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Statistics of fluorescent photons emitted near a phase conjugator

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Temporal photon correlations and photon count statistics of fluorescence radiation, emitted by an atom in the vicinity of a phase conjugator, are considered. The fluorescent intensity and the two-photon correlation function are expressed in terms of photon emission operators. An explicit evaluation shows their dependence on the phase-conjugate reflectivity of the surface. The variance of the photon count distribution for long counting times is expressed in terms of the Q-factor, and Q has been evaluated as a function of the reflectivity. It appears that for a reflectivity larger than unity the photon correlation displays antibunching and the statistics is sub-poissonian.

I. INTRODUCTION

Emission of photons by atoms during spontaneous decay is very nearly a purely random process, and hence the statistics of these random events is nearly Poissonian. Correlations between photons and deviations from Poisson statistics have been the subject of many studies. Of particular importance is the two-time intensity correlation $I_2(t_1, t_2)$, which equals the photon detection rate at time t_2 after the detection of a photon at time t_1, independent of any photon detections at other times. For uncorrelated photons, this quantity factorizes as $I_2(t_1, t_2) = I(t_1)I(t_2)$, where $I(t)$ equals the uncorrelated photon detection rate at time t. Since $I(t)$ is the detection rate, the average number $\mu(t)$ of detected photons in a counting time interval $[0, t]$ is given by

$$\mu(t) = \int_0^t dt_1\ I(t_1) \tag{1.1}$$

For Poisson statistics, the variiance $\sigma^2(t)$ of the photon count distribution in $[0, t]$ equals the average $\mu(t)$. The deviation from Poissonian statistics is determined by the two-time intensity correlation according to[1]

$$\sigma^2(t) - \mu(t) = \int_0^t dt_1 \int_0^t dt_2\ [I_2(t_1, t_2) - I(t_1)\ I(t_2)] \tag{1.2}$$

It can be shown[2] that for electromagnetic fields which have a classical analogue, the variance is always larger than the everage, This is called super-Poissonian statistics. Conversely, sub-Poissonian statistics, for which $\sigma^2 < \mu$, can only occur in photon fields which are essentially quantum mechanical in nature. It follows from Eq. (1.2) that the photon statistics is sub-Poissnian when $I_2(t_1, t_2)$ is smaller, on the everage, than $I(t_1)I(t_2)$. For fields with a classical analogue, the equal-time correlation $I_2(t, t)$ is necessarily larger than $I(t)^2$. This phonomonon is termed photon bunching.[3~5] since it implies that the detection of a photon at time t enhances the probability for a photon detection immediately afterwards, as compared to the uncorrelated detection probability. Photon antibunching, for which $I_2(t, t) < I(t)^2$, is again a pure quantum feature of radiation. Both sub-poissonian statistics and antibunching have been observed experimentally in single-atom resonance fluorescence.[2, 6]

II. ATOM NEAR A PHASE CONJUGATOR

We consider a two-state atom, with ground state $|g\rangle$ and excited state $|e\rangle$, separated by $h\omega_0$, which is positiioned near the surface of a phase conjugator (PC). It is assumed that the PC consists of a nonlinear medium which is pumped by two strong counterpropagating laser beams with frequency $\bar{\omega}$. When weak radiation is incident on the surface of the medium, a four-wave mixing process generates a phase-conjugate replica of the incident field. This time- reversed image propagates into the direction opposite to the incoming wave, rather than into the specular direction (as for reflection at a linear medium).[7~9] Since the process is based on four-

wave mixing, an incident wave with frequency ω will be reflected as a wave with frequency $2\bar{\omega}$-ω.

