} + [(\Omega_1 A_{10} + \Omega_2 A_{20}) + \text{H.c.}], \tag{15}$$

and the damping term is

$$\begin{aligned} \mathcal{L}\rho = & \frac{1}{2}\gamma_1(2A_{01}\rho A_{10} - A_{11}\rho - \rho A_{11}) \\ & + \frac{1}{2}\gamma_2(2A_{02}\rho A_{20} - A_{22}\rho - \rho A_{22}) \\ & + \frac{1}{2}\gamma_{12}(2A_{01}\rho A_{20} - A_{21}\rho - \rho A_{21}) \\ & + \frac{1}{2}\gamma_{12}(2A_{02}\rho A_{10} - A_{12}\rho - \rho A_{12}). \end{aligned} \tag{16}$$

In Eq. (15), $\Delta = \omega_2 - \omega_L$ is the detuning between the frequency ω_2 of the $|0\rangle \rightarrow |2\rangle$ transition and the driving laser frequency, $2\Omega_k (k=1,2)$ is the Rabi frequency of the kth transition, and ω_{12} is the level splitting between the excited sublevels. Here we assume that the excited sublevels can decay to the level $|0\rangle$ by spontaneous emission, whereas direct spontaneous transitions between the excited sublevels are dipole forbidden.

The master equation (14) leads to the following equations of motion for the density matrix elements:

$$\dot{\rho}_{10}=(\dot{\rho}_{01})^{*}=-i\Omega_1-\left[\frac{1}{2}\gamma_1+i(\Delta-\omega_{21})\right]\rho_{10}-\frac{1}{2}\gamma_{12}\rho_{20}+i\Omega_2\rho_{12}+i\Omega_1(2\rho_{11}+\rho_{22}),$$

$$\dot{\rho}_{20}=(\dot{\rho}_{02})^{*}=-i\Omega_2-\left(\frac{1}{2}\gamma_2+i\Delta\right)\rho_{20}-\frac{1}{2}\gamma_{12}\rho_{10}+i\Omega_1\rho_{21}+i\Omega_2(2\rho_{22}+\rho_{33}),$$

$$\dot{\rho}_{21}=(\dot{\rho}_{12})^{*}=-\left[\frac{1}{2}(\gamma_1+\gamma_2)+i\omega_{21}\right]\rho_{21}-\frac{1}{2}\gamma_{12}(\rho_{22}+\rho_{11})+i\Omega_1\rho_{20}-i\Omega_2\rho_{01},$$

$$\dot{\rho}_{11}=-\gamma_1\rho_{11}-\frac{1}{2}\gamma_{12}(\rho_{12}+\rho_{21})-i\Omega_1(\rho_{01}-\rho_{10}),$$

$$\dot{\rho}_{22}=-\gamma_2\rho_{22}-\frac{1}{2}\gamma_{12}(\rho_{12}+\rho_{21})-i\Omega_2(\rho_{02}-\rho_{20}). \tag{17}$$

The set of equations (17) can be written in matrix form as

$$\dot{\vec{X}}(t)=M\vec{X}(t)+\vec{I}, \tag{18}$$

where $\vec{X}(t)$ is a column vector composed of the density-matrix elements, $\vec{I}$ is a column vector composed of the inhomogeneous terms, and M is an 8×8 matrix obtained from the coefficients appearing in the equations of motion (17). Since we are interested in the time evolution of the density-matrix elements, we will need explicit expressions for the components X_i of the vector $\vec{X}(t)$ in terms of their initial values. This can be done by a direct integration of Eq. (18). Thus, if t_0 denotes an arbitrary initial time, the integration of Eq. (18) leads to the following formal solution for $\vec{X}(t)$:

$$\vec{X}(t)=\vec{X}(t_0)e^{Mt}-(1-e^{Mt})M^{-1}\vec{I}. \tag{19}$$

Solution (19) for the density-matrix elements at time t allows us, by using the quantum regression theorem [18], to find the density-matrix elements at time $t+\tau$ in terms of those at time t. In the following sections, we will use solution (19) to calculate the two-time normalized second-order correlation function for a strong driving field as well as a weak driving field.

