Spectral correlations in resonance fluorescence

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We study the photon correlations between the output of two spectral filters set within the fluorescence triplet of a two-state atom. The time uncertainty arising from the spectral resolution of the filters implies a possible interference between opposite orders of emission, contributing to the same detection order. Furthermore, the fluorescent emission is a quantum-mechanical process, and successive emissions in different components do not commute. The correlation functions are affected both by the memory time of the filters and by the noncommutativity of successive emissions. When the two filter bandwidths are larger than the widths of the components, this only modifies the short-time behavior of the correlation functions between two photons from different spectral components. For narrow filters, the entire correlation function is dominated by memory-time effects. Positive (bunching) correlations arise when the two filters are set at the same frequency, and also when they are positioned symmetrically at opposite sides of the driving frequency.

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I. INTRODUCTION

The measurement of spectral correlations between frequencies ω_1 and ω_2 of a light source requires the use of narrow spectral filters set at these frequencies. This filtering is only effective when the passband width λ of the filters is small compared with the spectral width of the source. When light is passed through such filters, a delay time of the order λ^{-1} arises between input and output. Fluctuations in the output light therefore reflect the input fluctuations in the recent past. In fact, the fluctuations can be qualitatively modified by the filter. For instance, phase noise can be converted into intensity noise [1]. In a recent experiment it has been demonstrated that the coherent phase-fluctuating light from a semiconductor laser is converted into light with chaotic intensity fluctuations by a narrow filter [2].

The time delay inherent in the spectral filtering can also have significant effects when the filter width λ is larger than the width of a component in the spectrum. An example is the time correlation between two successive photon detections from the components of the resonance fluorescence triplet of an atom. For the spectral separation of the components of the triplet it is sufficient to use filters with a bandwidth λ that is smaller than the line splittings, but larger than the linewidths. This means that for a certain instant of photon detection, the instant of emission is indeterminate with an uncertainty λ^{-1}. When two photons from different spectral components are detected with a time delay that is smaller than λ^{-1}, the order of emission may be different from the detection order. Since the two corresponding emission operators do not commute in general, this affects the correlation function for two photons from different spectral components for small time delays. It has recently been demonstrated that in the case of a photon from the central Rayleigh line and a photon from one of the sidebands, the commutators produce destructive interference, which leads to complete antibunching [3]. For two photons from opposite sidebands, the correlation function also displays a dip at zero time delay, which exactly counterbalances the photon bunching on the longer time scale of the inverse linewidths [4].

In the present paper we give a general description of spectral correlations within the fluorescence triplet. When the filters are narrower than the linewidths, the quantum effects of noncommuting emission operators combine with the effective integration over past emission times. We evaluate the correlation functions for all possible pairs of spectral components.

II. FLUORESCENCE TRIPLET

In this section, we summarize the properties of the fluorescence triplet of a two-state atom in a form that allows us to introduce the effect of spectral filters in a simple way. We consider an atom in a classical monochromatic radiation field at frequency ω. The field drives the transition between the ground state $|g\rangle$ and an excited state $|e\rangle$. Both states are supposed nondegenerate. In the rotating frame the evolution of the atomic density matrix is described by the equation

$$\frac{d\sigma}{dt} = -iL\sigma \ . \tag{2.1}$$

The Liouville operator L indicates the sum of the commutator with an effective Hamiltonian, and an operator Γ describing spontaneous decay, so that [5]

$$-iL\sigma = -i[H,\sigma]/\hbar - \Gamma\sigma \ , \tag{2.2}$$

with

$$H = -\hbar(\Delta S_z + \Omega S_x) \ . \tag{2.3}$$

The Rabi frequency Ω measures the strength of the atom-field coupling, and $\Delta = \omega - \omega_0$ is the detuning of the

light frequency from resonance. The operator Γ is defined by [6]

$$\Gamma\sigma=\tfrac{1}{2}A(S^{+}S^{-}\sigma+\sigma S^{+}S^{-}-2S^{-}\sigma S^{+}), \tag{2.4}$$

with A the spontaneous decay rate. In (2.3) and (2.4) we have used the Pauli matrices

$$\begin{aligned} S_x&=\tfrac{1}{2}(|e\rangle\langle g|+|g\rangle\langle e|),\\ S_y&=-\frac{i}{2}(|e\rangle\langle g|-|g\rangle\langle e|),\\ S_z&=\tfrac{1}{2}(|e\rangle\langle e|-|g\rangle\langle g|), \end{aligned} \tag{2.5}$$

and the atomic raising and lowering operators

$$S^{\pm}=S_x\pm iS_y . \tag{2.6}$$

The evolution equation (2.1) is equivalent to the optical Bloch equations [5].

The fluorescence radiation emitted by the atom is described by the Heisenberg electric-field operator with positive-frequency part $E^{+}(t)$, which is proportional to the lowering operator S^{-} [7]. It is well known that the fluorescence spectrum consists of three distinct lines at high intensity of the driving field, or at a large detuning Δ [8]. These lines can be viewed as arising from spontaneous transitions between the dressed states, which are the eigenstates of the combined Hamiltonian of the atom and the field [9]. These states form a ladder of pairs, where subsequent pairs are separated by one photon energy $\hbar\omega$ of the radiation field. In the present semiclassical treatment, we can find the energy separation of a pair by diagonalizing the Hamiltonian (2.3) [10]. These eigenstates $|1\rangle$ and $|2\rangle$ are related to the unperturbed atomic states by the simple rotations

$$|1\rangle=c|g\rangle-s|e\rangle,\quad |2\rangle=s|g\rangle+c|e\rangle , \tag{2.7}$$

with

$$c=\left[\frac{\Omega'+\Delta}{2\Omega'}\right]^{1/2},\quad s=\left[\frac{\Omega'-\Delta}{2\Omega'}\right]^{1/2}, \tag{2.8}$$

where

$$\Omega'=(\Omega^2+\Delta^2)^{1/2} \tag{2.9}$$

is the precession frequency, corresponding to the frequency separation of the eigenstates. The energy eigenvalues are

$$E_1=\tfrac{1}{2}\hbar\Omega',\quad E_2=-\tfrac{1}{2}\hbar\Omega' . \tag{2.10}$$