When the atom is in its excited state $|e\rangle$ then it will decay spontaneously to its ground state $|g\rangle$, under emission of a fluorescent photon with frequency ω_0. This process is illustrated schematically in diagram (a) of Fig. 1. An atom in empty space or near a linear medium would then remain in its ground state indefinitely, because there is no excitation mechanism to bring the atom back to state $|e\rangle$. For an atom near a PC, however, the situation is different. When the atom is in state $|g\rangle$, it can not emit any radiation due to energy conservation, but the atom is still surrounded by the electromagnetic field which is generated by the electromagnetic field which is generated by its dipole moment. This non-radiative field with frequency $-\omega_0$ serves as an incident field on the PC, which then generates a phase-conjugate image at frequency $2\bar{\omega}$-ω_0. In the quantum version of four-wave mixing, a complex conjugation (which is phase conjugation) becomes a Hermitian conjugation. Therefore, an annihilation field will be reflected as a creation field, and vice versa, and a non-radiative incident field will be reflected as a radiative field, containing photons. The energy for the production of these photons is, of course, provided by the pump beams. It can be shown[10] from the retardation in the emission time that these photons with frequency $2\bar{\omega}$-ω_0 appear to emanate from the site of the atom. Consequently, we can effectively regard the four--wave mixing to take place at the location of the atom, rather than in the medium. The corresponding energy-conserving process is shown in diagram (b) of Fig. 1. The atom is originally in its ground state. Then it absorbs a photon with frequency $\bar{\omega}$ in a stimulated excitation. Subsequently, the fluorescent photon with frequency $2\bar{\omega}$-ω_0 and a second absorption of an $\bar{\omega}$-photon leaves the atom in the excited state. The cycle can then be completed with ordinary spontaneous decay. In this fashion, an atom near a PC can continuously generate fluorescent photons. The occurrence of the three-photon process from diagram (b) should be amenable to experimental verification because it generates photons with frequency $2\bar{\omega}$-ω_0, rather then the ω_0-photons of ordinary fluorescence. Also the spontaneous excitation of the atom during this process should be observable.

III. RELAXATION

Spontaneous decay gives rise to relaxation of the atomic density operator p(t). From the expression for the vacuum electromagnetic field[10] near a PC the damping operator Γ can be derived. With $P_e = |e\rangle\langle e|$, $P_g = |g\rangle\langle g|$ the projectors on the atomic states and in terms of the raising operator $d = |e\rangle\langle g|$ and the lowering operator $d^+ = |g\rangle\langle g|$ the Liouvillian Γ attains the form

$$\Gamma\sigma = 1/2A_e\{P_e\sigma + \sigma P_e - 2d^+\sigma d\} + 1/2A_g\{P_g\sigma + \sigma P_g - 2d\sigma d^+\}, \tag{3.1}$$

which defines its action on an arbiitrary Hilbert-space operator σ. The relaxation constants for the excited state and ground state are

$$A_e = A(1 + 1/2|P|^2), \tag{3.2}$$

$$A_g = 1/2A|P|^2, \tag{3.3}$$

respectively. Here, P is the Fresnel reflection coefficient for a plane wave and A is the Einstein coefficient for spontaneous decay in empty space. The Liouvillian of free evolution L_a is defined as

$$L_a\sigma = \omega_b[P_e, \sigma] \tag{3.4}$$

for arbitrary σ. Then the equation of motion for the density operator becomes

$$i\frac{d\rho}{dt} = (L_a - i\Gamma)\rho, \tag{3.5}$$

subject to the constraints $\rho^+ = \rho$ and $\mathrm{Tr}\rho = 1$.

Any initial state $\rho(0)$ will evolve to the steady state $\bar{\rho} = \rho((t = \infty))$ on a time scale of 1/A, as can be checked by inspection from Eq. (3.5). The steady state is found to be

$$\bar{\rho} = n_e P_e + n_g P_g, \tag{3.6}$$

in terms of the populations of the atomic levels

$$n_e = \frac{1/2|P|^2}{1 + |P|^2}, \quad n_g = \frac{1 + 1/2|P|^2}{1 + |P|^2} \tag{3.7}$$

Due to the occurrence of the three-photon processes, the population n_e of the excited state is finite.

IV. FLUORESCENCE

During spontaneous decay and excitation, the atomic dipole μ emits fluorescence radiation. When detected under an angle θ with the normal to the surface and filtered with a polarizer for its e_d component, the positive frequency part of the radiation assumes the form[10]

$$G(t)^{(+)} = \frac{\omega^2 e^{-i(\omega h/c)\cos\theta}}{4\pi\varepsilon_0 rc^2}\langle e|\mu e_d^*|g\rangle\{d^+(t) - P^* e^{-2i\bar{\omega}t}d(t)\} \tag{4.1}$$

Here, h is the normal distance between the atom and the surface, and the time dependence of $d^+(t)$ and $d(t)$ signifies the Heisenberg picture. The transition dipole moment $\langle e|\mu|g\rangle$ is assumed to be real. In Eq. (4.1), the term proportional to $d^+(t)$ is ordinary fluorescence which is emitted during an $|e\rangle - |g\rangle$ transition, and the rerm with $d((t)$ represents radiation ernitted during a three-photon process. The factor $\exp(-2i\bar{\omega}t)$ reflects the absorption of two photons with frequency $\bar{\omega}$.