III. STRONG DRIVING FIELD

We first consider the second-order correlation function for the case of a strong driving field, and examine the effects of quantum interference on the photon correlations. We calculate the correlation function for the field emitted from the individual atomic transitions (distinguishable photons), as well as for the total emitted field (indistinguishable photons).

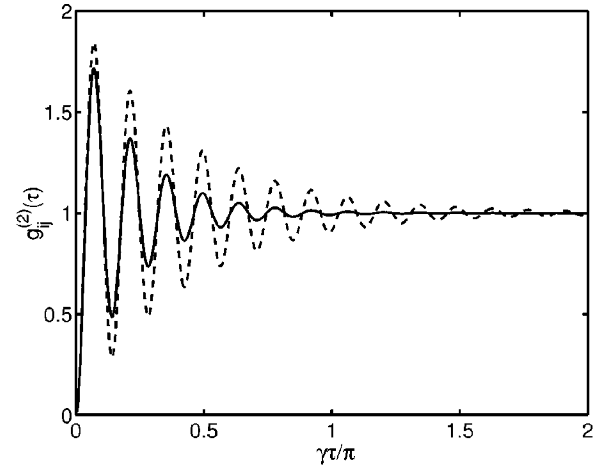

FIG. 2. Second-order correlation functions $g_{ij}^{(2)}(\tau)$ $(i,j=1,2)$ for the case of distinguishable photons. In this and subsequent figures, we take $\gamma_1=\gamma_2=\gamma$, and measure all quantities in terms of γ. In this figure we assume $\omega_{12}=0$, $\Delta=0$, and $\Omega=5\gamma$, and plot graphs for two different values of β: $\beta=0.99$ (solid line) and $\beta=0$ (dashed line).

A. Distinguishable photons

If the photons emitted from the excited states to the ground state are distinguishable, e.g., by having significantly different polarizations or frequencies, then the following normalized second-order correlation functions of the steady-state fluorescence intensity can be defined [14]:

$$g_{ij}^{(2)}(\tau)=\lim_{t\to\infty}g^{(2)}(\vec{r},t;\vec{r},t+\tau)=\frac{P_{0\to j}(\tau)}{P_j},\quad i,j=1,2,$$

where

$$P_{0\to j}(\tau)=\frac{\langle A_{i0}A_{j0}(\tau)A_{0j}(\tau)A_{0i}\rangle}{\langle A_{i0}A_{0i}\rangle} \tag{20}$$

is the probability that at time $t+\tau$ the atom is in the upper state $|j\rangle$ of the transition $|j\rangle\to|0\rangle$ if it was in the lower state $|0\rangle$ of the $|i\rangle\to|0\rangle$ transition at time t, and $P_i=\langle A_{i0}A_{0i}\rangle$ is the steady-state population of the state $|i\rangle$. In particular, we consider the following correlation functions:

$$g_{11}^{(2)}(\tau)=g_{21}^{(2)}(\tau)=\frac{P_{0\to 1}(\tau)}{P_1}, \tag{21}$$

$$g_{22}^{(2)}(\tau)=g_{12}^{(2)}(\tau)=\frac{P_{0\to 2}(\tau)}{P_2}. \tag{22}$$

In Fig. 2 we show the correlation functions (21) and (22) for $\gamma_1=\gamma_2$, $\omega_{12}=0$, $\Omega_1=\Omega_2=\Omega=5\gamma_1$, and $\Delta=0$, and two different values of β: $\beta=0$, corresponding to the case of perpendicular dipole moments; and $\beta=0.99$, corresponding to almost parallel dipole moments. We have chosen $\beta<1$ to avoid population trapping, which can appear for $\beta=1$ [1,2,4]. The correlations show the characteristic Rabi oscillations, which indicate that the detection of a photon at time

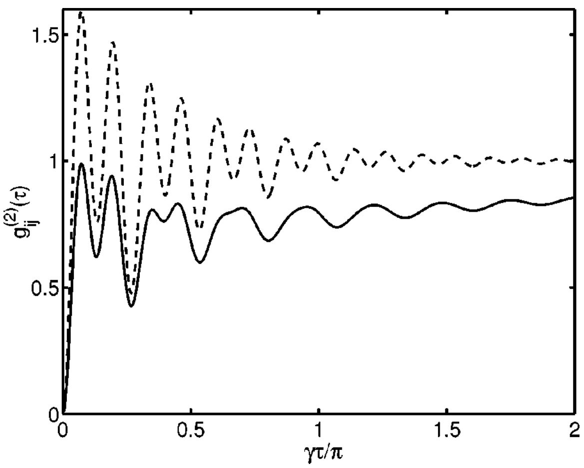

FIG. 3. Same as in Fig. 2, but $\omega_{12}=5\gamma$ and $\Delta=\omega_{12}/2$.

