Macroscopic quantum jumps from a two-atom system

K. Yamada and P. R. Berman
Department of Physics, New York University, 4 Washington Place, New York, New York 10003
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We analyze the macroscopic quantum jumps (sudden interruptions in the fluorescence on a macroscopic time scale) that are produced when a pair of two-level atoms separated by a distance d is irradiated by a strong laser having wavelength λ_0, with $\lambda_0 \gg d$. Included in the analysis is the dipole-dipole coupling of the atoms, the ac Stark effect, and the role played by a term that is present in the atom-laser field interaction Hamiltonian when $\mathbf{k}\cdot\mathbf{d}\neq 0$ ($\mathbf{k}$ is the wave vector of the laser field and $\mathbf{d}$ is the vector connecting the two atoms). Our treatment is based on frequency-resolved delay functions, an extension of a concept developed by Reynaud, Dalibard, and Cohen-Tannoudji [IEEE J. Quantum Electron. **24**, 1395 (1988)], which is shown to be useful to study frequency-resolved photon statistics. As examples, we study the statistics of the fluorescence produced by the two-atom system as well as those in the components of the fluorescent triplet produced by a single two-level atom.

I. INTRODUCTION

When identical two-level atoms are separated by a distance d that is smaller than their resonant wavelength λ_0, cooperative decay phenomena can occur. Dicke[1] and others[2,3] found that the exchange of photons between the two atoms produces new eigenstates with new decay rates. Denoting the ground and excited states of atom i by $|e_i\rangle$ and $|g_i\rangle$ $(i=1,2)$, these states are a triplet of symmetric states $[|E\rangle=|e_1e_2\rangle, |S\rangle=(1/\sqrt{2})(|e_1g_2\rangle+|g_1e_2\rangle), |G\rangle=|g_1g_2\rangle)$, and one antisymmetric state $(|A\rangle=(1/\sqrt{2})(|e_1g_2\rangle-|g_1e_2\rangle)]$. These states are shown in Fig. 1. When $\lambda_0 \gg d$, the system, initially excited by an incoherent or a weak coherent field to state $|E\rangle$, can decay to state $|G\rangle$ via state $|S\rangle$ with a rate $\Gamma_S \cong 2\Gamma$ (Γ is the decay rate of a single atom). As can be seen from Fig. 1, a two-peaked fluorescence spectrum centered at $\omega_0 \pm V$ is produced when the system undergoes the $|E\rangle \to |S\rangle \to |G\rangle$ cascade.

Cooperative effects in resonance fluorescence produced by the two-atom system when it is continuously excited by a strong coherent laser was studied by Senitzky[4] and others.[5] They searched for the existence of extra sidebands not present in the single-atom fluorescence spectrum (Mollow triplet).[6] An interpretation of the spectrum was provided by Freedhoff.[7] She calculated the fluorescence as arising from transitions between dressed states of the two-atom plus laser-field system and obtained a spectrum containing seven peaks.

In most treatments of the problem, the atoms have been considered to be so close as to render the antisymmetric state optically inactive in the sense that the decay rate Γ_A for the $|E\rangle \to |A\rangle$ and $|A\rangle \to |G\rangle$ transitions is set identically equal to zero. The decay rate Γ_A is approximately given by $\Gamma_A \cong (2\pi d/\lambda_0)^2\Gamma/5(\ll\Gamma)$.[2] As will be seen below, this small but finite decay rate can lead to macroscopic quantum jumps (MQJ).[8-14] The origin of the MQJ is the metastability of the antisymmetric state $|A\rangle$; once the system is shelved in this state the fluorescence produced by the $|E\rangle \to |S\rangle \to |G\rangle$ cascade is interrupted for a time interval on the order of Γ_A^{-1}.

In this paper, we analyze the MQJ produced by this cooperative atomic effect. Recently, this effect was partially incorporated into the problem "MQJ due to two three-level atoms" by Javanainen and Lewenstein.[15] We extend their work by including effects relating to the energy shifts of states $|S\rangle$ and $|A\rangle$ resulting from the atomic dipole-dipole interaction. Moreover, we allow for an additional mixing of the symmetric and antisymmetric states produced by a term in the laser-field–atom interaction Hamiltonian that is present when $\mathbf{k}\cdot\mathbf{d}\neq 0$ ($\mathbf{k}$ is the wave vector of the laser field and $\mathbf{d}$ is the vector connecting the two atoms). The former effect is important because the $|E\rangle \leftrightarrow |S\rangle$ and $|S\rangle \leftrightarrow |G\rangle$ transitions are no longer resonant with a laser tuned to ω_0; the energy shift

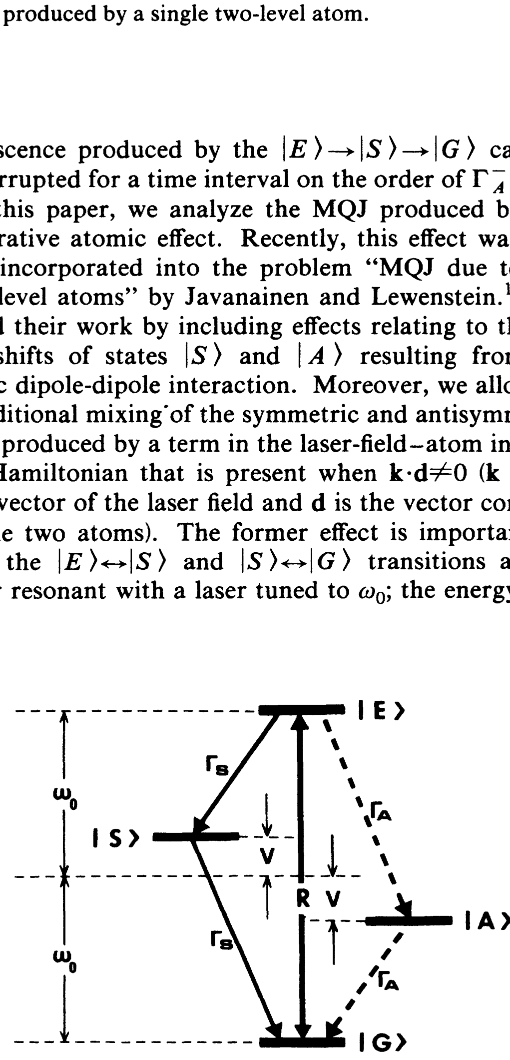

FIG. 1. Energy diagram for a two-atom composite system. When the atomic separation $d \ll \lambda_0 = 2\pi d/\omega_0$, $\Gamma_S \cong 2\Gamma$, $\Gamma_A \cong (2\pi d/\lambda_0)^2\Gamma/5$, and $V=(2\pi d/\lambda_0)^{-3}3\Gamma/4$, where Γ is the decay rate and ω_0 is the resonant frequency of a single atom. Transitions between states $|E\rangle$ and $|G\rangle$ (at rate R) are produced by an incoherent pump field. State $|A\rangle$ acts as a shelving state in the problem of macroscopic quantum jumps.

$V=(2\pi d/\lambda_0)^{-3}3\Gamma/4$ is much larger than Γ, in the same limit where state $|A\rangle$ becomes metastable. The latter effect is also important because it drastically alters the duration of the bright and dark periods. Our analysis is based on so-called frequency-resolved delay functions (an extension of a concept introduced by Reynaud, Dalibard, Cohen-Tannoudji, and others),[11-13] which is shown to be very convenient in analyzing quantities such as frequency-resolved photon statistics (FRPS).