In terms of $G(t)^{(+)}$ and its Hermitian conjugate $G(t)^{(-)}$,

the fluorescent intensity can be expressed as

$$I(t)=\zeta<G(t_1)^{(-)}G(t)^{(+)}>, \tag{4.2}$$

where ζ is a detector parameter, which accounts for the efficiency, the aperture, atc. Similarly, the two-photon correlation becomes

$$I_2(t_1, t_2)=\zeta^2<G(t_1)^{(-)}G(t_2)^{(-)}G(t_2)^{(+)}G(t_1)^{(+)}>, \tag{4.3}$$

for $t_2 \geq t_1$. In the steady state, $I(t)$ will be independent of time, and I_2 can only depend on t_1 and t_2 through $\gamma=t_2-t_1$.

V. INTENSITY

After transformation to the Schrödinger picture, the steady-state intensity can be written as

$$I=\zeta Tr(R_e+R_g)\bar{\rho}, \tag{5.1}$$

where we have absorbed an overall factor in ζ. The Liouville operators R_e and R_g are defined as

$$R_e\sigma=P_g<e \mid \sigma \mid e>, \tag{5.2}$$

$$R_g\sigma=\mid P \mid ^2P_e<g \mid \sigma \mid g>, \tag{5.3}$$

for arbitrary σ. The intensity acquires two contributions,

$$I=I_e+I_g, \tag{5.4}$$

with

$$I_\alpha=\zeta TrR_\alpha\bar{\rho},\ \alpha=e,\ g, \tag{5.5}$$

and with Eqs. (5.2) and (5.3) we readily obtain

$$I_e=\zeta n_e \tag{5.6}$$

$$I_g=\zeta \mid P \mid ^2n_g, \tag{5.7}$$

in terms of the level populations.

The total intensity as a function of the reflectivity $\mid P \mid ^2$ is then

$$I=1/2\zeta\chi\frac{3+x}{1+x}, \tag{5.8}$$

where we have set $\mid P \mid ^2=x$, Figure 2 illustrated the behavior of I with $\mid P \mid ^2$. Notice that for $x\rightarrow\infty$, I does not saturate but increases indefinitely.

Operators R_e and R_g have a clear physical interprctation. As follows from Eq. (5.2), the action of R_e on a density operator projects the atomic state onto $\mid g>$, and produces a factor equal to the population of $\mid e>$, Hence the effect of R_e on a density operator is to induce an $\mid e>\rightarrow \mid g>$ transition, and the process has a probability equal to the population of the excited state. This process corresponds to diagram (a) in Fig. 1. Alternatively, we could interpret R_e as the operator which accounts for the emission of a photon with frequency ω_0 during an $\mid e>\rightarrow \mid g>$ transition, We shall call these photons "e-photons".

Similarly, operator R_g represents the emission of a "g-photon", with frequency $2\bar{\omega}-\omega_0$, during a $\mid g>$ to $\mid e>$ excitation. This mechanism is depicted in diagram (b) of Fig. 1.

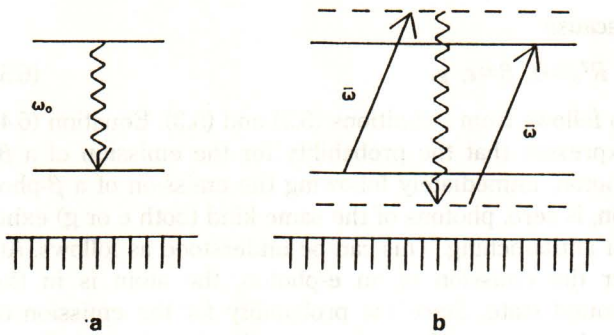

Fig. 1. Diagram (a) represents ordinary spontaneous decay from $\mid e>$to $\mid g>$, which is accompanied by the emission of a photon with frequency ω_0, In diagram (b), the atom is initially in the ground state. Then is absorbs a photona line misscing with frequency $2\bar{\omega}$-ω_0. To complete the energy-conserving diagram a second $\bar{\omega}$-photon is absorbed, which brings the atom to the excited state.

VI. TWO-PHOTON CORRELATION

The two-time intensity correlation from Eq. (4.3) can be evaluated in a similar way. Transforming into the Schrödinger picture and taking the stationary limit yields

$$I_2(0,\ \gamma)=\zeta^2Tr(R_e+R_g)e^{-i(L-ir)\gamma}(R_e+R_g)\bar{\rho} \tag{6.1}$$