τ, after the detection of a photon at time 0, is impossible if $\tau=0$, and is unlikely until τ increases to a value of the order of $(2\Omega)^{-1}\pi$. For both values of β the correlation function oscillates with the Rabi frequency of the driving field and there is little difference between the plots for $\beta=0$ and 0.99. The shape of the oscillations resembles that known for a two-level atom, which indicates that the atomic dipole moments oscillate independently, regardless of the value of β.

In Fig. 3, we show the correlation functions for the same parameters as in Fig. 2, but now $\omega_{12}=5\gamma_1$ and $\Delta=\omega_{12}/2$. Here the behavior of the correlation functions is qualitatively different to the case where $\omega_{12}=0$. For correlated dipole moments with $\beta=0.99$, the values of $g^{(2)}_{11}(\tau)$ and $g^{(2)}_{22}(\tau)$ remain below unity for all times. This shows that for any τ the probability of emission of two photons from levels $|1\rangle$ or $|2\rangle$ is very small. We can interpret this as extended *simultaneous* periods of darkness in the fluorescence from the two atomic transitions: after detection of a photon at time $\tau=0$, detection of another photon at time $\tau>0$, emitted from levels $|1\rangle$ or $|2\rangle$, is very unlikely. We point out that the simultaneous periods of darkness appear only for correlated transitions with $\beta\neq 0$. Dark periods of fluorescence were predicted before [15,16] in a three-level atom with $\beta=0$ and significantly different transition rates γ_1 and γ_2. However, the predicted dark periods appear on only one of the two atomic transitions, whereas the extended dark periods, predicted here for the correlated transitions, appear simultaneously on both transitions. This indicates that in the presence of quantum interference the atomic states $|1\rangle$ and $|2\rangle$ are not the preferred radiative states of the atom.

It is apparent that there are oscillations at more than one frequency present in Fig. 3. In fact, there are oscillations at the Rabi frequency 2Ω as well as at Ω. The origin of these frequencies is discussed in Sec. IV.

B. Indistinguishable photons

We are concerned here with the situation in which the photons emitted from the two atomic transitions are not distinguishable. This can happen when the atomic transition dipole moments are exactly or almost parallel. Then the detector responds to the total field (4), for which the correlation functions are given by Eqs. (11) and (12). However, even for $\beta\approx 1$ we can still distinguish between photons emitted from the $|s\rangle\rightarrow|0\rangle$ and $|a\rangle\rightarrow|0\rangle$ transitions, as they can have different polarizations. It is easy to see from Eqs. (9) and (10) that the dipole moments $\vec{\mu}_s$ and $\vec{\mu}_a$ of the respective $|s\rangle\rightarrow|0\rangle$ and $|a\rangle\rightarrow|0\rangle$ transitions are oriented in different directions unless $\vec{\mu}_1=\vec{\mu}_2$, and then $\vec{\mu}_a=0$.