Spontaneous transitions from state $|1\rangle$ to state $|1\rangle$ or from state $|2\rangle$ to state $|2\rangle$ give rise to emission at frequency ω, and two sidebands at $\omega\pm\Omega'$ arise from the transitions $|1\rangle\rightarrow|2\rangle$ and $|2\rangle\rightarrow|1\rangle$. This is illustrated in Fig. 1. Hence the sideband photons are emitted in sequences $T,F,T,F,\ldots$, while the state changes accordingly. The central line at frequency ω is the Rayleigh line (R). For $\Delta>0$, the low-frequency sideband at $\omega-\Omega'$ is nearest to the atomic resonance frequency ω_0, and it is called the fluorescence line (F). The opposite sideband can be understood to arise from at least a three-photon excitation process [9], and it is denoted as the three-photon line (T). It is convenient to express the raising and lowering operators in terms of the dressed states, and we obtain

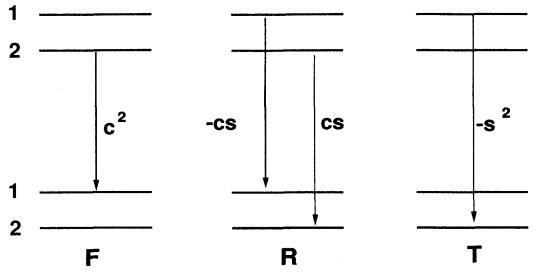

FIG. 1. Three components of the fluorescence spectrum, arising from spontaneous decay down a ladder of pairs of dressed states. The multiplication factors in the transition amplitudes are indicated. These factors correspond to Eq. (2.12).

$$S^{\pm}=S_F^{\pm}+S_T^{\pm}+S_R^{\pm} , \tag{2.11}$$

with

$$\begin{aligned} S_F^{-}&=c^2|1\rangle\langle 2|,\quad S_T^{-}=-s^2|2\rangle\langle 1| ,\\ S_R^{-}&=cs[|2\rangle\langle 2|-|1\rangle\langle 1|] , \end{aligned} \tag{2.12}$$

and likewise for the Hermitian conjugates. Hence atomic decay from $|e\rangle$ to $|g\rangle$ contributes to all three components. The branching ratios are determined by the relative amplitudes, which are given by the scalar factors in (2.12). These amplitudes are indicated in Fig. 1.

The linewidths are of the order of the spontaneous decay rate A, so that they are well separated when $A\ll\Omega'$. Then the evolution equation can be simplified, and the coupling between the dressed-atom populations and coherences may be neglected. The resulting approximate evolution equation takes the form (2.1), with the spontaneous decay operator replaced by Γ_d, defined as [11]

$$\Gamma_d\sigma=\tfrac{1}{2}A\sum_{\alpha=F,T,R}[S_\alpha^{+}S_\alpha^{-}\sigma+\sigma S_\alpha^{+}S_\alpha^{-}-2S_\alpha^{-}\sigma S_\alpha^{+}] . \tag{2.13}$$

This expression results after substituting the expansions (2.11) into (2.4), and neglecting the mixed terms with $\alpha\neq\alpha'$. The resulting approximate evolution equations for the density-matrix elements on the basis of the dressed states (2.8) take the explicit form

$$\begin{aligned} \frac{d}{dt}\sigma_{11}&=-\frac{d}{dt}\sigma_{22}=A[c^4\sigma_{22}-s^4\sigma_{11}] ,\\ \frac{d}{dt}\sigma_{21}&=-(\gamma-i\Omega')\sigma_{21} ,\\ \frac{d}{dt}\sigma_{12}&=-(\gamma+i\Omega')\sigma_{12} , \end{aligned} \tag{2.14}$$

with

$$\gamma=\frac{A}{2}(1+2c^2s^2) . \tag{2.15}$$

We summarize the evolution (2.14) by writing

$$\frac{d}{dt}\sigma = -iL_d\sigma \ , \tag{2.16}$$

which defines the dressed-atom Liouville operator L_d. The normalized steady-state solution $\bar{\sigma}$ of (2.14) has matrix elements

$$\bar{\sigma}_{11}=\frac{c^4}{c^4+s^4}, \quad \bar{\sigma}_{22}=\frac{s^4}{c^4+s^4}, \quad \bar{\sigma}_{12}=\bar{\sigma}_{21}=0 \ . \tag{2.17}$$

It is typical for the situation of well-separated lines that the dressed-state populations only depend on the ratio Δ/Ω. The strengths of the three separate lines (expressed in the number of photons per unit time) can be found from the expressions

$$I_\alpha = A\langle S_\alpha^+(t)S_\alpha^-(t)\rangle \ . \tag{2.18}$$

In the steady state this is equal to

$$I_\alpha = A\,\mathrm{Tr}\bar{\sigma}S_\alpha^+S_\alpha^- \ , \tag{2.19}$$

which gives the result

$$I_F=I_T=A\frac{c^4s^4}{c^4+s^4}, \quad I_R+Ac^2s^2 \ . \tag{2.20}$$

The equal strength of the sidebands is an obvious consequence of the fact that sideband photons alternate. The sidebands are Lorentzians located at $\omega\pm\Omega'$, with half width at half maximum (HWHM) equal to γ. The central Rayleigh line at the frequency ω is the superposition of a coherence line with zero width and strength [9,10]

$$I_{Rc}=I_R\left[\frac{c^4-s^4}{c^4+s^4}\right]^2 , \tag{2.21}$$

and an incoherent Lorentzian line with width

$$\gamma_0 = A(c^4+s^4) \tag{2.22}$$

and strength

$$I_{Ri}=I_R\frac{4c^4s^4}{(c^4+s^4)^2} \ . \tag{2.23}$$

III. FILTERED INTENSITIES

Before turning to the intensity correlation functions, we describe in this section the intensity of the output of a single filter. The filter width λ can be smaller or larger than the linewidths of the components, which are of the order of A. When the photons emitted in one of the components of the triplet are detected with a narrow spectral filter with setting frequency ω_1, the fluorescence field operator $E^+(t)$ must be replaced by the filtered field [12]

$$\bar{E}^+(t)=\int_0^\infty d\tau\,\lambda e^{-\lambda\tau-i\omega_1\tau}E^+(t-\tau) \ . \tag{3.1}$$