The paper is organized as follows. In Sec. II, we assume an incoherent pumping of the $|E\rangle\leftrightarrow|G\rangle$ transition and analyze the statistics of the bright and dark periods of the fluorescence. The FRPS in a bright period is calculated in Sec. III utilizing the method developed in Sec. II. In Sec. IV, we analyze the statistics of bright and dark periods for the case of strong coherent pumping when $\mathbf{k}\cdot\mathbf{d}=0$. In Sec. V, we consider the application of the method developed in Sec. IV to two problems in FRPS in a dressed-atom picture: fluorescence photons produced by a single two-level atom and by the two-atom system considered in this paper. Finally, in Sec. VI, we consider a coupling of the antisymmetric state to the symmetric states by the laser field when $\mathbf{k}\cdot\mathbf{d}\neq 0$ and show how this alters the statistics of the bright and dark periods considered in Sec. IV.

II. INCOHERENT PUMP

We first consider the two-atom system irradiated by a strong incoherent pump which couples states $|E\rangle$ and $|G\rangle$ directly with rate R (see Fig. 1). It can be shown that this is a good model when a reasonably strong broadband laser whose center is tuned to the transition frequency of each atom is used [see the argument below Eq. (28) in Sec. IV]. When $\Gamma_S, R \gg \Gamma_A$, this system exhibits two phases in its fluorescence: a bright period (BP), in which repeated cycling through the channel $|E\rangle\to|S\rangle\to|G\rangle$ produces many fluorescence photons, and a dark period (DP), in which the system is shelved in the metastable state $|A\rangle$. A bright period begins (ends) when the system jumps from state $|A\rangle$ to state $|G\rangle$ (state $|E\rangle$ to state $|A\rangle$). Therefore, to determine the probability distributions $P_B(\tau)$ or $P_D(\tau)$ of the duration τ of a single BP or DP, respectively, we must find the time delay between *successive* transitions. The distribution $P_B(\tau)$ is determined by the delay between an $|A\rangle\to|G\rangle$ transition and the *next* $|E\rangle\to|A\rangle$ transition, while $P_D(\tau)$ is determined by the delay between an $|E\rangle\to|A\rangle$ transition and the *next* $|A\rangle\to|G\rangle$ transition. The calculation starts with rate equations

$$\frac{d}{dt}\Pi_i(t)=-\Pi_i(t)\sum_j \Gamma_{ij}+\sum_j \Pi_j(t)\Gamma_{ji} , \tag{1}$$

where $\Pi_i(t)$ is the population of state $|i\rangle$ at time t and Γ_{ij} is the transition rate from state $|i\rangle$ to state $|j\rangle$ $(i,j=E,S,G,A)$, which can be written in matrix form as

$$\Gamma_{ij}=\begin{bmatrix} 0 & \Gamma_S & R & \Gamma_A \\ 0 & 0 & \Gamma_S & 0 \\ R & 0 & 0 & 0 \\ 0 & 0 & \Gamma_A & 0 \end{bmatrix} . \tag{2}$$

The solution for $\Pi_i(t)$ includes contributions from any number of possible pathways leading to a final state population Π_i at time t. For example, suppose that the system decays from state $|A\rangle$ to state $|G\rangle$ at time $t=0$ (a BP begins at time $t=0$). The population $\Pi_A(t)$ obtained by solving Eqs. (1) and (2) with the initial condition $\Pi_G(0)=1$ includes contributions from pathways such as $|G\rangle \xrightarrow{t_1} |E\rangle \xrightarrow{t_2} |A\rangle$, $|G\rangle \xrightarrow{t_1} |E\rangle \xrightarrow{t_2} |A\rangle \xrightarrow{t_3} |G\rangle \xrightarrow{t_4} |E\rangle \xrightarrow{t_5} |A\rangle$, etc. $(0\le t_1<t_2\cdots\le t)$. In the $|G\rangle \xrightarrow{t_1} |E\rangle \xrightarrow{t_2} |A\rangle$ pathway, the first BP ends and the first DP begins at time t_2, with no further transition between times t_2 and t. In the $|G\rangle \xrightarrow{t_1} |E\rangle \xrightarrow{t_2} |A\rangle \xrightarrow{t_3} |G\rangle \xrightarrow{t_4} |E\rangle \xrightarrow{t_5} |A\rangle$ pathway, the second BP ends and the second DP begins at time $t=t_5$, with no further transition between times t_5 and t. Note that in both pathways, any number of transitions $|E\rangle\to|S\rangle\to|G\rangle\to|E\rangle$ is allowed before each transition to state $|A\rangle$. In order to calculate the duration of a single BP we need to separate out the contribution of the first decay to $\Pi_A(t)$. To do this, we pretend that the system will never escape from state $|A\rangle$ after the first $|E\rangle\to|A\rangle$ decay. This corresponds to setting $\Gamma_{AG}=0$ in Eq. (2), and solving Eq. (1) for $\Pi_A(t)$ with the initial condition $\Pi_G(0)=0$. We designate this population with a prime, $\Pi'_A(t)$, to distinguish from the true population of state $|A\rangle$. The crucial point in setting $\Gamma_{AG}=0$ in Eq. (2) is that it in no way influences the dynamics of the system for times before the first $|E\rangle\to|A\rangle$ decay. A quantity $W(E\to A/G;t)$ defined as the probability per unit time that, starting from state $|G\rangle$, the system decays to state $|A\rangle$ for the first time at t, can be found through

$$W(E\to A/G;t)=\frac{d}{dt}\Pi'_A(t) . \tag{3}$$

When $\Gamma_S, R\gg\Gamma_A$, a simple calculation using Eqs. (1), (3), and (2) with $\Gamma_{AG}=0$ yields

$$\begin{aligned} W(E\to A/G;t)=&\frac{\Gamma_A R(R-B)}{2B(R+\Gamma_S-B)}\exp[-(R+\Gamma_S-B)t] \\ &-\frac{\Gamma_A R(R+B)}{2B(R+\Gamma_S+B)} \\ &\times\exp[-(R+\Gamma_S+B)t] \\ &+\frac{R\Gamma_A}{3R+\Gamma_S}\exp[-R\Gamma_A t/(3R+\Gamma_S)] , \end{aligned} \tag{4}$$

where

$$B=(R^2-R\Gamma_S)^{1/2} .$$

From Eq. (4), it follows that $W(E\to A/G;t)\cong 0$ in the transient regime $t\ll R^{-1}$, reflecting the fact that it takes a time $\sim R^{-1}$ for the system to be pumped from state $|G\rangle$ to state $|E\rangle$, from which it can then decay to state $|A\rangle$. In the third term of Eq. (4), the factor $R/(3R+\Gamma_S)$ can be interpreted as the quasi-steady-state population (Π_E^{qs}) that state $|E\rangle$ would have if a decay to state $|A\rangle$ were forbidden. The appearance of this factor is related to the fact that fast transitions among states

$|E\rangle$, $|S\rangle$, and $|G\rangle$ drive their populations to the quasi-steady-state values long before any $|E\rangle \to |A\rangle$ decay occurs.

The probability distribution function $P_B(\tau)$ for the duration τ of a single BP is equal to the probability that, starting from state $|G\rangle$, the system will not decay to state $|A\rangle$ until time τ. Thus we can write $P_B(\tau)$ in terms of $W(E \to A/G;t)$ as

$$P_B(\tau) = 1 - \int_0^\tau W(E \to A/G;t)dt \ . \qquad (5)$$

By using Eqs. (4) and (5), one sees immediately that both the first and the second terms in Eq. (4) do not contribute in the limit $\Gamma_S, R \gg \Gamma_A$; in this limit, we find

$$P_B(\tau) = \exp(-\tau/\tau_B), \quad \tau_B = \frac{3R + \Gamma_S}{R\Gamma_A} \ , \qquad (6)$$

where τ_B is the average duration of a single BP. The inverse of the average duration of a single BP, τ_B^{-1}, is simply equal to the quasi-steady-state population of state $|E\rangle$ [given by $\Pi_E^{qs} = R/(3R + \Gamma_S)$] multiplied by the rate Γ_A for the $|E\rangle \to |A\rangle$ decay. As R/Γ_S increases, τ_B decreases, and eventually saturates at a value equal to $3\Gamma_A^{-1}$.