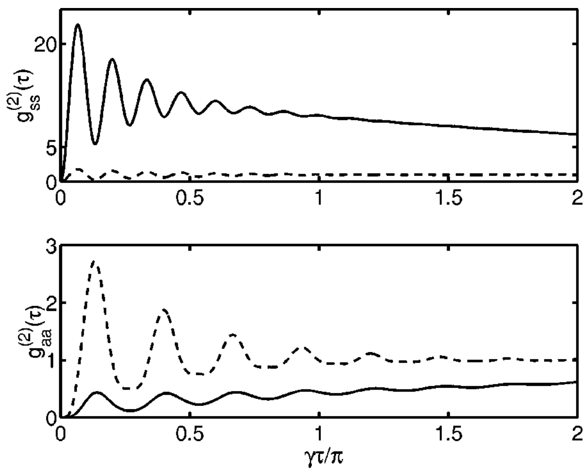

FIG. 4. Second-order correlation functions for the case of indistinguishable photons for $\omega_{12}=5\gamma$, $\Delta=\omega_{12}/2$, and $\Omega=5\gamma$. In the upper plot we present $g^{(2)}_{ss}(\tau)$, and in the lower plot $g^{(2)}_{aa}(\tau)$. The solid line is for $\beta=0.99$, and the dashed line for $\beta=0$. If we change β and Δ to $\beta=1$ and $\Delta=0.4\omega_{12}/2$, the graphs are almost identical.

Therefore, we separately consider the following correlation functions:

$$g^{(2)}_{ss}(\tau)=\frac{P_{0\rightarrow s}(\tau)}{P_s}, \tag{23}$$

$$g^{(2)}_{aa}(\tau)=\frac{P_{0\rightarrow a}(\tau)}{P_a}. \tag{24}$$

In essence, the correlation function $g^{(2)}_{ss}(\tau)$ corresponds to that of the total fluorescence field, as the contribution from the asymmetric state, which is proportional to $(1-\beta)$, is negligible for $\beta\approx 1$.

In Fig. 4, we plot the correlation functions (23) and (24) for $\gamma_1=\gamma_2$, $\Omega_1=\Omega_2=\Omega=5\gamma_1$, $\omega_{12}=5\gamma_1$, and $\Delta=\omega_{12}/2$. Again, the solid line is for $\beta=0.99$ and the dashed line for $\beta=0$. It is apparent from the graphs that with quantum interference ($\beta=0.99$), there are very strong correlations of photons on the $|s\rangle\rightarrow|0\rangle$ transition, whereas the photons are strongly anticorrelated on the $|a\rangle\rightarrow|0\rangle$ transition. The correlation function $g^{(2)}_{ss}(\tau)$ oscillates with the frequency $2\sqrt{2}\Omega$, which is the Rabi frequency in the symmetric basis, and attains a maximum value at time $\tau=(2\sqrt{2}\Omega)^{-1}\pi$. Moreover, the correlations decay at a very low rate, and it takes a time in excess of $300\pi/\gamma_1$ before $g^{(2)}$ is close to unity. The correlation function $g^{(2)}_{aa}(\tau)$ oscillates with frequency $\sqrt{2}\Omega$, and, in the presence of quantum interference, is less than unity for all times, whereas for $\beta=0$ the values can exceed

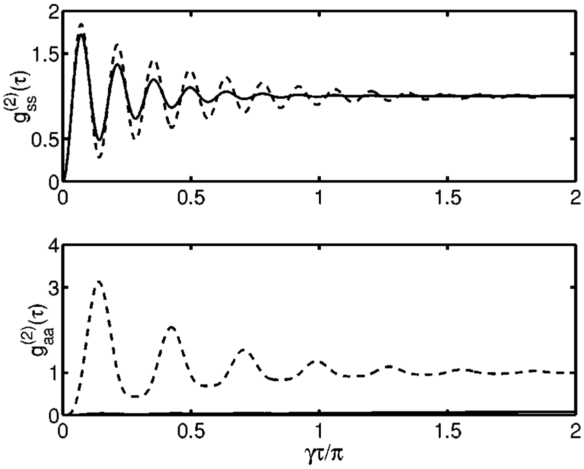

FIG. 5. Same as in Fig. 4, but $\omega_{12}=0.1\gamma$.

unity, with a maximum value of around 2.8. It is worth pointing out that very large values of $g_{ss}^{(2)}(\tau)$ are possible for $\beta\simeq 1$, whereas the maximum value of $g_{ss}^{(2)}(\tau)$ remains of the order of unity for $\beta=0$. Thus in Fig. 4 it is seen that the maximum value is about 22.5 for $\beta\simeq 1$. Even larger values are possible: if we reduce the value of Ω to $0.5\gamma_1$, leaving other parameters unchanged, then the maximum value increases to almost 1500.