Since the emitted field E^+ is proportional to the Heisenberg lowering operator S^-, a similar relation holds between $\bar{S}^-$ and S^-. We consider the limit of well-separated spectral lines, so that both the linewidths and the passband width of the filter are small compared with the line separation Ω'. Hence we assume throughout this paper that

$$A \ll \Omega', \quad \lambda \ll \Omega' \ . \tag{3.2}$$

In this limit and for a setting frequency ω_1 within the width of the component α ($\alpha=F,T,R$), only this component contributes. Then the lowering operator S^- may be replaced by S_α^-, and we obtain the relation

$$\bar{S}_\alpha^-(t)=\int_0^\infty d\tau\,\lambda e^{-\lambda\tau-i\omega_1\tau}S_\alpha^-(t-\tau) \ . \tag{3.3}$$

The integral illustrates that a photon detection at time t implies an indeterminacy of the instant of emission. The filtered intensity is then obtained by replacing in (2.18) the raising and lowering operators by their filtered counterparts, so that

$$\bar{I}(\omega_1,\alpha)=A^2\int_0^\infty d\tau\int_0^\infty d\tau'\lambda^2e^{-\lambda(\tau+\tau')}e^{-i\omega_1(\tau-\tau')} \times\langle S_\alpha^+(t-\tau')S_\alpha^-(t-\tau)\rangle \ . \tag{3.4}$$

The right-hand side of (3.4) contains a twofold correlation function, which in the steady state depends only on the time difference $\tau-\tau'$. According to the quantum regression theorem [13], the decay of the correlation function is governed by the same evolution operator that describes the evolution of the density matrix. An explicit expression for the steady-state correlation function is found to be

$$\langle S_\alpha^+(t-\tau')S_\alpha^-(t-\tau)\rangle = e^{i\omega(\tau-\tau')}\mathrm{Tr}S_\alpha^+e^{-iL_d(\tau-\tau')}(S_\alpha^-\bar{\sigma}) \tag{3.5}$$

for $\tau>\tau'$, and

$$\langle S_\alpha^+(\tau-\tau')S_\alpha^-(t-\tau)\rangle = e^{i\omega(\tau-\tau')}\mathrm{Tr}\{e^{-iL_d(\tau'-\tau)}(\bar{\sigma}S_\alpha^+)\}S_\alpha^- \ , \tag{3.6}$$

for $\tau'>\tau$. The oscillatory exponential accounts for the transformation back to the nonrotating frame. It is useful to notice that the correlation function obeys the symmetry relation

$$\langle S_\alpha^+(t-\tau')S_\alpha^-(t-\tau)\rangle=\langle S_\alpha^+(t-\tau)S_\alpha^-(t-\tau')\rangle^* \ , \tag{3.7}$$

which shows that its real part is an even function of the time difference $\tau-\tau'$, whereas the imaginary part is odd. This implies that the contributions to (3.4) from the region where $\tau>\tau'$ and from the region where $\tau<\tau'$ are each other's complex conjugate. Hence it is sufficient to evaluate only the first contribution and take twice the real part of the result. Next we use as new integration variables τ and $s=\tau-\tau'$. This yields the expression

$$\bar{I}(\omega_1,\alpha)=\lambda A^2\mathrm{Re}\int_0^\infty ds\,e^{-\lambda s}e^{-i(\omega_1-\omega)s} \times\mathrm{Tr}S_\alpha^+e^{-iL_ds}(S_\alpha^-\bar{\sigma}) \ . \tag{3.8}$$

If we introduce the detunings of the setting frequency ω_1 from the line centers,

$$\Delta_{1\alpha}=\omega_1-\omega_\alpha \ , \tag{3.9}$$

for $\alpha=F,T,R$, with

$$\omega_F=\omega-\Omega',\quad \omega_T=\omega+\Omega',\quad \omega_R=\omega\ , \tag{3.10}$$

we arrive at the results

$$\begin{aligned}\bar{I}(\omega_1,F)&=I_F\mathrm{Re}\frac{\lambda}{\lambda+\gamma+i\Delta_{1F}},\\ \bar{I}(\omega_1,T)&=I_T\mathrm{Re}\frac{\lambda}{\lambda+\gamma+i\Delta_{1T}}\ ,\\ \bar{I}(\omega_1,R)&=I_{Ri}\mathrm{Re}\frac{\lambda}{\lambda+\gamma_0+i\Delta_{1R}}+I_{Rc}\mathrm{Re}\frac{\lambda}{\lambda+i\Delta_{1R}}\ .\end{aligned} \tag{3.11}$$

When the filter bandwidth λ is much larger than the linewidths γ and γ_0, we may neglect γ and γ_0 in the denominators in (3.11), and the intensity as a function of the filter setting frequency ω_1 of each component has a Lorentzian shape with width λ. For zero detunings $\Delta_{1\alpha}$ the intensities are equal to the line strengths I_α, given in (2.20). For values of λ that are smaller than the linewidths, the filtered intensities (3.11) follow the line profiles of the components. In particular, the coherent and the incoherent part of the Rayleigh line become separately visible.