In order to find the analogous probability function $P_D(\tau)$, we apply a similar method [setting $\Gamma_{GE} = 0$ in Eq. (2) and solving Eq. (1) for $\Pi_G(t)$ with the initial condition $\Pi_A(0) = 1$] to find

$$W(A \to G/A;t) = \Gamma_A \exp(-\Gamma_A t) \qquad (7)$$

and

$$P_D(\tau) = 1 - \int_0^\tau W(A \to G/A;t)dt = \exp(-\tau/\tau_D) \ ,$$

$$\tau_D = \frac{1}{\Gamma_A} \ , \qquad (8)$$

where τ_D is the average duration of a single dark period. Note that the ratio $\tau_B/\tau_D = (3R + \Gamma_S)/R \Longrightarrow 3$ when $R \gg \Gamma_S$. This result is larger than that obtained in a single three-level atom (MQJ)[8-13] because of the additional active level $|S\rangle$ which is present in our problem.

In this section, we found $W(E \to A/G;t)$ and $W(A \to G/A;t)$ in order to calculate $P_B(\tau)$ or $P_D(\tau)$. The beginning or ending of a BP or DP corresponds to a distinctive decay $|E\rangle \to |A\rangle$ or $|A\rangle \to |G\rangle$, respectively. Depending on the W function to be evaluated, we alter Eq. (2) by setting some of the Γ's equal to zero. It is a generalization of a method developed by others.[11-13] As is shown in Sec. III, this approach is useful for calculating the frequency-resolved photon statistics.

In this problem and related problems, the quasi-steady-state populations in each period determine the probability distributions for the durations of a single BP and DP. Specifically, it has the form $P_i(\tau) = \exp(-\tau/\tau_i)$, where τ_i is the average duration of the period i ($i = B$ or D). As a simple application of this, we consider the three-level system depicted in Fig. 2 which has been studied extensively.[8-13] In Fig. 2, R_1 and R_2 represent incoherent pumps and Γ_1 and Γ_2 represent spontaneous decay rates. When $R_1, \Gamma_1 \gg R_2, \Gamma_2$, this three-level system, with state $|2\rangle$ metastable, exhibits BP and DP in its fluorescence: the BP involves transitions between states $|1\rangle$ and $|3\rangle$, and the DP is triggered by the rare excitation $|3\rangle \to |2\rangle$. When the system is in a DP, the next BP will start following by the transition $|2\rangle \to |3\rangle$. A simple calculation yields the quasi-steady-state population of state $|3\rangle$ in a BP as $\Pi_3^{qs} = (R_1 + \Gamma_1)/(2R_1 + \Gamma_1)$. Since the next DP is triggered by the transition $|3\rangle \to |2\rangle$ with a rate $\Gamma_{32} = R_2$, one finds

$$\tau_B = (\Pi_3^{qs}\Gamma_{32})^{-1} = [(R_1 + \Gamma_1)R_2/(2R_1 + \Gamma_1)]^{-1} \ ,$$

which is equal to $(R_2/2)^{-1}$ when $R_1 \gg \Gamma_1$. A similar argument produces that $\tau_D = (R_2 + \Gamma_2)^{-1}$, which reduces to R_2^{-1} when $R_2 \gg \Gamma_2$. These results agree with those that have been obtained previously.[10]

III. PHOTON STATISTICS IN A BRIGHT PERIOD

In this section, we study the FRPS in a BP by using the method developed in the previous section. We calculate the delay function $D(\omega_{ES}, \tau)$, the probability distribution describing the delay time τ between *successive* emissions of ω_{ES} photons (produced by the $|E\rangle \to |S\rangle$ decays) in a BP. As is seen in Fig. 1, these photons contribute to the peak in the spectrum centered at $\omega_0 - V$ and can be distinguished from ω_{SG} photons (produced by the $|S\rangle \to |G\rangle$ transitions). In this section, we ignore the existence of state $|A\rangle$ [all Γ_A's in Eq. (2) are set equal to zero] because of the time scale involved ($\Gamma_S^{-1}, R^{-1} \ll \Gamma_A^{-1}$).

We assume that an emission of a ω_{ES} photon takes place at time $t = 0$ leaving the system in state $|S\rangle$. In order to emit the next photon at time τ, the system decays from state $|S\rangle$ to state $|G\rangle$ at any instant t, is excited to state $|E\rangle$ at any instant after t, t', and then undergoes an $|E\rangle \to |S\rangle$ decay at time τ ($0 \le t \le t' \le \tau$). Therefore the delay function for the ω_{ES} photon should be written in

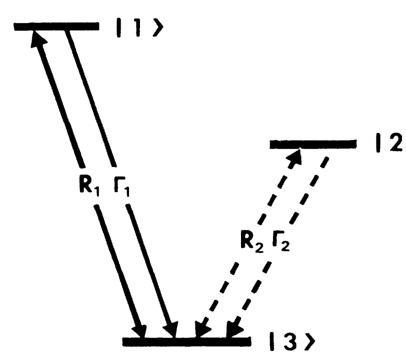

FIG. 2. Energy-level scheme for a single three-level atom producing macroscopic quantum jumps. The transition between states $|1\rangle$ and $|3\rangle$ is strongly driven at rate R_1, while that between states $|2\rangle$ and $|3\rangle$ is weakly driven at rate R_2. The decay rate from state $|1\rangle$ to state $|3\rangle$ is Γ_1 and the decay rate from state $|2\rangle$ to state $|3\rangle$ is Γ_2. These rates satisfy the condition $R_1, \Gamma_1 \gg R_2, \Gamma_2$. State $|2\rangle$ is the shelving state.

terms of two types of W functions [$W(S\to G/S;t)$ and $W(E\to S/G;t)$] as

$$D(\omega_{ES},\tau)=\int_0^\tau W(S\to G/S;t)W(E\to S/G;\tau-t)dt \; , \tag{9}$$

where the function $W(S\to G/S;t)$ is the probability per unit time that, starting from state $|S\rangle$, the system undergoes the *first* $|S\rangle\to|G\rangle$ transition at time t and the function $W(E\to S/G;t)$ is the probability per unit time that, starting from state $|G\rangle$, the system is pumped up to state $|E\rangle$ at any time $t'(0\le t'\le t)$ and makes the *first* $|E\rangle\to|S\rangle$ transition at time t. The way of finding these W functions from Eq. (1) and a modified form of Eq. (2) is the same as in Sec. I, and is not repeated here. A straightforward calculation yields

$$W(S\to G/S;t)=\Gamma_S\exp(-\Gamma_S t) \; ,$$

$$W(E\to S/G;t)=\frac{R\Gamma_S}{C}\exp\left[\left[-R-\frac{\Gamma_S}{2}+\frac{C}{2}\right]t\right] - \frac{R\Gamma_S}{C}\exp\left[\left[-R-\frac{\Gamma_S}{2}-\frac{C}{2}\right]t\right] \; , \tag{10}$$

where

$$C=(4R^2+\Gamma_S^2)^{1/2} \; .$$

Substitution of Eq. (10) into Eq. (9) yields

$$D(\omega_{ES},\tau)=\frac{\Gamma_S^2 R}{C}\left[\frac{\exp(-\Gamma_S\tau)-\exp[(-R-\Gamma_S/2-C/2)\tau]}{-R+\Gamma_S/2-C/2}\right]+(C\to -C) \; , \tag{11}$$

where $(C\to -C)$ represents a term substituting $-C$ for C. This function clearly manifests an antibunching effect[16] [$D(\omega_{ES},0)=0$]. (It is impossible to emit the second ω_{ES} photon right after the first emission of a ω_{ES} photon.) In the limit of strong pumping $R\gg\Gamma_S$ Eq. (11) reduces to a form

$$D(\omega_{ES},\tau)=\Gamma_S[\exp(-\Gamma_S\tau/2)-\exp(-\Gamma_S\tau)] \; , \tag{12}$$

which still exhibits antibunching. This can be expected from the cascade structure of the system.[17] In other words, the system still takes a time Γ_S^{-1} to decay out of state $|S\rangle$ even though the pumping R is so strong that the system can be rapidly pumped to state $|E\rangle$ from state $|G\rangle$.