If we reduce the value of ω_{12}, the difference between the $\beta=0$ and 0.99 graphs for $g_{ss}^{(2)}(\tau)$ becomes less pronounced. This is shown in Fig. 5, where we plot $g_{ss}^{(2)}(\tau)$ for the same parameters as in Fig. 4, but $\omega_{12}=0.1\gamma_1$. We see that indeed for sufficiently small ω_{12} the correlation functions for $\beta=0$ and 0.99 oscillate in a similar fashion with $g_{ss}^{(2)}(\tau)<2$ for all times τ. The dominant frequency in $g_{ss}^{(2)}(\tau)$ is the Rabi frequency $2\sqrt{2}\Omega$, whereas in $g_{aa}^{(2)}(\tau)$ it is $\sqrt{2}\Omega$, as we discuss in Sec. IV.

IV. INTERPRETATION OF THE RESULTS

The effect of quantum interference on the second-order correlation function, shown in Figs. 2–5, is very sensitive to the splitting ω_{12} of the excited levels. For degenerate excited levels ($\omega_{12}=0$) or small splittings ($\omega_{12}\approx 0$), the photon emissions are similar to those of a two-level atom, independent of quantum interference. For large splittings, the correlation functions $g_{ij}^{(2)}(\tau)$ $(i,j=1,2)$ and $g_{aa}^{(2)}(\tau)$ are smaller than unity for all times τ, while $g_{ss}^{(2)}(\tau)$ exhibits strong correlations $[g_{ss}^{(2)}(\tau)\gg 2]$ for $\tau\approx(2\sqrt{2}\Omega)^{-1}\pi$, which decay at a very low rate.

We can explain these features by considering the master equation (14) and the equations of motion (17). For $\omega_{12}=0$ the states $|1\rangle$ and $|2\rangle$ are equally driven by the laser, and the coherences ρ_{10} and ρ_{20} oscillate in phase with frequency Δ. The coherences are directly coupled by the cross-damping term γ_{12}. However, for a strong driving field $(\Omega\gg\gamma_i,\gamma_{12})$ the Rabi oscillations dominate over the spontaneous exchange of photons, resulting in independent oscillations of the atomic dipole moments.

The situation is different when $\omega_{12}\neq 0$ and $\Delta=\omega_{12}/2$. In this case the coherences oscillate with opposite phases, indicating that there is an exchange of photons between states $|1\rangle$ and $|2\rangle$ which prevents photons being emitted from the atomic levels. The coherences oscillate with $\pm\omega_{12}/2$, which introduces the modulation of the Rabi oscillations seen in Fig. 3. The exchange of photons between the atomic levels is better seen in the basis of the symmetric and antisymmetric states (9) and (10). In terms of these states, setting $\gamma_1=\gamma_2=\gamma$ for simplicity, the master equation (14) and the Hamiltonian (15) take the form

$$\dot{\rho}=-i[\rho,H]+\frac{1}{2}\gamma(1+\beta)(2A_{0s}\rho A_{s0}-A_{ss}\rho-\rho A_{ss})+\frac{1}{2}\gamma(1-\beta)(2A_{0a}\rho A_{a0}-A_{aa}\rho-\rho A_{aa}), \tag{25}$$

with

$$H=\left(\Delta-\frac{1}{2}\omega_{12}\right)(A_{ss}+A_{aa})-\frac{1}{2}\omega_{12}(A_{sa}+A_{as})+\sqrt{2}\Omega(A_{s0}+A_{0s}), \tag{26}$$

where $\Omega=\Omega_1=\Omega_2$.

We see that the laser field couples only to the symmetric state, and both states decay independently to the ground state with different decay rates. For $\omega_{12}\neq 0$ the antisymmetric state is coupled to the symmetric state by the Hamiltonian H. This coupling introduces periodical oscillations of the population between the symmetric and antisymmetric states. This is seen in the equations of motion for the populations

$$\dot{\rho}_{ss}=-\frac{1}{2}\gamma(1+\beta)\rho_{ss}-\frac{1}{2}i\omega_{12}(\rho_{sa}-\rho_{as})-i\sqrt{2}\Omega(\rho_{s0}-\rho_{0s}), \tag{27}$$

$$\dot{\rho}_{aa}=-\frac{1}{2}\gamma(1-\beta)\rho_{aa}+\frac{1}{2}i\omega_{12}(\rho_{sa}-\rho_{as}). \tag{28}$$