IV. INTENSITY CORRELATIONS

We consider the case that the fluorescence is observed through two filters with setting frequencies ω_1 and ω_2, with the same linewidth λ. The filtered fields are denoted by $\bar{E}_1^+$ and $\bar{E}_2^+$. The standard expression for the intensity correlation function between the filtered fields is proportional to $\langle \bar{E}^-(0)\bar{E}^-(t)\bar{E}^+(t)\bar{E}^+(0)\rangle$, indicating a successive detection of a photon at time 0 and time t. When ω_1 falls within the linewidth of the component α, and ω_2 within the width of β, we can replace $\bar{E}_1^+$ by $\bar{S}_\alpha^-$, and $\bar{E}_2^+$ by $\bar{S}_\beta^-$, where the filtered operators $\bar{S}_\alpha^-$ and $\bar{S}_\beta^-$ are expressed by relations as (3.2). Since the operators S_α^- and S_β^- are noncommuting quantum-mechanical operators, their order is essential. As has been demonstrated by various authors [14,15], in the resulting expression for the intensity correlation function the time-dependent operators have to be arranged in a pyramidical order, so that the earlier lowering operators S_α^- or S_β^- are moved to the right, and the earlier raising operators S_α^+ or S_β^+ are moved to the left. This simply represents the time ordering of Heisenberg operators, where operators at earlier time have to operate first.

The correlation function is basically the average of the square of the amplitude for two successive detections. Such an amplitude is the sum of two terms, since the first detected photon could have been emitted before or after the emission of the second detected photon. Correspondingly, the correlation function can be expressed in the form

$$\bar{I}_2(\omega_1,\alpha;\omega_2,\beta;t)=A^2\langle F^\dagger(\alpha\beta,t)F(\alpha\beta,t)\rangle\ , \tag{4.1}$$

in terms of the two-photon detection operator

$$\begin{aligned}F(\alpha\beta,t)=\lambda^2\int_0^\infty d\tau_1\int_0^\infty d\tau_2 e^{-\lambda(\tau_1+\tau_2)}e^{-i\omega_1\tau_1-i\omega_2\tau_2}\\ \times[\Theta(t-\tau_2+\tau_1)S_\beta^-(t-\tau_2)S_\alpha^-(-\tau_1)+\Theta(-t+\tau_2-\tau_1)S_\alpha^-(-\tau_1)S_\beta^-(t-\tau_2)]\ .\end{aligned} \tag{4.2}$$

Here Θ is the Heaviside step function. When the time delay t between the two detections is significantly larger than λ^{-1}, the second term in (4.2) becomes negligible, and the order of detection must correspond to the order of emission. Substituting (4.2) into (4.1) gives for the two-time correlation function for detection of a photon passing filter 1 at time 0, and detection of a photon passing filter 2 at time $t\geq 0$,

$$\begin{aligned}&\bar{I}_2(\omega_1,\alpha;\omega_2,\beta;t)\\ &=A^2\int_0^\infty d\tau_1 d\tau_2 d\tau_1' d\tau_2'\lambda^4 e^{-\lambda(\tau_1+\tau_2+\tau_1'+\tau_2')}e^{-i\omega_1(\tau_1-\tau_1')}e^{-i\omega_2(\tau_2-\tau_2')}\\ &\quad\times[\Theta(t-\tau_2+\tau_1)\Theta(t-\tau_2'+\tau_1')\langle S_\alpha^+(-\tau_1')\langle S_\beta^+(t-\tau_2')S_\beta^-(t-\tau_2)S_\alpha^-(-\tau_1)\rangle\\ &\qquad+\Theta(t-\tau_2+\tau_1)\Theta(-t+\tau_2'-\tau_1')\langle S_\beta^+(t-\tau_2')S_\alpha^+(-\tau_1')S_\beta^-(t-\tau_2)S_\alpha^-)(-\tau_1)\rangle\\ &\qquad+\Theta(-t+\tau_2-\tau_1)\Theta(t-\tau_2'+\tau_1')\langle S_\alpha^+(-\tau_1')S_\beta^+(t-\tau_2')S_\alpha^-(-\tau_1)S_\beta^-(t-\tau_2)\rangle\\ &\qquad+\Theta(-t+\tau_2-\tau_1)\Theta(-t+\tau_2'-\tau_1')\langle S_\beta^+(t-\tau_2')S_\alpha^+(-\tau_1')S_\alpha^-(-\tau_1)S_\beta^-(t-\tau_2)\rangle]\ .\end{aligned} \tag{4.3}$$

The first term corresponds to the case that the order of emission corresponds to the order of detection, whereas the last term expresses the situation that the last detected photon was emitted first. The other two terms represent quantum-mechanical interference between these two cases. This interference arises from the uncertainty of the order of λ^{-1} in the instants of emission for known detection instants. It is customary to present results for the normalized correlation function

$$\begin{aligned}&g_2(\omega_1,\alpha;\omega_2,\beta;t)\\ &\quad=\bar{I}_2(\omega_1,\alpha;\omega_2,\beta;t)/[\bar{I}(\omega_1,\alpha)\bar{I}(\omega_2,\beta)]\ ,\end{aligned} \tag{4.4}$$

which approaches unity in the limit of large delay time t.