A knowledge of the delay function $D(\omega_{ES},\tau)$ is enough to determine the complete photon statistics. The quantities $\overline{m}_T$ and Δm_T, defined as the average number and the dispersion of ω_{ES} photons emitted during a time period T, can be determined from[12]

$$\overline{m}_T=T/\overline{\tau}, \quad \Delta m_T^2=\overline{m}_T\Delta\tau^2/\overline{\tau}^2 \; , \tag{13}$$

where $\overline{\tau}$ and $\Delta\tau^2$ (the average and the dispersion of the delay time τ between successive photon emissions) can be calculated from Eq. (11) to be

$$\overline{\tau}=\frac{3R+\Gamma_S}{R\Gamma_S}, \quad \Delta\tau^2=\frac{5R^2+2R\Gamma_S+\Gamma_S^2}{R^2\Gamma_S^2} \; . \tag{14}$$

It follows from Eqs. (13) and (14) that

$$\overline{m}_T=T\frac{R\Gamma_S}{\Gamma_S+3R}, \quad \Delta m_T^2=\overline{m}_T\frac{5R^2+2R\Gamma_S+\Gamma_S^2}{(\Gamma_S+3R)^2} \; , \tag{15}$$

where the factor $R/(\Gamma_S+3R)$ can again be interpreted as the quasi-steady-state population of state $|E\rangle$. Here, we find that $\Delta m_T^2<\overline{m}_T$ and the photon statistics are sub-Poissonian. This reduced fluctuation (as compared to a coherent state where $\Delta m_T^2=\overline{m}_T$) can be explained by noting that $D(\omega_{ES},\tau)$ is a more sharply peaked function of τ than the corresponding D function for a coherent state [$D(\tau)=\Gamma\exp(-\Gamma\tau)$] so that the time delay between successive photon emissions is approximately constant. Thus we should detect roughly the same number of photons in each time interval T. Sub-Poissonian, Poissonian, or super-Poissonian statistics are usually characterized by Mandel's Q factor,[18] which is defined in terms of $\overline{m}_T$ and Δm_T^2 as

$$Q=\Delta m_T^2/\overline{m}_T-1 \; , \tag{16}$$

where

$$Q\begin{cases}<0, & \text{sub-Poissonian}\\ =0, & \text{Poissonian}\\ >0, & \text{super-Poissonian} \; .\end{cases} \tag{17}$$

In our problem, we find, by using Eqs. (15) and (16), that

$$Q=-\frac{4R(\Gamma_S+R)}{(\Gamma_S+3R)^2}\cong-\frac{4}{9} \quad (\text{when } R\gg\Gamma_S) \; . \tag{18}$$

IV. COHERENT PUMP

The incoherent pump field is now replaced by a strong coherent pump (laser) field. The laser field is assumed to be resonant with the transition frequency of each atom and is strong enough to saturate the two-photon $|G\rangle\leftrightarrow|E\rangle$ transition. We use a dressed-atom approach,[19] not only because it can account for quantum-mechanical coherence effects such as the ac Stark splitting, but also because it can permit us to interpret the results in a relatively simple manner. In this picture, we first neglect the coupling of the two-atom system to the vacuum which is responsible for spontaneous emission. After the laser field is quantized, the dynamics of the total system (two-atom plus the laser field) is governed by the time-independent Hamiltonian[20]

$$H=\hbar\omega_0(\sigma_1^+\sigma_1^-+\sigma_2^+\sigma_2^-+a^\dagger a)+\hbar V(\sigma_1^+\sigma_2^-+\sigma_2^+\sigma_1^-)$$
$$+i\hbar g(e^{-i\mathbf{k}\cdot\mathbf{d}/2}\sigma_1^+ + e^{i\mathbf{k}\cdot\mathbf{d}/2}\sigma_2^+)a+\text{H.c.} , \qquad (19)$$

where σ_i^+ (σ_i^-) is the atomic raising (lowering) operator of the ith atom ($i=1,2$) and $a^\dagger(a)$ is the creation (annihilation) operator of the laser photons. The first line in Eq. (19) represents the free energy of the atoms and the laser field. The second line represents the atom-atom interaction which is responsible for the energy shifts V shown in Fig. 1. The third line represents the atom-field dipole interaction, with coupling constant $g\exp(\mp i\mathbf{k}\cdot\mathbf{d}/2)$, where $\pm\mathbf{d}/2$ are the positions of the atoms and $\mathbf{k}$ is the wave vector of the laser field. The eigenstates for the Hamiltonian (19), called dressed states, are superpositions of products of atomic states and field states. In this section, we assume that $\mathbf{k}\cdot\mathbf{d}=0$, so that the atom-field coupling is identical for both atoms. When $\mathbf{k}\cdot\mathbf{d}=0$, the coupling in Eq. (19) involves only the symmetrical atomic states [i.e., only $i\hbar g(\sigma_1^+ +\sigma_2^+)a+\text{H.c.}$ appears]. In this case, the dressed states are found to be[7]

$$|1,n\rangle=\frac{\cos\theta}{\sqrt{2}}(|E,n-2\rangle-|G,n\rangle)-i\sin\theta|S,n-1\rangle ,$$
$$|2,n\rangle=\frac{1}{\sqrt{2}}(|E,n-2\rangle+|G,n\rangle) ,$$
$$|3,n\rangle=\frac{\sin\theta}{\sqrt{2}}(|E,n-2\rangle-|G,n\rangle)+i\cos\theta|S,n-1\rangle , \qquad (20)$$
$$|4,n\rangle=|A,n-1\rangle ,$$

with the corresponding eigenenergies

$$E_{1,n}=\hbar\{n\omega_0+\tfrac{1}{2}[(V^2+\Omega_R^2)^{1/2}+V]\} ,$$
$$E_{2,n}=\hbar(n\omega_0) ,$$
$$E_{3,n}=\hbar\{n\omega_0-\tfrac{1}{2}[(V^2+\Omega_R^2)^{1/2}-V]\} , \qquad (21)$$
$$E_{4,n}=\hbar(n\omega_0-V) ,$$

where n ($n=1,2,\dots$) is the occupation number for the laser field state and Ω_R is the usual Rabi frequency which can be written in terms of g and $\bar{n}$ (the average number of laser photons) as

$$\Omega_R=4g\bar{n}^{1/2} . \qquad (22)$$

Note that we exploited the quasiclassical character of the laser field by evaluating Ω_R at $\bar{n}$.[19] The factors $\cos\theta$ and $\sin\theta$ are functions of Ω_R and V

$$\cos\theta=\left[\frac{(V^2+\Omega_R^2)^{1/2}-V}{2(V^2+\Omega_R^2)^{1/2}}\right]^{1/2} ,$$
$$\sin\theta=\left[\frac{(V^2+\Omega_R^2)^{1/2}+V}{2(V^2+\Omega_R^2)^{1/2}}\right]^{1/2} . \qquad (23)$$

Energy levels of these states are shown in Fig. 3 and form an infinite ladder of nearly degenerate four-state multiplets. Adjacent multiplets are separated by the laser frequency ω_0.