It is evident that the antisymmetric state is populated by the coupling to the symmetric state. Since the decay rate of the antisymmetric state, $\gamma(1-\beta)$, is very small for $\beta\approx 1$, the population stays in this state for a long time. If $\omega_{12}=0$, the state is decoupled from the symmetric state, and $\rho_{aa}(t)$ is zero if its initial value is zero. In the latter case the system reduces to a two-level atom. In the former case the transfer of the population to a slowly decaying state leaves the symmetric state almost unpopulated even if the driving field is strong. This is shown in Fig. 6, where we plot the steady-state populations ρ_{ss}, ρ_{aa}, and ρ_{00} as functions of Δ for $\Omega=5\gamma_1$, $\omega_{12}=5\gamma_1$, and $\beta=0.99$. It is evident that the symmetric state is almost unpopulated for $\Delta=\omega_{12}/2$. This indicates that in the presence of quantum interference, the driving field does not saturate the transition $|0\rangle\to|s\rangle$, even for very large Rabi frequencies. The lack of population in the state $|s\rangle$ increases the probability of returning the atom to this state from the ground state by the driving field. Conse-

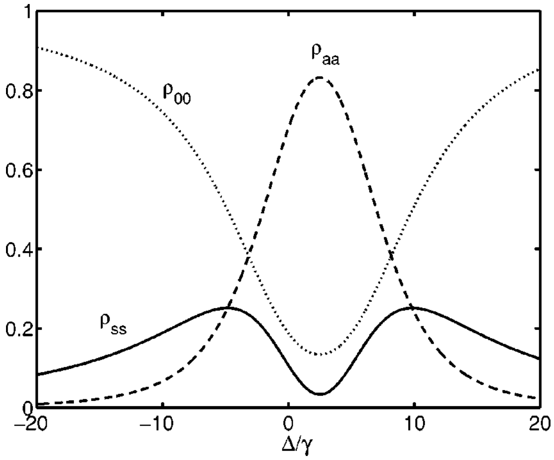

FIG. 6. The steady-state populations ρ_{ss}, ρ_{aa}, and ρ_{00} as functions of Δ/γ for $\Omega=5\gamma$, $\omega_{12}=5\gamma$, and $\beta=0.99$.

quently, $g_{ss}^{(2)}(\tau)$ attains a very large value at time $\tau=(2\sqrt{2}\Omega)^{-1}\pi$ corresponding to half of the Rabi cycle between $|0\rangle$ and $|s\rangle$. However, $g_{aa}^{(2)}(\tau)$ attains a maximum at $\tau\approx(\sqrt{2}\Omega)^{-1}\pi$, i.e., at the Rabi period. This results from the fact that the driving laser takes the population from $|0\rangle$ to $|s\rangle$ in a time equal to half of the Rabi period. Then the population can be transferred to $|a\rangle$ in a time equal to that in which the population will stay in $|s\rangle$, i.e., a time equal to half of the Rabi period. Therefore, the total time of transferring the population from $|0\rangle$ to $|a\rangle$ is equal to the Rabi period.

V. WEAK DRIVING FIELD

The previous discussion shows that in the presence of a strong driving field, quantum interference significantly affects the second-order correlation function of the emitted fluorescence field. Here we consider the correlation functions for a weak driving field. In Fig. 7, we plot $g_{ij}^{(2)}(\tau)(i,j=1,2)$ for $\gamma_1=\gamma_2$, $\Omega=0.5\gamma_1$, $\Delta=\omega_{12}/2$, and two values of ω_{12}. All four correlation functions are identical for these parameter values. For $\omega_{12}=0$ the correlation functions increase monotonically with τ, and there is not much difference between $\beta=0$ and 0.99. When ω_{12} is different from zero, there is a small difference between the $\beta=0$ and 0.99 plots, but what is interesting that the $g_{ij}^{(2)}(\tau)$ show strong correlations with a maximum greater than 2 at $\tau=(\omega_{12}/2)^{-1}\pi$. This is in contrast to the case of the strong driving field, shown in Fig. 3, where quantum interference leads to photon anticorrelation appearing for all τ. Thus, for a weak driving field, quantum interference does not significantly affect the correlation functions. The strong photon correlations, seen in Fig. 7, can be interpreted as arising from a three-wave mixing process, which dominates when the laser is detuned from the atomic transition frequencies. For example, the strong correlations in $g_{22}^{(2)}(\tau)$ result from an absorption from the laser field of two photons of frequency $\omega_2-\Delta$ and the emission of a correlated pair of photons of frequencies $\omega_L-\Delta$ and $\omega_L+\Delta$.

