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OFFICE OF NAVAL RESEARCH

Grant N00014-90-J-1193

TECHNICAL REPORT No. 63

Conditions for Sub-Poissonian Photon Statistics in Phase-Conjugated Resonance
Fluorescence

by

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Prepared for publication

in

Optics Communications

Departments of Chemistry and Physics
Washington State University
Pullman, WA 99164-1046

September 1991

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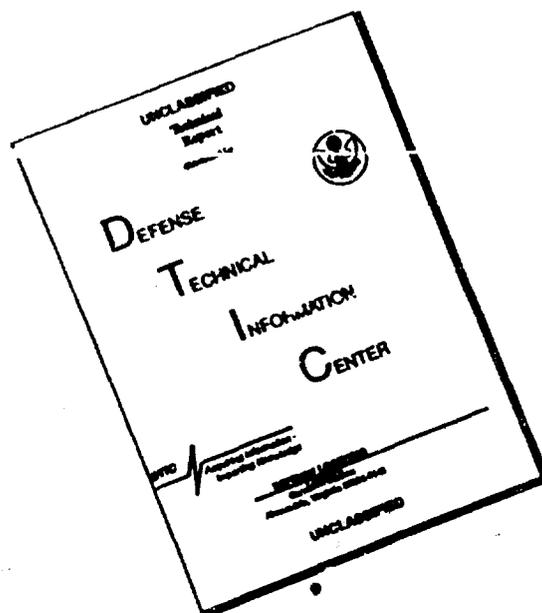
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REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. REPORT SECURITY CLASSIFICATION Unclassified		1b. RESTRICTIVE MARKINGS	
2. SECURITY CLASSIFICATION AUTHORITY		3. DISTRIBUTION / AVAILABILITY OF REPORT Approved for public release; distribution unlimited	
3. DECLASSIFICATION / DOWNGRADING SCHEDULE			
PERFORMING ORGANIZATION REPORT NUMBER(S) WSU/DC/91/TR-63		5. MONITORING ORGANIZATION REPORT NUMBER(S)	
4a. NAME OF PERFORMING ORGANIZATION Depts. Chemistry & Physics Washington State University	6b. OFFICE SYMBOL (if applicable)	7a. NAME OF MONITORING ORGANIZATION	
4c. ADDRESS (City, State, and ZIP Code) 428 French Administration Building Pullman, WA 99164-1046		7b. ADDRESS (City, State, and ZIP Code) Chemistry Program 800 N. Quincy Street Arlington, Virginia 22217	
5a. NAME OF FUNDING / SPONSORING ORGANIZATION Office of Naval Research	8b. OFFICE SYMBOL (if applicable)	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER Grant N00014-90-J-1193	
5c. ADDRESS (City, State, and ZIP Code) Chemistry Program 800 N. Quincy Street Arlington, Virginia 22217		10. SOURCE OF FUNDING NUMBERS	
		PROGRAM ELEMENT NO.	PROJECT NO.
		TASK NO.	WORK UNIT ACCESSION NO.
1. TITLE (Include Security Classification) Conditions for Sub-Poissonian Photon Statistics in Phase-Conjugated Resonance Fluorescence			
2. PERSONAL AUTHOR(S) Henk F. Arnoldus and Thomas F. George			
3a. TYPE OF REPORT	13b. TIME COVERED FROM _____ TO _____	14. DATE OF REPORT (Year, Month, Day) September 1991	15. PAGE COUNT 23
6. SUPPLEMENTARY NOTATION Prepared for publication in <u>Optics Communications</u>			
7. COSATI CODES		18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)	
FIELD	GROUP	SUB-GROUP	
			PHOTON STATISTICS
			SUB-POISSONIAN
			RESONANCE FLUORESCENCE
			CORRELATIONS
			PHASE CONJUGATED
			REFLECTIVITY
9. ABSTRACT (Continue on reverse if necessary and identify by block number) Photon correlations and statistics of phase-conjugated resonance fluorescence of a two-state atom is considered. The Q-factor, as a function of the incident laser power, the detuning, the laser linewidth and the phase-conjugate reflectivity, has been calculated. It is shown that for small and large reflectivity the statistics is predominantly sub-poissonian. For unit reflectivity the statistics appears to be exactly poissonian ($Q = 0$) for all values of the optical parameters.			
20. DISTRIBUTION / AVAILABILITY OF ABSTRACT <input checked="" type="checkbox"/> UNCLASSIFIED/UNLIMITED <input checked="" type="checkbox"/> SAME AS RPT <input type="checkbox"/> DTIC USERS		21. ABSTRACT SECURITY CLASSIFICATION Unclassified	
22a. NAME OF RESPONSIBLE INDIVIDUAL Dr. David L. Nelson		22b. TELEPHONE (Include Area Code) (202) 696-4410	22c. OFFICE SYMBOL