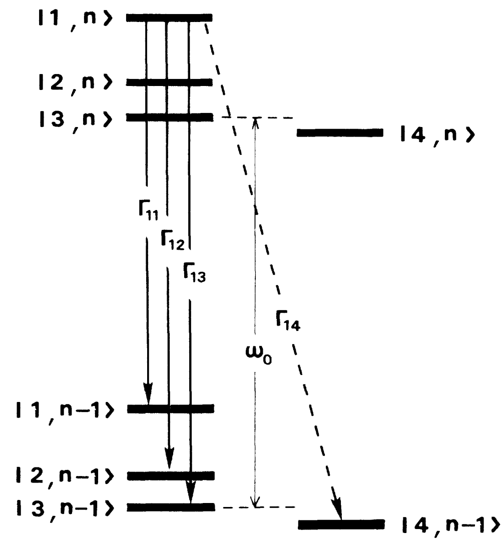

FIG. 3. Energy levels of the dressed states $|\alpha,n\rangle$ for the two-atom plus laser field system. Adjacent multiplets are separated by the laser frequency ω_0. Decay rates $\Gamma_{1\alpha}$ from state $|1,n\rangle$ to the lower lying states $|\alpha,n-1\rangle$ ($\alpha=1,2,3,4$) are indicated in the figure. When $d\ll\lambda_0$, the decay rate Γ_{14} to state $|4,n-1\rangle$ (represented by a dashed arrow) is much smaller than the decay rates to the other states (represented by solid arrows).

Coupling the states to the vacuum produces states having bandwidths on the order of Γ_S or Γ_A, and results in the system's cascade down the quantum ladder in which the decays between adjacent multiplets occur with rates Γ_S or Γ_A. Each of these decays corresponds to the creation of a fluorescence photon whose frequency is determined by the energy separation between the dressed states, within the uncertainty given by the bandwidths. It is assumed that the states in a given multiplet do not overlap (secular approximation),[19] i.e.,

$$(E_{\alpha,n}-E_{\beta,n})/\hbar\gg 2\Gamma(\cong\Gamma_S\gg\Gamma_A) \quad \text{for all } \alpha,\beta . \qquad (24)$$

Under this condition, general relaxation theory dictates that the cascade of the system can be described as a rate process among the dressed states

$$\frac{d}{dt}\Pi_{\alpha,n}(t)=-\Pi_{\alpha,n}(t)\sum_{\beta=1}^{4}\Gamma_{\alpha\beta}^{(n)}$$
$$+\sum_{\beta=1}^{4}\Pi_{\beta,n+1}(t)\Gamma_{\beta\alpha}^{(n+1)} , \qquad (25)$$

where $\Pi_{\alpha,n}(t)$ is the time-dependent population of the state $|\alpha,n\rangle$ and $\Gamma_{\alpha\beta}^{(n)}$ is the decay rate for the $|\alpha,n\rangle\to|\beta,n-1\rangle$ ($\alpha,\beta=1,2,3,4$) transition calculated from[20]

$$\Gamma_{\alpha\beta}^{(n)}=\frac{\Gamma_S}{2}|\langle\alpha,n|(\sigma_1^+ +\sigma_2^+)|\beta,n-1\rangle|_{n=\bar{n}}^2$$
$$+\frac{\Gamma_A}{2}|\langle\alpha,n|(\sigma_1^+ -\sigma_2^+)|\beta,n-1\rangle|_{n=\bar{n}}^2 . \qquad (26)$$

Using Eqs. (20) and (26), we find $\Gamma_{\alpha\beta}^{(n)}$ in matrix form as

$$\Gamma_{\alpha\beta}^{(n)}=\begin{bmatrix} \frac{1-D^2}{2}\Gamma_S & \frac{1-D}{4}\Gamma_S & \frac{D^2}{2}\Gamma_S & \frac{1+D}{4}\Gamma_A \\ \frac{1-D}{4}\Gamma_S & 0 & \frac{1+D}{4}\Gamma_S & \frac{1}{2}\Gamma_A \\ \frac{D^2}{2}\Gamma_S & \frac{1+D}{4}\Gamma_S & \frac{1-D^2}{2}\Gamma_S & \frac{1-D}{4}\Gamma_A \\ \frac{1+D}{4}\Gamma_A & \frac{1}{2}\Gamma_A & \frac{1-D}{4}\Gamma_A & 0 \end{bmatrix}, \quad (27)$$

where

$$D=\cos^2\theta-\sin^2\theta=-V/(V^2-\Omega_R^2)^{1/2},$$

and $\cos\theta$ and $\sin\theta$ are given in Eq. (23). The rows and columns in matrix (27) are labeled according to Eq. (26). Again, we evaluated $\Gamma_{\alpha\beta}^{(n)}$ at $n=\bar{n}$, so that $\Gamma_{\alpha\beta}^{(n)}$ is the same for any pair of adjacent multiplets. The first term in Eq. (26) represents decays among the atomic symmetrical states and constitutes the 3×3 subblock in matrix (27), while the second term represents decays between the atomic symmetrical and antisymmetrical states and constitutes the 1×3 and 3×1 subblocks in matrix (27).

As is seen from Eq. (27), there are 14 possible transitions corresponding to the creation of photons with at most 13 different frequencies (the $|1,n\rangle\to|1,n-1\rangle$ and $|3,n\rangle\to|3,n-1\rangle$ transitions create photons which have the same frequency ω_0). However, when the two atoms are very close ($d\ll\lambda_0$), six of the decays (those involving decays to or from states $|4,n\rangle$) are relatively improbable. Consequently, one expects bright and dark periods in the fluorescence owing to the metastable states $|4,n\rangle$. A BP corresponds to a fast cascade of the system among the short-lived states $|\alpha,n\rangle$ ($\alpha=1,2,3$). When a transition $|\alpha,n\rangle\to|4,n-1\rangle$ ($\alpha=1$, 2, or 3) occurs, this fast cascade is interrupted and the BP ends; once a $|4,n\rangle\to|\alpha,n-1\rangle$($\alpha=1$, 2, or 3) decay occurs, the next BP starts.

The steady-state and quasi-steady-state populations [obtained by setting $\Gamma_A=0$ in Eq. (27)] of the dressed states are found from Eq. (25) to be

$$\Pi_\alpha^{\text{ss}}=\left[\sum_{n=0}^{\infty}\Pi_{\alpha,n}\right]^{\text{ss}}=\tfrac{1}{4}\quad(\alpha=1,2,3,4),$$

$$\Pi_\alpha^{\text{qs}}=\left[\sum_{n=0}^{\infty}\Pi_{\alpha,n}\right]^{\text{qs}}=\tfrac{1}{3}\quad(\alpha=1,2,3)\text{ in a BP}. \quad (28)$$

In this limit, we also see from Eq. (20) that in a BP the quasi-steady-state populations of the bare atom states are equal $[\Pi_i^{\text{qs}}=(\sum_{n=0}^{\infty}\Pi_{i,n})^{\text{qs}}=\frac{1}{3}$ $(i=E,S,G,)]$. The only approximation made leading to Eq. (28) is the secular approximation (24), which can be written in terms of Ω_R and V using Eq. (21) as $\sqrt{V\Gamma}\ll\Omega_R$ when $V\gg\Omega_R$. Therefore $\sqrt{V\Gamma}\ll\Omega_R\ll V$ is the condition under which the pumping scheme considered in Sec. II is valid. Note that when $\Omega_R\ll V$, the $|S\rangle\leftrightarrow|G\rangle$ transition is not appreciably driven by the laser field.