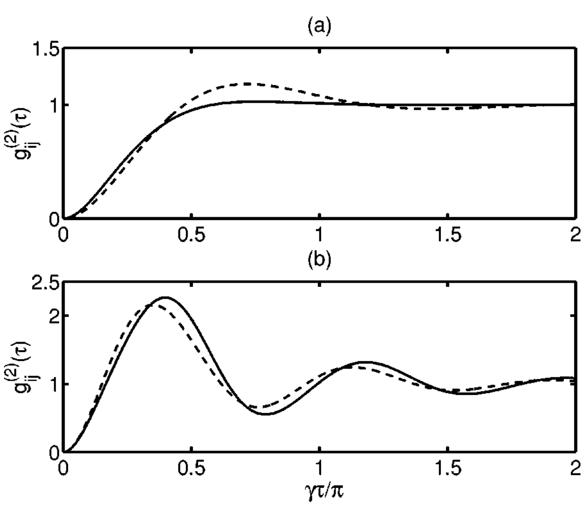

FIG. 7. Second-order correlation functions $g_{ij}^{(2)}(\tau)(i,j=1,2)$ for distinguishable photons with $\Omega=0.5\gamma$, $\Delta=\omega_{12}/2$, and two values of ω_{12}. In the upper plot $\omega_{12}=0$, and in the lower plot $\omega_{12}=5\gamma$. The solid line is for $\beta=0.99$, and the dashed line for $\beta=0$.

VI. SUMMARY

In this paper we have examined the effect of quantum interference on the two-time correlation functions of the fluorescence field emitted by a V-type three-level atom driven by a single-mode laser field. We have used the master equation of the system, and have applied the quantum regression theorem to calculate various correlation functions $g_{ij}^{(2)}(\tau)$. We have found that for the case of degenerate atomic transitions the photon correlations are not significantly affected by quantum interference. For nondegenerate transitions, the photon correlations depend strongly on the intensity of the driving field. When a strong driving field is tuned to the middle of the two excited levels, the correlations of photons emitted from the atomic transitions exhibit anticorrelations which persist for all times. Thus the excited atomic levels are not the preferred radiative states: the atom emits strongly correlated photons from a symmetric superposition of the excited levels. The correlations result from a coherent transfer of populations to the antisymmetric state, which leaves the symmetric state unpopulated even for very strong driving fields. For a weak driving field, the photon correlations are not strongly affected by quantum interference, but the atom can emit strongly correlated pairs of photons arising from a three-wave mixing process.

We conclude with a brief discussion on the possibility of experimental detection of these unusual features. The essential conditions for this is that the V system should be slightly detuned from the optimum conditions necessary for full quantum interference. In the figures, we have achieved this by assuming the dipole moments to be slightly misaligned from parallel ($\beta=0.99$), while the laser detuning is taken to be optimum for quantum interference: $\Delta=\omega_{12}/2$. It has been predicted that these conditions may be realizable in a cavity system [19]. An alternative arrangement would be to utilize parallel dipole moments ($\beta=1$) with the laser frequency detuned from the optimum for quantum interference. For example, in Fig. 4, we have taken $\beta=0.99$ and $\Delta=\omega_{12}/2$. We find that spectra — almost indistinguishable from those presented, at least for the first few Rabi oscillations — are obtained for the alternative parameter values $\beta = 1$ and $\Delta = 0.4 \times \omega_{12}/2$. A system with parallel dipole moments was employed in the experiments of Xia *et al.* [20]. It seems that an experimental observation of these effects is feasible.

ACKNOWLEDGMENTS

This research was supported by the United Kingdom Engineering and Physical Sciences Research Council. We acknowledge helpful conversations with S.-Y. Zhu and M. S. Zubairy.

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