CONDITIONS FOR SUB-POISSONIAN PHOTON STATISTICS IN
PHASE-CONJUGATED RESONANCE FLUORESCENCE

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ABSTRACT

Photon correlations and statistics of phase-conjugated resonance fluorescence of a two-state atom is considered. The Q-factor, as a function of the incident laser power, the detuning, the laser linewidth and the phase-conjugate reflectivity, has been calculated. It is shown that for small and large reflectivity the statistics is predominantly sub-poissonian. For unit reflectivity the statistics appears to be exactly poissonian ($\langle n \rangle = 0$) for all values of the optical parameters.

PACS: 42.65.H, 32.80, 52.50.D

Such sub-poissonian statistics was predicted [4] and demonstrated experimentally [5] in single-atom resonance fluorescence.

The Q-factor can be expressed in terms of the two-photon correlation function $I_2(t_1, t_2)$. By definition, $I_2(t_1, t_2)dt_1dt_2$ is the probability for the detection of a photon in $[t_1, t_1+dt_1]$ and a photon in $[t_2, t_2+dt_2]$, irrespective of photon detections at other times. For stationary fields, $I_2(t_1, t_2)$ depends only on t_1 and t_2 through $\tau = t_2 - t_1$, and the Q-factor assumes the form [6]

$$Q(\tau) = \frac{2}{I\tau} \int_0^\tau d\tau \{ (\tau - \tau)I_2(0, \tau) - \tau I^2 \} . \quad (3)$$

In terms of the normalized correlation function

$$h(\tau) = \frac{I_2(0, \tau) - I^2}{I^2} , \quad (4)$$

the Q-factor can be written as

$$Q(\tau) = \frac{2I}{\tau} \int_0^\tau d\tau (\tau - \tau)h(\tau) . \quad (5)$$

Of particular interest is the limit of long counting times. For $\tau \rightarrow \infty$, Eq. (5) reduces to

$$Q(\infty) = 2I \int_0^\infty d\tau h(\tau) , \quad (6)$$

provided that the limit exists. When we adopt a Laplace transform

$$\bar{h}(s) = \int_0^{\infty} dr e^{-sr} h(r) \quad , \quad (7)$$

the long-time Q-factor becomes

$$Q(\infty) = 2I\bar{h}(0) \quad . \quad (8)$$

2. Phase-conjugated fluorescence

We consider a two-state atom, with excited state $|e\rangle$, ground state $|g\rangle$ and level separation $\hbar\omega_0$, which is positioned near the surface of a four-wave mixing phase conjugator (PC). The medium is pumped by two strong counterpropagating lasers with frequency $\bar{\omega}$. A laser beam with frequency ω_L is parallel to the surface and irradiates the atom. Resonance fluorescence is emitted by the atom and detected in the far field by a photomultiplier. It can be shown [7] that the negative-frequency part of the detected radiation is proportional to the Heisenberg operator

$$b(t) = d(t) - e^{2i\bar{\omega}t} P d^\dagger(t) \quad , \quad (9)$$

where $d = |e\rangle\langle g|$ is the atomic raising operator. The (complex) number P is the Fresnel reflection coefficient for a plane wave with frequency ω_0 , which is incident on the surface of the PC under the same angle as the location of the detector. This coefficient P depends in a complicated way on ω_0 , the angle of incidence and the polarization of the pump beams [8]. The term $d(t)$

in Eq. (9) represents ordinary fluorescence, and the term proportional to P is the phase-conjugated image which is reflected by the PC. We shall assume that the medium of the four-wave mixer is transparent, so that there is no ordinary specular reflection of the incident fluorescence radiation. In terms of $b(t)$, the intensity of the detected radiation is

$$I(t) = \xi \langle \langle b(t)b(t)^\dagger \rangle \rangle , \quad (10)$$

and the two-photon correlation is given by

$$I_2(t_1, t_2) = \xi^2 \langle \langle b(t_1)b(t_2)b(t_2)^\dagger b(t_1)^\dagger \rangle \rangle . \quad (11)$$

The parameter ξ contains the proportionality factor between $b(t)$ and the detected field, and ξ is proportional to the detector efficiency. An overall time retardation between emission and detection has been suppressed. The finite linewidth of the driving laser will be considered to be brought about by a stochastically fluctuating phase. The notation $\langle \dots \rangle$ in Eqs. (10) and (11) indicates an average over the random laser phase, whereas the notation $\langle \dots \rangle$ represents a quantum average.