The exponential for the time regression is identical to the time evolution operator for the atomic density operator. Equation (6.1) can be written as

$$I_2(0,\ \gamma)=\sum_{\zeta\beta}f_{\beta\alpha}(\gamma)I_\alpha, \tag{6.2}$$

where the summation runs over $\alpha=e, g$ and $\beta=e, g$. The four functions $f_{\beta\alpha}(\gamma)$ are defined as

$$f_{\beta\alpha}(\gamma)=\zeta^2I_\alpha{}^{-1}TrR_\beta e^{-1(L_\alpha-ir)\gamma}R_\alpha\bar{\rho}, \tag{6.3}$$

for $\gamma\geq 0$ the significance of $f_{\beta\alpha}(\gamma)$ follows from reading this expression from right to left. Operator R_α acts on $\bar{\rho}$, and it has the effect that an α-photon is emitted. It also leaves the atom in either $\mid e>$or $\mid g>$, according to Eqs. (5.2) and (5.3). Then the exponential lets this state evolve for a time γ, after which the action of R_β results in an emission of a β-photon. Division by I_α than renormalises the probability for the emission of an α-photon at time $\gamma=0$ to unity. The parameter ζ^2 relates the emission probabiilities for the two photons to the detection rate of β-photons at a time γ after the detection of an α-photon.

When we set $\gamma=0$ and $\alpha=\beta$ in Eq. (6.3), we obtain

$$f_{\beta\beta}(0)=0,\ \beta=e,\ g, \tag{6.4}$$

bacause

$$R^2_\beta=0,\ \beta=e,\ g, \tag{6.5}$$

as follows from definitions (5.2) and (5.3). Equation (6.4) expresses that the probability for the emission of a β-photon, immediately following the emission of a β-photon, is zero, photons of the same kind (both e or g) exhibit antibunching. This can be understood as follows. After the emission of an e-photon, the atom is in the ground state. Since the probability for the emission of a subsequent e-photon is proportional to the population of the excited state there must necessarily be a finite time delay γ during which the atom performs a $|g\rangle \rightarrow |e\rangle$ transition. Antibunching of g-photons has a similar explanation.

For long time delays between the two photons we obtain

$$f_{\beta\alpha}(\infty)=I_\beta, \tag{6.6}$$

e.g., the conditional detection rate for β-photons equals the uncorrelated intensity I_β. This implies that for $\gamma\rightarrow\infty$ the memory to the detection of an α-photon at $\gamma=0$ is lost, as could be expected.

When we set $\gamma=0$ and $\alpha=\beta$ in Eq. (6.3) then it is easy to show that

$$f_{\beta\alpha}(0)>I_\beta,\ \alpha\neq\beta. \tag{6.7}$$

This inequality expresses that the probability for the detection of a β-photon, immediately following an α-photon, is larger than the uncorrelated detection rate of β-photons. In other words, unlike photons tend to bunch. This property can readily be understood from Eqs. (5.2) and (5.3). After the emission of an e-photon at $\gamma=0$, the atom is in the ground state. The probability for the emission of a subsequent g-photon is proportional to the population of $|g\rangle$, and this is larger when the atom is in the ground state, as compared to an atom in the steady state ρ. Therefore, the emission of an e-photon enhances the probability for the subsequent emission of a g-photon. Bunching of e-photons after g-photons follows similarly.

Functions $f_{ee}(\gamma)$ and $f_{eg}(\gamma)$ are explicitly

$$f_{ee}(\gamma)=I_e(1-e^{-(A_e+A_g)\gamma}), \tag{6.8}$$

$$f_{eg}(\gamma)=I_e(1+(n_g/n_e)e^{-(A_e+A_g)\gamma}), \tag{6.9}$$

and $f_{gg}(\gamma)$ and $f_{ge}(\gamma)$ follow after interchanging e and g in these expressions. Combining everything then gives for the intensity correlation

$$I_2(0,\ \tau)=I^2\{1+(1-x)\frac{x^2+3x+4}{x(x+3)^2}\,e^{-(A_e+A_g)\tau}\}, \tag{6.10}$$

where $x=|P|^2$. This function is shown in Fig. 3 for three values of $|P|^2$. Notice that for unit reflectivity ($|P|^2=1$), this function factories as $I_2(0,\ \gamma)=I^2$ for all γ.

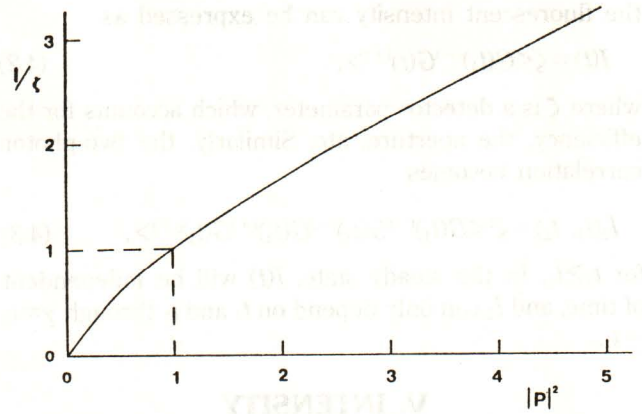

Fig. 3. Plot of QA/ζ as a function of $|P|^2$. At $|P|^2=1$ have $Q=0$, and this corresponds to the correlation function (b) in Fig. 3.