We now study the duration of a single BP and DP. Suppose that at time $t=0$ the system decays from state $|4,n+1\rangle$ to state $|\alpha,n\rangle$ and the state $|\alpha,n\rangle$ is populated with a probability p_α ($\alpha=1,2,3$) (the beginning of a BP). The probability p_α is the branching ratio

$$p_\alpha=\frac{\Gamma_{4\alpha}}{\sum_{\alpha'=1}^{3}\Gamma_{4\alpha'}}, \quad (29)$$

where the Γ's are given in Eq. (27). In order to obtain the probability describing the duration τ of a single BP, $P_B(\tau)$, we need to know when the *next* $|\beta,n'\rangle\to|4,n'-1\rangle$ ($\beta=1$, 2, or 3; $n'\le n$) transition occurs. Analogous to Eq. (5), we can write $P_B(\tau)$ as

$$\begin{aligned}P_B(\tau)&=1-\int_0^\tau dt\left[\sum_{\alpha,\beta=1}^{3}p_\alpha W(\beta\to4/\alpha;t)\right]\\&=1-\int_0^\tau dt\left[\sum_{\alpha,\beta=1}^{3}p_\alpha\sum_{f=0}^{\infty}W^{(f)}(\beta\to4/\alpha;t)\right],\end{aligned} \quad (30)$$

where the function $W^{(f)}(\beta\to4/\alpha;t)$ is the probability per unit time that, starting from state $|\alpha,n\rangle$, the system makes f successive decays *excluding* those of type $|\mu,n'\rangle\to|4,n'-1\rangle$ ($\mu=1,2,3$; $n\ge n'\ge n-f+1$) between times 0 and t, and then decays from state $|\beta,n-f\rangle$ to state $|4,n-f-1\rangle$ ($\beta=1$, 2, or 3) for the first time at t (see Fig. 4). It is tempting to try to calculate the function $W^{(f)}(\beta\to4/\alpha;t)$ by setting $\Gamma_{\mu4}^{(n')}$

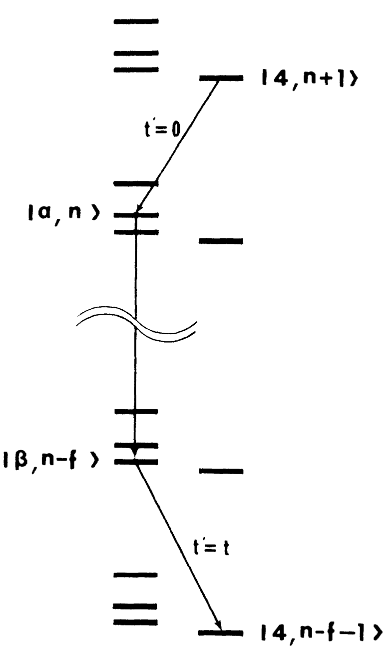

FIG. 4. The diagram shows a particular cascade which is characterized by the function $W^{(f)}(\beta\to4/\alpha;t)$, defined as a probability per unit time that, starting from state $|\alpha,n\rangle$, the system makes f successive decays *excluding* those of type $|\mu,n'\rangle\to|4,n'-1\rangle$ ($\mu=1,2,3$; $n\ge n'\ge n-f+1$) between times 0 and t, then decays from state $|\beta,n-f\rangle$ to state $|4,n-f-1\rangle$ ($\beta=1$, 2, or 3). The initial and final transitions in the cascade correspond to those out of and into metastable states.

($\mu=1,2,3$; $n\geq n'\geq n-f+1$) equal to zero in Eqs. (25) and (27). Tempting as it may be, this procedure leads to incorrect results since it modifies the dynamics of the system at times *before* the transition to the shelving level of interest. For this purpose, we introduce a more elementary branching function $w_{\mu\nu}(t)$, which is defined as the probability per unit time that, starting from state $|\mu,n'\rangle$, the system decays to state $|\nu,n'-1\rangle$ ($\mu,\nu=1,2,3,4$) at a time t. Since the branching function above involves only transitions between adjacent multiplets, it can be calculated by the same method used in Sec. II. Terms in Eqs. (25) and (27) corresponding to decays to states in the ($n'-2$) multiplet are set equal to zero. Note that we still have a right to set these Γ's equal to zero because this does not alter the dynamics of the system *until* the system decays to states $|\nu,n'-1\rangle$. The branching function is found to be

$$w_{\mu\nu}(t)\left[=\frac{d}{dt}\Pi'_{\nu,n'-1}(t)\right]=\Gamma_{\mu\nu}\exp\left[-\sum_{\nu'=1}^{4}\Gamma_{\mu\nu'}t\right] . \quad (31)$$

In Eq. (31), the prime of the population denotes the modified population of state $|\nu,n-1\rangle$ as before. We now write $W^{(f)}(\beta\rightarrow 4/\alpha;t)$ using Eq. (31) by considering all possible paths during f successive decays as

$$W^{(f)}(\beta\rightarrow 4/\alpha;t)=\sum_{(\epsilon,\zeta,\ldots,\xi)=1}^{4}\int_0^t dt_f\int_0^{t_f}dt_{f-1}\cdots\int_0^{t_2}dt_1[w_{\alpha\epsilon}(t_1)w_{\epsilon\zeta}(t_2-t_1)\cdots w_{\xi\beta}(t_f-t_{f-1})w_{\beta 4}(t-t_f)] , \quad (32)$$

where the summation $\sum$ excludes paths involving transitions $|\mu,n'\rangle\rightarrow|4,n'-1\rangle$ ($\mu=1,2,3$; $n\geq n'\geq n-f+1$). In order to proceed further, we first take the Laplace transform of Eq. (32). This yields a simple product of f functions of the form $w_{\mu\nu}(s)=\Gamma_{\mu\nu}/(s+\sum_{\nu'=1}^{4}\Gamma_{\mu\nu'})$; each being the Laplace transform of Eq. (31). Now, the summation over all possible intermediate states can be done easily by treating the $w_{\mu\nu}(s)$ function as the component of a matrix $\underline{W}(s)$. We see from Eqs. (27) and (31) that these matrices are identical so that the final result is the matrix $\underline{W}(s)$ raised to the fth power. However, the matrix must be modified to account for the fact that some of the paths are excluded from the summation. Thus we introduce a modified matrix, $\underline{W}_B(s)$ as

$$[\underline{W}_B(s)]_{\mu\nu}=\begin{cases}w_{\mu\nu}(s), & \text{when } (\mu,\nu)\neq(\mu',4),\ (\mu'=1,2,3)\\ 0, & \text{when } (\mu,\nu)=(\mu',4),\ (\mu'=1,2,3) .\end{cases} \quad (33)$$

Note that we did not set $\Gamma_{\mu'4}$ ($\mu'=1,2,3$) equal to zero, which would alter matrix elements of $\underline{W}(s)$ corresponding to allowed transitions. Thus we write Eq. (32) as

$$W^{(f)}(\beta\rightarrow 4/\alpha;t)=\mathcal{L}_t^{-1}\{([\underline{W}_B(s)]^f)_{\alpha\beta}w_{\beta 4}(s)\} , \quad (34)$$

where the symbol $\mathcal{L}^{-1}$ represents the inverse Laplace transform. Then, substitution of Eqs. (29) and (34) into Eq. (30) yields

$$P_B(\tau)=1-\sum_{\alpha,\beta=1}^{3}\frac{\Gamma_{4\alpha}}{\sum_{\alpha'=1}^{3}\Gamma_{4\alpha'}}\int_0^{\tau}dt\,\mathcal{L}_t^{-1}\left[\sum_{f=0}^{\infty}([\underline{W}_B(s)]^f)_{\alpha\beta}w_{\beta 4}(s)\right] . \quad (35)$$

This formula is exact. However, in the limit $\Gamma_A\ll\Gamma_S$, this reduces to

$$P_B(\tau)=\exp\left[-\left[\sum_{\beta=1}^{3}\tfrac{1}{3}\Gamma_{\beta 4}\right]\tau\right]=\exp(-\tau/\tau_B) ,$$

$$\tau_B=\left[\sum_{\beta=1}^{3}\tfrac{1}{3}\Gamma_{\beta 4}\right]^{-1}=3\Gamma_A^{-1} . \quad (36)$$

The factor of $\frac{1}{3}$ in the exponential can be interpreted as the quasi-steady-state populations (28). The reason for the appearance of this factor is the same as before [see the argument below Eq. (4) in Sec. II].