Each of the four terms contains a correlation function of the type $\langle S^+(t_1')S^+(t_2')S^-(t_2)S^-(t_1)\rangle$, with $t_2'\geq t_1'$ and $t_2\geq t_1$. These terms can in principle be evaluated by repeated use of the quantum regression theorem. The explicit expression for a four-time correlation function still

depends on the time order of the four arguments. One notices that there are six possible orders, namely, $t_2' \geq t_1' \geq t_2 \geq t_1$, $t_2' \geq t_2 \geq t_1' \geq t_1$, $t_2' \geq t_2 \geq t_1 \geq t_1'$, and these orders with t_i and t_i' interchanged. Accordingly, the integration region separates into six subregions, each corresponding to one of the possible time orders. For instance, when $t_2' \geq t_1' \geq t_2 \geq t_1$, the steady-state four-time correlation function is

$$\langle S_\alpha^+(t_1')S_\beta^+(t_2')S_\beta^-(t_2)S_\alpha^-(t_1)\rangle = e^{i\omega(t_1'+t_2'-t_1-t_2)}\mathrm{Tr}\{e^{-iL_d(t_2'-t_1')}[(e^{-iL_d(t_1'-t_2)}\{S_\beta^- e^{-iL_d(t_2-t_1)}[S_\alpha^-\bar{\sigma}]\})S_\alpha^+]S_\beta^+\}\ . \tag{4.5}$$

In total, the integrand in (4.3) contains 24 such terms. In each term, the four instants of time embrace three time intervals, and the integrals can be performed by integrating the lengths of these intervals from 0 to ∞, while still accounting for the step functions in (4.3). An explicit evaluation of the intensity correlation is complicated in general, but simple results arise in special cases.

V. LARGE FILTER WIDTHS

The general expressions (4.3) and (3.11) are valid in the limit of well-separated lines, as indicated by the inequalities (3.2). Explicit results still depend on the ratio of the filter passband width λ and the linewidths, which are of the order A. In this section, we discuss the intensity correlation functions in the case of a sufficiently large filter width, which samples an entire component. Hence we assume that $\lambda \gg A$. This is the common case treated so far. For later comparison we briefly recall and generalize the results in this section. We distinguish the short-time and the long-time behavior of the correlation functions.

A. Long-time behavior

First we consider the case of long correlation times, where

$$t \gg \lambda^{-1}\ . \tag{5.1}$$

Then the difference t between the two detection times is large compared with the time delay between emission and detection, so that this time delay becomes irrelevant. In particular, the order of emission and detection is the same, so that only the first term in (4.3) contributes. Results for this situation have been obtained by several authors [16,11,17,18]. The intensity correlation function $\bar{I}_2$ is then correctly described by the simple picture of two instantaneous emissions, separated by free evolution. The emission corresponds to a transition between two dressed states, as indicated in Fig. 1. The free evolution in between the two emissions describes the transition between the final state of the first emission to the initial state of the second emission. The normalized correlation g_2 is then independent of the setting frequencies ω_1 and ω_2, when these are within a frequency distance of the order of λ from line center. The time dependence is determined by the slow evolution described by the spontaneous emission operator Γ_d in between two spontaneous emissions. The result can be expressed as [18]

$$g_2(\omega_1,\alpha;\omega_2,\beta;t) = A^2\mathrm{Tr}S_\beta^-[e^{-\Gamma_d t}(S_\alpha^-\bar{\sigma}S_\alpha^+)]S_\beta^+/I_\alpha I_\beta\ . \tag{5.2}$$

For two photons from a single sideband the correlation is given by [16]

$$g_2(\omega_1,F;\omega_2,F;t) = g_2(\omega_1,T;\omega_2,T;t) = 1-e^{-\gamma_0 t}\ . \tag{5.3}$$

For a time difference $t=0$, g_2 vanishes in this case. This antibunching behavior results from the alternating character of sideband photons, which implies the fact that after the first emission in a sideband, an emission in the other sideband must occur before the second emission is possible. For a similar reason, the correlation between two photons from opposite sidebands displays bunching, since the first emission puts the atom in the dressed state where the second emission starts. The correlation functions in this case take the form [16]

$$\begin{aligned} g_2(\omega_1,F;\omega_2,T;t) &= 1+\frac{s^4}{c^4}e^{-\gamma_0 t}\ , \\ g_2(\omega_1,T;\omega_2,F;,t) &= 1+\frac{c^4}{s^4}e^{-\gamma_0 t}\ . \end{aligned} \tag{5.4}$$

For $\Delta \geq \Omega'$, the dressed state $|1\rangle$ is mainly the ground state $|g\rangle$, with an admixture of the excited state $|e\rangle$. Then we have $c^4 > s^4$, so that the steady-state population of state $|1\rangle$ is larger than that of state $|2\rangle$, and the bunching is strongest for an F photon following a T photon. This time asymmetry has been observed experimentally [19]. The time dependence of (5.3) and (5.4) is determined by the decay rate of the dressed-state populations to their steady-state value. Finally, photons from the central Rayleigh line are uncorrelated to other emissions [18], and one derives from (5.1)

$$g_2(\omega_1,R;\omega_2,\beta;t) = g_2(\omega_1,\alpha;\omega_2,R;t) = 1\ . \tag{5.5}$$

This is understandable from the dressed-state picture, since emission of an R photon does not affect the atomic state, and the emission probability of an R photon is independent of the density matrix.

B. Short-time behavior

The correlation functions for a detection time difference t obeying the inequality

$$t \ll A^{-1} \tag{5.6}$$

have received attention only recently [4]. In this case the typical time delay λ^{-1} between emission and detection is small compared with A^{-1}, and we may conclude that all time arguments occurring in the integrand in (4.3) are close together compared with the dressed-state evolution time A^{-1}. On this time scale, we can ignore the damp-

ing terms of order A in the dressed-state evolution, and only the Rabi precession remains. The intensity correlation function is given by (4.1), where now the two-photon detection operator takes the explicit form

$$F(\alpha\beta,t)=\frac{\lambda^2}{\lambda+i\Delta_{2\beta}}\left[\left[\frac{1}{\lambda+i\Delta_{1\alpha}}-\frac{e^{-(\lambda+i\Delta_{2\beta})t}}{2\lambda+i(\Delta_{1\alpha}+\Delta_{2\beta})}\right]S_\beta^- S_\alpha^- + \frac{e^{-(\lambda+i\Delta_{2\beta})t}}{2\lambda+i(\Delta_{1\alpha}+\Delta_{2\beta})}S_\alpha^- S_\beta^-\right] . \quad (5.7)$$

For two photons from the same line ($\alpha=\beta$), the time dependence in (5.7) disappears, and the intensity correlation function does not vary on the short-time scale of the order λ^{-1}.