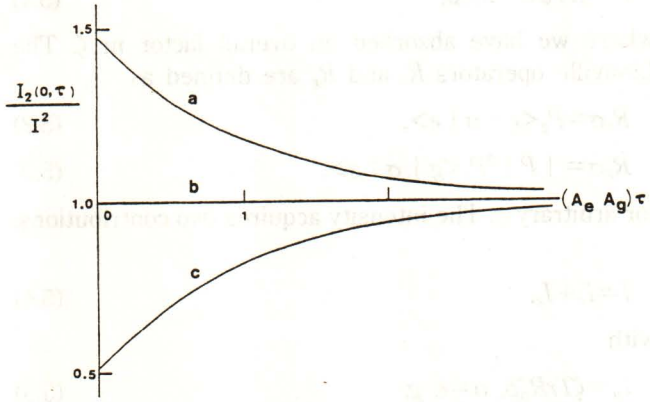

Fig. 2. Total intensity, divided by ζ, as a function of the reflectivity $|P|^2$. For $|P|^2\rightarrow 0$ the fluorescence vanishes, and for $|P|^2\rightarrow\infty$ the intensity increases unlimited.

VII. STATISTICS

With expression (6.10) for the two-photon correlation function, the steady-state variance can readily be obtained from Eq. (1.2). Of particular importance is the value of $\sigma^2(t)$ for long counting times (much larger than $1/(A_e+A_g)$). It is easy to verify that for $t\rightarrow\infty$ the variance becomes proportional to t. With Eq. (1.1) we find the average number of detected photons in $[0,\ t]$ to be $\mu(t)=It$, with I the counting rate in the steady state. Therefore, a normalized measure for the variance can be defined as

$$Q=\lim_{t\rightarrow\infty}\frac{\sigma^2(t)-\mu(t)}{\mu(t)}, \tag{7.1}$$

which is Mandel's Q-factor[2] for $t\rightarrow\infty$. From this definition it follows that Q is bounded by $Q\geq -1$. A negative value of Q reflects sub-poissonian statics, and $Q>0$ corresponds to super Poisson statistics of the photon counts.

The Q-factor of the fluorescence turns out to be

$$Q=\frac{\zeta}{A}\ \frac{1-x}{(1+x)^2}\ \frac{x^2+3x+4}{x+3} \tag{7.2}$$

where $x=|P|^2$. Here, ζ/A is merely an efficiency factor which relates the number of detected photons to the number of emitted photons. Therefore, the value of Q depends essentially only the intensity reflection coefficient $|P|^2$. The value of QA/ζ as a function of $|P|^2$ is shown in Fig. 4. It follows from Eq. (7.2) that the statistics of the fluorescent photons is sub-poissonian whenever the reflectivity exceeds unit ($x>1$). For $|P|^2 \to \infty$ the value of QA/ζ reaches its ultimate lower limit of -1. Since for $|P|^2 \to \infty$ the intensity increases unbounded, as illustrated in Fig. 2, this non-classical behavior should be amenable to experimental verification.

VIII. CONCLUSIONS

We have studied the statistical properties of photons in fluorescence radiation, which is emitted by an atom near the surface of a PC. The intensity of emission and the temporal correlations could be expressed in terms of the emission operators R_e and R_g for e-photons and g-photons, respectively. The time regression of the correlation function is brought about by the time-evolution operator for the atomic density operator. Emission of an e-photon is accompanied by ordinary spontaneous decay, whereas an emission of a g-photon requires the occurrence of a three-photon event. During such a process, two $\bar{\omega}$-photons are absorbed, a fluorescent photon with frequency $2\bar{\omega}-\omega_0$ is emitted, and the atom makes a transition from the ground state to the excited state.

It was shown that like photons exhibit antibunching and unlike photons tend to bunch. Without resolution with respect to the photon kind (by means of their frequency), the temporal behavior can either be bunching or antibunching, depending on the phase-conjugate reflectivity $|P|^2$. For $|P|^2<1$ bunching prevails, and the statistics is super-Poissonian, whereas for a reflectivity larger than unity the photons antibunch, leading to sub-Poissonian statistics. Equation (1.2) implies that antibunching (on the average) must necessarily yield a sub-Poisson photon count distribution.

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