A similar calculation yields

$$P_D(\tau)=1-\sum_{\beta=1}^{3}\int_0^{\tau}dt\,\mathcal{L}_t^{-1}\left[\sum_{f=0}^{\infty}([\underline{W}_D(s)]^f)_{44}w_{4\beta}(s)\right]$$

$$=\exp(-\tau/\tau_D) .$$

$$\tau_D=\left[\sum_{\beta=1}^{3}\Gamma_{4\beta}\right]^{-1}=\Gamma_A^{-1} . \quad (37)$$

This time, matrix $\underline{W}_D(s)$ is defined as

$$[\underline{W}_D(s)]_{\mu\nu}=\begin{cases}w_{\mu\nu}(s),\\ \text{when } (\mu,\nu)\neq(4,\nu')\ (\nu'=1,2,3)\\ 0,\\ \text{when } (\mu,\nu)=(4,\nu')\ (\nu'=1,2,3) .\end{cases} \quad (38)$$

We find that $P_B(\tau)$ and $P_D(\tau)$ are completely independent of the Rabi frequency Ω_R and the energy shift V.

This results in part from the secular approximation (24), and, in part, owing to the fact that state $|4,n\rangle$ is completely decoupled from the laser due to our choice of geometry ($\mathbf{k}\cdot\mathbf{d}=0$). Coupling of state $|A\rangle$ to the symmetrical states occurs via spontaneous decay only and does not involve the Rabi frequency Ω_R.

V. PHOTON STATISTICS IN A DRESSED-ATOM PICTURE

We have formulated the probability distributions of durations of a single BP and DP based on an elementary branching function $w_{\mu\nu}(t)$. However, the real power of this method will become apparent when frequency-resolved photon statistics are considered in a dressed-atom picture.

In this section, we consider a delay function $D(\omega_{\alpha\beta},\tau)$ defined as the probability distribution for the time delay τ between successive emissions of $\omega_{\alpha\beta}$ photons. For simplicity, let us suppose that the transition $|\alpha,n+1\rangle\rightarrow|\beta,n\rangle$ for fixed pairs of (α,β) and any n creates a photon with a definite frequency $\omega_{\alpha\beta}$. This assumption corresponds to the secular approximation, such as Eq. (24), and the quasiclassical character of the laser field. The first emission of a $\omega_{\alpha\beta}$ photon leaves the system in state $|\beta,n\rangle$ (n can be any integer) at time $t=0$. Therefore the function $D(\omega_{\alpha\beta},\tau)$ can be interpreted as a probability per unit time that from this initial state $|\beta,n\rangle$, the system makes any number (f) of successive decays while creating many types of photons *exclusive* of $\omega_{\alpha\beta}$ photons, and eventually reaches a state $|\alpha,n-f\rangle$, from which the system makes the final decay $|\alpha,n-f\rangle\rightarrow|\beta,n-f-1\rangle$ at time τ. Analogous to Eq. (34), we thus write $D(\omega_{\alpha\beta},\tau)$ as

$$D(\omega_{\alpha\beta},\tau)=\mathcal{L}_\tau^{-1}\left[\sum_{f=0}^{\infty}([\underline{W}_{(\alpha\beta)}(s)]^f)_{\beta\alpha}w_{\alpha\beta}(s)\right], \tag{39}$$

where matrix $\underline{W}_{(\alpha\beta)}(s)$ is defined as

$$[\underline{W}_{(\alpha\beta)}(s)]_{\mu\nu}=\begin{cases}w_{\mu\nu}(s), & \text{when } (\mu,\nu)\neq(\alpha,\beta)\\ 0, & \text{when } (\mu,\nu)=(\alpha,\beta)\,.\end{cases} \tag{40}$$

As a first example of an application of Eqs. (39) and (40), we consider the FRPS of the fluorescence photons produced by a single two-level atom irradiated by a strong coherent laser with Rabi frequency Ω_R and detuning δ.[21,22] The frequency spectrum of the fluorescence photons is the well-known Mollow triplet.[6] These photons are produced when the system cascades down the ladder consisting of states $|\alpha,n\rangle$, executing the four types of transitions $|\alpha,n\rangle\rightarrow|\beta,n-1\rangle$ ($\alpha,\beta=1,2$; $n=1,2,\ldots$). The decay rates $\Gamma_{\alpha\beta}$ between the adjacent multiplets were calculated to be[23]

$$\Gamma_{\alpha\beta}=\frac{\Gamma}{4(\Omega_R^2+\delta^2)}\begin{bmatrix}\Omega_R^2 & [(\Omega_R^2+\delta^2)^{1/2}-\delta]^2\\ [(\Omega_R^2+\delta^2)^{1/2}+\delta]^2 & \Omega_R^2\end{bmatrix}. \tag{41}$$

Let us calculate the delay function of the ω_{21} photons. The $\underline{W}_{(\alpha\beta)}(s)$ matrix in Eq. (40) takes the form

$$\underline{W}_{(21)}(s)=\begin{bmatrix}\dfrac{\Gamma_{11}}{s+\Gamma_{11}+\Gamma_{12}} & \dfrac{\Gamma_{12}}{s+\Gamma_{11}+\Gamma_{12}}\\ 0 & \dfrac{\Gamma_{22}}{s+\Gamma_{21}+\Gamma_{22}}\end{bmatrix}. \tag{42}$$

Substitution of Eqs. (42) and $w_{21}(s)=\Gamma_{21}/(s+\Gamma_{21}+\Gamma_{22})$ into Eq. (39) with the use of the formula

$$\begin{bmatrix}a & b\\ 0 & c\end{bmatrix}^f=\begin{bmatrix}a^f & b\dfrac{a^f-c^f}{a-c}\\ 0 & c^f\end{bmatrix}$$

yields

$$D(\omega_{21},\tau)=\frac{\Gamma_{12}\Gamma_{21}}{\Gamma_{12}-\Gamma_{21}}[\exp(-\Gamma_{21}\tau)-\exp(-\Gamma_{12}\tau)]\,. \tag{43}$$

This function clearly manifests the antibunching effect $D(\omega_{21},0)=0$, simply explained by the fact that the emission of a ω_{21} photon at time $t=0$ projects the wave function of the system to one of the dressed states $|1,n\rangle$ so that the system is unable to emit a photon created by the same type of transition $|2,n\rangle\rightarrow|1,n-1\rangle$ without any time delay. From Eq. (43), we find the average and the dispersion of the delay time τ to be

$$\bar{\tau}=\frac{\Gamma_{21}+\Gamma_{12}}{\Gamma_{21}\Gamma_{12}},\quad \Delta\tau^2=\frac{\Gamma_{21}^2+\Gamma_{12}^2}{\Gamma_{21}^2\Gamma_{12}^2}\,. \tag{44}$$

Substitution of Eq. (44) into Eq. (13) yields

$$\begin{aligned}\bar{m}_T&=T\frac{\Gamma_{12}\Gamma_{21}}{\Gamma_{12}+\Gamma_{21}}\\ &=\frac{\Gamma T}{8}\frac{\Omega_R^4}{(\Omega_R^2+\delta^2)(\Omega_R^2+2\delta^2)}\,,\end{aligned} \tag{45}$$

$$\begin{aligned}\Delta m_T&=\left[\bar{m}_T\frac{\Gamma_{12}^2+\Gamma_{21}^2}{(\Gamma_{12}+\Gamma_{21})^2}\right]^{1/2}\\ &=\left[\bar{m}_T\frac{2(\Omega_R^2+2\delta^2)^2-\Omega_R^4}{2(\Omega_R^2+2\delta^2)^2}\right]^{1/2},\end{aligned} \tag{46}$$

where we used Eq. (41) to write Γ's in terms of Ω_R and δ. As can be seen from Eq. (46), the photon statistics are sub-Poissonian. Mandel's Q factor can be calculated from Eqs. (16), (45) and (46) to be

$$Q=-\frac{\Omega_R^4}{2(\Omega_R^2+2\delta^2)^2}\geq-\frac{1}{2}\ , \tag{47}$$

which is in complete agreement with previously obtained result.[22]