The short-time correlations between a photon detection from a sideband and a photon from the central Rayleigh line are easily evaluated if we use the identities

$$S_\alpha^- S_R^- = -S_R^- S_\alpha^- , \quad (5.8)$$

for $\alpha=F$ or T. The relations (5.8), which follow from (2.12), show that the amplitude for successive emission of an R photon and a photon from one of the sidebands is just the opposite of the amplitude for the twofold emission in the opposite order [3]. If we substitute (5.8) and (5.7) into (4.1), we obtain an explicit result for the short-time behavior of the normalized correlation function between the Rayleigh line and a sideband in the form

$$g_2(\omega_1,\alpha;\omega_2,\beta;t)=\left|1-\frac{2(\lambda+i\Delta_{1\alpha})}{2\lambda+i(\Delta_{1\alpha}+\Delta_{2\beta})}\times e^{-(\lambda+i\Delta_{2\beta})t}\right|^2 \quad (5.9)$$

for the cases $(\alpha,\beta)=(R,F)$, (R,T), (F,R), or (T,R). This expression generalizes an earlier result that was obtained in the special case that $\Delta_{1\alpha}=\Delta_{2\beta}=0$ [4]. The correlation function (5.9) vanishes for $t=0$ when the two detunings of the setting frequencies from line center are equal, so that $\Delta_{1\alpha}=\Delta_{2\beta}$. This complete antibunching arises from fully destructive interference between the opposite orders of emission that contribute to the detection of the two photons at the same time instant. For unequal detuning, the antibunching is incomplete. When the two detunings are each other's opposite, so that $\Delta_{1\alpha}=-\Delta_{2\beta}=\delta$, then (5.9) is equal to

$$g_2(\omega_1,\alpha;\omega_2,\beta;t)=f(\delta,t) , \quad (5.10)$$

where the function f is defined by

$$f(\delta,t)=\left|1-\frac{\lambda+i\delta}{\lambda}e^{-(\lambda-i\delta)t}\right|^2 . \quad (5.11)$$

The behavior of f for various values of δ is illustrated in Fig. 2.

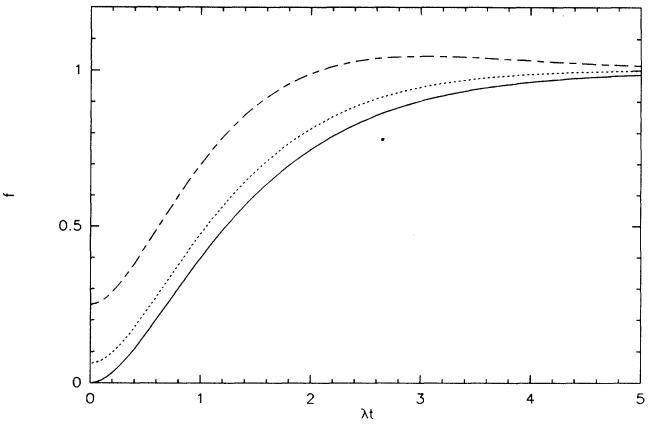

FIG. 2. Short-time behavior of the two-photon correlation function between a photon from the Rayleigh line and a sideband photon, in the case that the filter width λ is larger than the linewidths. The two filter detunings from line center are taken to be each other's opposite, so that $\Delta_{1\alpha}=-\Delta_{2\beta}=\delta$. Solid line: $\delta=0$; dotted line: $\delta=\lambda/4$; dashed line: $\delta=\lambda/2$. When the driving frequency is on resonance, the same function determines also the correlation between opposite sidebands, again for opposite filter detunings. This is expressed in Eq. (5.14).

Finally we turn to the case of two photons from opposite sidebands, so that $(\alpha,\beta)=(F,T)$ or (T,F). After substitution of (5.7) and its complex conjugate into (4.1), the cross terms disappear. This results from the fact that $S_F^+ S_T^- = S_T^- S_F^+ = 0$, according to (2.12). The one-time averages can be directly expressed in the steady-state populations $\bar{\sigma}_{11}$ and $\bar{\sigma}_{22}$. We obtain for the normalized correlation function

$$g_2(\omega_1,F;\omega_2,T;t)=\left[1+\frac{s^4}{c^4}\right]\left|1-\frac{\lambda+i\Delta_{1F}}{2\lambda+i(\Delta_{1F}+\Delta_{2T})}e^{-(\lambda+i\Delta_{2T})t}\right|^2+\left[1+\frac{c^4}{s^4}\right]\frac{\lambda^2+\Delta_{1F}^2}{4\lambda^2+(\Delta_{1F}+\Delta_{2T})^2}e^{-2\lambda t} , \quad (5.12)$$

$$g_2(\omega_1,T;\omega_2,F;t)=\left[1+\frac{c^4}{s^4}\right]\left|1-\frac{\lambda+i\Delta_{1T}}{2\lambda+i(\Delta_{1T}+\Delta_{2F})}e^{-(\lambda+i\Delta_{2F})t}\right|^2+\left[1+\frac{s^4}{c^4}\right]\frac{\lambda^2+\Delta_{1T}^2}{4\lambda^2+(\Delta_{1T}+\Delta_{2F})^2}e^{-2\lambda t} . \quad (5.13)$$

Again these expressions generalize the results of Ref. [4]. For a nonzero value of the detuning Δ of the driving field, the short-time behavior described by (5.12) and (5.13) provides a smooth connection between the different values of the correlation for opposite orders of detection inherent in the expressions (5.4). In the case that both detunings $\Delta_{1\alpha}$ and $\Delta_{2\beta}$ are zero, we plot both the short-time behavior of the correlation functions $g_2(\omega_1,F;\omega_2,T;t)$ and $g_2(\omega_1,T;\omega_2,F;t)$ for a few values of Δ/Ω in Fig. 3. Note that for $t=0$, the functions $g_2(FT)$ and $g_2(TF)$ coincide.