Transforming Eq. (10) to the Schrödinger picture yields

$$I(t) = \xi \langle \text{Tr} \mathcal{B}(t) \rho(t) \rangle , \quad (12)$$

where we introduced the Liouville operator $\mathcal{B}(t)$, which is defined by its action on an arbitrary Liouville vector Π according to

$$\mathcal{B}(\tau)\Pi = (d^\dagger - e^{-2i\bar{\omega}\tau}P^*d)\Pi(d - e^{2i\bar{\omega}\tau}Pd^\dagger) \quad (13)$$

In expression (12), $\rho(\tau)$ is the atomic density operator. Its time evolution operator will be indicated by the Liouvillian $X(\tau, \tau')$, which is defined as

$$\rho(\tau) = X(\tau, \tau')\rho(\tau') \quad , \quad \tau \geq \tau' \quad (14)$$

With this notation, the two-photon correlation from Eq. (11) becomes in the Schrödinger picture

$$I_2(\tau_1, \tau_2) = \xi^2 \langle \text{Tr} \mathcal{B}(\tau_2) X(\tau_2, \tau_1) \mathcal{B}(\tau_1) \rho(\tau_1) \rangle \quad , \quad \tau_2 \geq \tau_1 \quad (15)$$

The terms inside the brackets $\langle \dots \rangle$ in both Eq. (12) and Eq. (15) are randomly fluctuating functions. Only their average over the stochastic laser phase will reach a steady state.

3. Equation of motion

The Hamiltonian of the two-state atom is given by

$$H_a = \hbar\omega_e P_e + \hbar\omega_g P_g \quad , \quad (16)$$

in terms of the projectors $P_e = |e\rangle\langle e|$ and $P_g = |g\rangle\langle g|$ on the excited state and ground state, respectively. The interaction between the atom and the laser field, in the rotating-wave approximation, is represented by the Hamiltonian

$$H_{ar}(t) = -\frac{1}{2} \Omega d e^{-i(\omega_L t + \phi(t))} + \text{H.c.} , \quad (17)$$

where Ω is the (complex) Rabi frequency of the dipole coupling and $\phi(t)$ is the stochastic laser phase. Then the equation of motion for the atomic density operator $\rho(t)$ becomes

$$i\hbar \frac{d\rho}{dt} = [H_a + H_{ar}(t), \rho] - i\hbar \Gamma \rho . \quad (18)$$

Spontaneous decay and excitation is accounted for by the Liouvillian Γ , given by

$$\Gamma \Pi = \frac{1}{2} A_e (P_e \Pi + \Pi P_e - 2d^\dagger \Pi d) + \frac{1}{2} A_g (P_g \Pi + \Pi P_g - 2d \Pi d^\dagger) , \quad (19)$$

which defines its action on an arbitrary Liouville vector Π . The rate constants A_e and A_g are [9]

$$A_e = A(1 + \frac{1}{2}|P|^2) , \quad A_g = \frac{1}{2}A|P|^2 , \quad (20)$$

with A the Einstein coefficient for spontaneous decay of an atom in empty space.

Oscillations with the laser frequency ω_L in the Hamiltonian can be eliminated with a transformation. With the Liouvillian L_g defined as

$$L_g \Pi = [P_g, \Pi] , \quad (21)$$

for Π arbitrary, the transformed density operator $\sigma(t)$ is given by [10]

$$\sigma(t) = e^{-i(\omega_L t + \phi(t))L_g} \rho(t) \quad (22)$$

From Eq. (18) we then obtain the equation of motion for $\sigma(t)$,

$$i \frac{d\sigma}{dt} = (L_d + \dot{\phi}(t)L_g - i\Gamma)\sigma \quad (23)$$

Here, the dressed-atom Liouvillian L_d equals

$$L_d \Pi = \Delta L_g \Pi - \frac{1}{2}[\Omega d + \Omega^* d^\dagger, \Pi] \quad (24)$$

with $\Delta = \omega_L - \omega_0$ the detuning from resonance.