As a second example, we consider the FRPS in a BP of the fluorescence of two-atom system considered in Sec. IV, but now in a dressed-atom picture. This is more complicated owing to the level structure of the dressed states. For simplicity, we completely ignore the existence of state $|4,n\rangle$. In order to study the FRPS, we again need to know the delay function $D(\omega_{\alpha\beta},\tau)$ for a fixed pair of (α,β) $(\alpha,\beta=1,2,3)$. We must evaluate the fth power of a 3×3 $\underline{W}_{(\alpha\beta)}$ matrix, whose analytical expression is sometimes impossible to find. However, we can still find an approximate expression for $D(\omega_{\alpha\beta},\tau)$ by truncating the sum in Eq. (39), corresponding to the *minimum* delay time or *minimum* number of successive decays between the dressed states required for the emission of the *next* $\omega_{\alpha\beta}$ photon. As an example, let us consider ω_{12} photons. Since the first nonvanishing $([\underline{W}_{(12)}(s)]^f)_{21}$ occurs when $f=1$ in Eq. (39), we predict that

$$\begin{aligned}D(\omega_{12},\tau)&\cong\mathcal{L}_\tau^{-1}[w_{21}(s)w_{12}(s)]\\&=\left[\frac{(3-D)\Gamma_S}{4}\right]^2\tau\exp\left[-\frac{(3-D)\Gamma_S}{4}\tau\right]\ ,\end{aligned} \tag{48}$$

where $D(\omega_{12},\tau)$ is renormalized as $\int_0^\infty D(\omega_{12},\tau)d\tau=1$. Using Eqs. (13) and (48), we find that $\overline{m}_T=T\Gamma_S(3-D)/8$ and $\Delta m=(\overline{m}_T/2)^{1/2}$ (sub-Poissonian).

VI. COHERENT PUMPING WITH ARBITRARY GEOMETRY ($\mathbf{k}\cdot\mathbf{d}\neq 0$)

In Sec. IV, we considered the special case when $\mathbf{k}\cdot\mathbf{d}=0$. We found that only the transitions $|E\rangle\leftrightarrow|S\rangle$ and $|S\rangle\leftrightarrow|G\rangle$ involving the symmetrical atomic states were excited by the laser field. This result is also implied by Eq. (20) where the state $|4,n\rangle$ is the direct product $|A\rangle|n-1\rangle$. However, when $\mathbf{k}\cdot\mathbf{d}\neq 0$, the situation is modified because the interatomic separation d now results in an atom–laser-field interaction Hamiltonian that varies as $i\hbar g[\cos(\mathbf{k}\cdot\mathbf{d}/2)](\sigma_1^+ +\sigma_2^+)a+\hbar g[\sin(\mathbf{k}\cdot\mathbf{d}/2)](\sigma_1^+ -\sigma_2^+)a+\text{H.c.}$ It is seen that the coupling now contains antisymmetric as well as symmetric components. When $\mathbf{k}\cdot\mathbf{d}\ll 1$, we find the approximate dressed states to be

$$\begin{aligned}|1,n\rangle'&=|1,n\rangle-\frac{\Omega_R\Lambda\mathbf{k}\cdot\mathbf{d}}{2(\Lambda+3V)}|4,n\rangle\ ,\\|2,n\rangle'&=|2,n\rangle\ ,\\|3,n\rangle'&=|3,n\rangle+\frac{\Omega_R\Lambda\mathbf{k}\cdot\mathbf{d}}{2(\Lambda-3V)}|4,n\rangle\ ,\\|4,n\rangle'&=|4,n\rangle+\frac{\Omega_R\Lambda\mathbf{k}\cdot\mathbf{d}}{2(\Lambda+3V)}|1,n\rangle\\&\quad-\frac{\Omega_R\Lambda\mathbf{k}\cdot\mathbf{d}}{2(\Lambda-3V)}|3,n\rangle\ ,\end{aligned} \tag{49}$$

where

$$\Lambda=(V^2+\Omega_R^2)^{1/2}\ .$$

The shift of eigenenergies $\Delta E_{\alpha,n}$ $(\alpha=1,2,3,4)$ from the energies given in Eq. (21) are expected to be small and on the order of $\Omega_R(\mathbf{k}\cdot\mathbf{d})^2$.

Modified decay rates between adjacent multiplets can be found from Eqs. (26) and (49). It is easy to see that the decay rates $\Gamma'_{\alpha\beta}$ $(\alpha,\beta=1,2,3)$ involving the short-lived states $|\alpha,n\rangle'$ $(\alpha=1,2,3)$ are not changed significantly from the values given in the 3×3 subblock in matrix (27), leading to the same quasi-steady-state populations (28). The rates $\Gamma'_{\alpha 4}$ or $\Gamma'_{4\alpha}$ involving decays to or from the metastable states $|4,n\rangle'$, are different from the values given in the 1×3 and 3×1 subblocks in matrix (27). In particular, we find the modified total decay rate Γ'_4 to or from state $|4,n\rangle$ to be

$$\begin{aligned}\Gamma'_4&\equiv\sum_{\beta=1}^{3}\Gamma'_{4\beta}=\sum_{\beta=1}^{3}\Gamma'_{\beta 4}\\&=\Gamma_A+\frac{(\mathbf{k}\cdot\mathbf{d})^2}{4}\frac{\Omega_R^2(\Omega_R^2+8V^2)}{(\Omega_R^2-8V^2)^2}\Gamma_S\\&\cong\Gamma_A\left[1+\frac{5(\hat{\mathbf{k}}\cdot\hat{\mathbf{d}})^2}{2}\frac{\Omega_R^2(\Omega_R^2+8V^2)}{(\Omega_R^2-8V^2)^2}\right]\ ,\end{aligned} \tag{50}$$

where approximate values for Γ_A and Γ_S appropriate to the limit $d\ll\lambda_0$ were used (see Sec. I). Analogous to Eqs. (36) and (37), we find

$$\begin{aligned}\tau'_B&=\left[\sum_{\beta=1}^{3}\frac{\Gamma'_{\beta 4}}{3}\right]^{-1}=3(\Gamma'_A)^{-1}\ ,\\\tau'_D&=\left[\sum_{\beta=1}^{3}\Gamma'_{4\beta}\right]^{-1}=(\Gamma'_A)^{-1}\ .\end{aligned} \tag{51}$$

The reason for these shortened periods (τ'_D,τ'_B) is understood as follows: the laser excites not only the transitions $|E\rangle\leftrightarrow|S\rangle$ and $|S\rangle\leftrightarrow|G\rangle$ but also the transitions $|E\rangle\leftrightarrow|A\rangle$ and $|A\rangle\leftrightarrow|G\rangle$. Consequently, transitions to or from the shelving state $|A\rangle$ can be caused by stimulated emission and absorption (of the laser photons) to the short-lived states $|E\rangle$, $|S\rangle$, or $|G\rangle$ as well as by spontaneous emission.

VII. CONCLUSION

We have shown that it is possible to observe MQJ in the fluorescence through a cooperative atomic interaction. The jumps or discontinuous changes of the fluorescent intensity occurs on a time scale $\Gamma_A^{-1}\sim\Gamma^{-1}(\lambda_0/d)^2$. A restriction is imposed on the interatomic separation needed to observe MQJ by the finite response time of the detector. In order to make Γ_A^{-1} larger than the response time of the detector, d must be $\sim\lambda_0/100$. Current technology has yet to surmount this difficulty. Ion traps cannot be used because the Coulomb repulsion is too strong. Using neutral atom traps or confining atoms in a solid host may be the best hope of observing the two-atom MQJ described in this paper.

The formalism developed in this paper can be applied to any four-level or more complicated scheme in which one or more states is metastable. Additionally, our formalism can be used to study the FRPS of any multilevel atom in a simple way.

Note added in proof. After submitting this article, we learned of a recent calculation of the dynamics of a two-atom composite system interacting with a weak incident field.[24] In that work, the time evolution of the symmetric states was studied, including the level shift of the intermediate symmetric state, but neglecting the transitions between symmetric and the antisymmetric states.

ACKNOWLEDGMENTS

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