In the case that $\Delta=0$, the factors $s^4=c^4=\frac{1}{4}$ are equal.

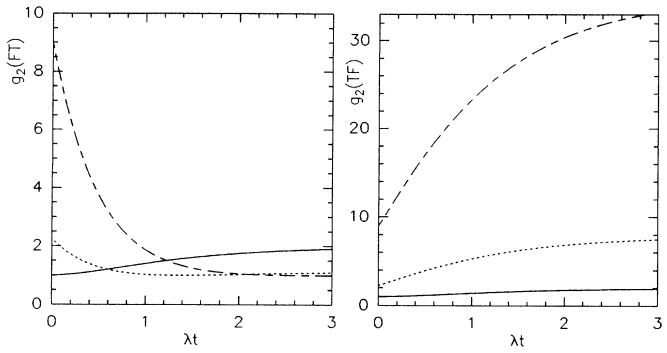

FIG. 3. Short-time behavior of the correlation functions between opposite sidebands, for large filter widths, and for filter setting frequencies on line centers. Solid line: detuning $\Delta=0$; dotted line: $\Delta=\Omega/2$; dashed line: $\Delta=\Omega$. Note the difference in scale.

Then the physical difference between the F line and the T line vanishes, and the two correlation functions (5.12) and (5.13) become identical. Furthermore, one checks that in this case the result (5.12) or (5.13) is identical to the result (5.9) plus 1. When, moreover, the two detunings are each other's opposite, so that $\Delta_{1F}=-\Delta_{2T}=\delta$, or $\Delta_{1T}=-\Delta_{2F}=\delta$, we obtain

$$g_2(\omega_1,F;\omega_2,T;t)=g_2(\omega_1,T;\omega_2,F;t)$$
$$=1+f(\delta,t)\ . \tag{5.14}$$

For equal detunings, so that $\Delta_{1\alpha}=\Delta_{2\beta}$, we find that $g_2(\omega_1,F;\omega_2,T;0)=g_2(\omega_1,T;\omega_2,F;0)=1$, and the dip exactly counterbalances the bunching behavior on the long-time scale, as described by (5.4) [4].

One should notice that the validity conditions (5.1) and (5.6) for the long-time and the short-time behavior overlap, so that together they cover all possible values of the detection time difference t.

VI. NARROW FILTERS

Now we consider the opposite case that the filter widths λ are small compared with the linewidths, so that $\lambda \ll A$. Then the filtering process implies a time integration over the past that is long compared with the correlation times of the Heisenberg emission operators. The quantum nature of successive noncommuting emissions is then important over the entire decay time of the intensity correlations. Therefore all 24 terms of the type (4.5) contribute to the correlation function (4.3). We have evaluated the correlation functions for all pairs of lines up to the lowest order in λ/A. The calculations are straightforward, but a bit tedious. Fortunately, the results are remarkably simple.

A. Correlation within sidebands

When both filters are set within the same sideband, the normalized correlation function g_2 is found to be

$$g_2(\omega_1,\alpha;\omega_2,\alpha;t)=1+\frac{4\lambda^2}{4\lambda^2+(\Delta_{1\alpha}-\Delta_{2\alpha})^2}e^{-2\lambda t}\ , \tag{6.1}$$

for $\alpha=F$ or T. When the two setting frequencies coincide, so that $\Delta_{1\alpha}=\Delta_{2\alpha}$, the correlation function attains the maximum value 2 for zero time difference t. The same results hold obviously when both detected photons emerge from a single filter. This demonstrates that the intensity correlation function of the output of a narrow filter set at a fluorescence sideband displays the bunching behavior that is characteristic for thermal light. When two filters are used with different frequency settings, the bunching behavior is diminished. This frequency dependence of (6.1) indicates that spectral correlations within a sideband are detected only insofar as the two filters overlap. It is interesting to notice that this expression (6.1) of the intensity correlation between the output of two filters coincides exactly with the situation of two narrow filters set within the bandwidth of a coherent light source described as a classical field with purely phase-fluctuating light [20]. In that case the input field has vanishing intensity fluctuations, so that the correlation function g_2 is unity. However, in the present case the input field of the filters is a fluorescence sideband, which displays antibunching, as indicated by the result (5.3). Apparently, this quantum characteristic of the input field is lost in the time integration inherent in the filtering process. Still, in the calculation of (6.1) the noncommutativity of the Heisenberg emission operators was accounted for.

B. Correlations between opposite sidebands

Now we consider the case that the two filters are set within opposite sidebands. According to (5.4), this means that the correlations between the two input fields are of the bunching type. Then the evaluation of (4.3) leads to the result

$$g_2(\omega_1,\alpha;\omega_2,\beta;t)=1+G\frac{4\lambda^2}{4\lambda^2+(\Delta_{1\alpha}+\Delta_{2\beta})^2}e^{-2\lambda t}\ , \tag{6.2}$$

for $\alpha=F$, $\beta=T$, or $\alpha=T$, $\beta=F$. Since the correlation is important only when the two detunings are each other's opposite, it is sufficient to specify the factor G in the case that $\Delta_{1\alpha}\cong-\Delta_{2\beta}\cong\delta$, and we obtain the expression in terms of the steady-state populations of the dressed states,

$$G=\frac{1+\delta^2(\bar\sigma_{11}-\bar\sigma_{22})^2/\gamma^2}{4\bar\sigma_{11}\bar\sigma_{22}}\ . \tag{6.3}$$

Substituting (2.17) then gives the result

$$G=\frac{\gamma^2(c^4+s^4)^2+\delta^2(c^4-s^4)^2}{4\gamma^2c^4s^4}\ . \tag{6.4}$$

When the excitation is on resonance, so that $\Delta=0$, G is equal to unity. When, moreover, the filter detunings are exactly each other's opposite, the value of g_2 at $t=0$ is equal to 2. When $\Delta\geq\Omega$, the population of the dressed state $|1\rangle$ is larger than the population of $|2\rangle$ and $c^4>s^4$. Then G can be much larger than unity, and the bunching can become correspondingly larger than 2. In Fig. 4 we plot the quantity G as a function of Δ/Ω, for various values of δ/A. Notice that the time asymmetry expressed in (5.4) in the order of detection of an F photon and a T photon has disappeared in the present case of a narrow filter.

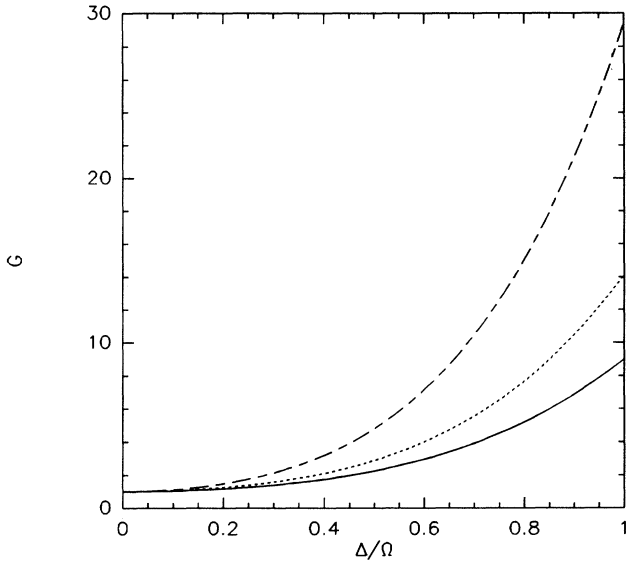

FIG. 4. Bunching factor G, characterizing the equal-time correlation between opposite sidebands for narrow filters, as a function of the detuning Δ of the driving light from resonance. The two detunings of the filter setting frequencies from line center are taken to be each other's opposite, so that $\Delta_{1\alpha}=-\Delta_{2\beta}=\delta$. Solid line: $\delta=0$; dotted line: $\delta=A/2$; dashed line: $\delta=A$.

C. Correlation between sidebands and the Rayleigh line

In calculating correlation functions involving a Rayleigh photon, we have to distinguish the narrow coherent Rayleigh line and the broader incoherent line with width γ_0. When the filter setting frequency ω_i is detuned from the driving frequency ω by an amount that is of the order of the filter width λ or less, then the output is dominated by the coherent line, and the contribution from the incoherent component can be ignored. If this detuning is larger than λ, the contribution from the coherent line can be neglected. In either case we find up to zeroth order in λ/γ that detection of a Rayleigh photon is uncorrelated to the detection of a photon in the sideband. Hence we find

$$g_2(\omega_1,\alpha;\omega_2,R;t)=g_2(\omega_1,R;\omega_2,\beta;t)=1\ , \tag{6.5}$$

for $\alpha=F$ or T, $\beta=F$ or T. This result may be compared with the corresponding expression (5.9), which is valid for short correlation times and $\lambda\geq\gamma$. The antibunching behavior due to destructive interference has disappeared in the present case of a narrow filter width λ.

D. Correlations within the Rayleigh line

Finally we turn to the case that both filters are set within the central Rayleigh line. When one or both of the setting frequencies are separated from the driving frequency ω by less than the filter width λ, so that the corresponding filter output is dominated by the coherent Rayleigh line, the two photon detections are fully uncorrelated, and the correlation function g_2 is equal to unity for all time separations t. The only remaining case occurs when both setting frequencies ω_i are separated from the line center ω by more than the filter width λ. If we evaluate (4.3) to lowest order in λ/γ, we obtain for the correlation function the result

$$g_2(\omega_1,R_i;\omega_2,R_i;t) = 1+\left[\frac{4\lambda^2}{4\lambda^2+(\Delta_{1\alpha}-\Delta_{2\beta})^2}+\frac{4\lambda^2}{4\lambda^2+(\Delta_{1\alpha}+\Delta_{2\beta})^2}\right]e^{-2\lambda t}\ . \tag{6.6}$$

Hence the incoherent Rayleigh line displays spectral correlations of the bunching type when both setting frequencies coincide, and also when the two detunings from line center ω are each other's opposite. The maximal value of the correlation function is 2 in either case.

VII. CONCLUSIONS

We have given a description of the photon correlations between the output of two spectral filters with passband widths λ set within the resonance fluorescence triplet. As a result of the time delay inherent in the filtering process, the time order in which the two photons are detected may differ from the order in which they have been emitted, provided that the difference t in detection time is smaller than λ^{-1}. Moreover, interference between opposite emission orders can occur. When the filter width λ is larger than the widths of the components in the spectrum, we have evaluated the short-time behavior of the correlation function, thereby generalizing previous results. Additional results are obtained when the filter width λ is small compared with the widths of the components in the triplet. This situation may be viewed as a measurement of spectral correlations of resonance fluorescence. Correlations are obtained both when the two setting frequencies are sufficiently close, so that the two filter widths overlap, and also when the setting frequencies are symmetrically positioned at opposite sides of the central frequency. All correlations are of the bunching type. The correlations can be particularly strong when the filters are set within opposite sidebands, provided that the detuning of the driving frequency is larger than the Rabi frequency. Spectral correlations of a light source provide information that may be viewed as complementary to the information contained in the intensity correlation without spectral resolution.

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