Equation (23) is a stochastic differential equation for $\sigma(t)$. We shall take $\phi(t)$ to be the independent-increment process [11], which has the phase-diffusion model as its gaussian limit. Then Eq. (23) can be solved for the average, with the formal result [10]

$$\langle \sigma(t) \rangle = e^{-i(L_d - iW - i\Gamma)(t - t_0)} \langle \sigma(t_0) \rangle \quad (25)$$

The operator W accounts for the phase fluctuations and is given by

$$W = \lambda L_g^2 \quad (26)$$

with λ the half-width at half-maximum of the Lorentzian laser profile. The steady-state value $\langle \sigma(\infty) \rangle$ will be indicated by $\bar{\sigma}$, and is the solution of

$$(L_d - iW - i\Gamma)\bar{\sigma} = 0 \quad (27)$$

This equation is easily solved for the matrix elements of $\bar{\sigma}$. For the population of the excited state we obtain

$$\bar{n}_e = \langle e | \bar{\sigma} | e \rangle = \frac{1}{2} \frac{\Omega_0^2 \eta + A |P|^2 (\Delta^2 + \eta^2)}{\Omega_0^2 \eta + 2\bar{A} (\Delta^2 + \eta^2)} \quad (28)$$

where we have set $\bar{A} = \frac{1}{2}(A_e + A_g)$, $\eta = \bar{A} + \lambda$ and $\Omega_0 = |\Omega|$. The population of the ground state is $\bar{n}_g = 1 - \bar{n}_e$, and also the coherence $\langle e | \bar{\sigma} | g \rangle$ can readily be found (but is not needed here).

4. Intensity

With Eq. (22), expression (12) for the intensity can be transformed to the σ -representation, and with Eq. (13) this gives four separate terms. Then we take the stochastic average and the limit $t \rightarrow \infty$. In the σ -representation, the terms proportional to P and P^* acquire phase factors of the form $\exp(\pm 2i\phi(t))$. With the identity [12]

$$\lim_{t \rightarrow \infty} \langle e^{\pm 2i\phi(t)} \sigma(t) \rangle = 0 \quad (29)$$

these cross terms vanish identically in the steady state. When we introduce the Liouville operators \mathcal{R}_e and \mathcal{R}_g as

$$\mathcal{R}_e \Pi = d^\dagger \Pi d = P_g \langle e | \Pi | e \rangle \quad (30)$$

$$\mathcal{R}_g \Pi = |P|^2 d \Pi d^\dagger = |P|^2 P_e \langle g | \Pi | g \rangle \quad (31)$$

then the steady-state intensity can be written as

$$I = \xi \text{Tr}(\mathcal{R}_e + \mathcal{R}_g)\bar{\rho} \quad (32)$$

For the two contributions we write

$$I_\alpha = \xi \text{Tr} \mathcal{R}_\alpha \bar{\rho} \quad , \quad \alpha = e, g \quad (33)$$

and with Eqs. (30) and (31) this is $I_e = \xi \bar{n}_e$ and $I_g = \xi |P|^2 \bar{n}_g$, respectively.

The total intensity is

$$I = \sum_{\alpha} I_{\alpha} \quad (34)$$

and with Eq. (28) this becomes

$$I = \frac{1}{2} \frac{\Omega_0^2 \eta (1 + |P|^2) + A |P|^2 (\Delta^2 + \eta^2) (3 + |P|^2)}{\Omega_0^2 \eta + 2\bar{A}(\Delta^2 + \eta^2)} \quad (35)$$

The significance of the two contributions follows from Eqs. (30) and (31). The part I_e is brought about by the action of \mathcal{R}_e on the density operator $\bar{\rho}$, which gives $\mathcal{R}_e \bar{\rho} = \bar{n}_e |g\rangle\langle g|$. Therefore, the probability for the emission of an "e-photon" is proportional to the population of the excited state, and after the emission the atom is left in the ground state. Similarly, the action of \mathcal{R}_g produces a "g-photon". According to Eq. (31), the probability for this process is proportional to the population of the ground state, and after the emission the atom is in the excited state. It can be shown [7] that

this stimulated transition actually involves a three-photon process: two photons with frequency $\bar{\omega}$ are absorbed and a photon (the g-photon) with frequency $2\bar{\omega} - \omega_0$ is emitted as fluorescence. An atomic transition from $|g\rangle$ to $|e\rangle$ then guarantees conservation of energy. The e-photons are ordinary fluorescence in an $|e\rangle \rightarrow |g\rangle$ transition, and they have frequency ω_0 (in the weak-field limit).

5. Two-photon correlation

The two-photon correlation function from Eq. (15) can be worked out in the same way as the intensity. In the steady state ($t_1 \rightarrow \infty$, $t = t_2 - t_1$ fixed) the cross terms vanish identically, and we obtain

$$I_2(0, \tau) = \xi^2 \text{Tr}(\mathcal{R}_e + \mathcal{R}_g) e^{-i(L_d - iW - i\Gamma)\tau} (\mathcal{R}_e + \mathcal{R}_g) \bar{\sigma} . \quad (36)$$

This can be written as

$$I_2(0, \tau) = \sum_{\alpha, \beta} f_{\beta\alpha}(\tau) I_\alpha \quad (37)$$

in terms of the four functions

$$f_{\beta\alpha}(\tau) = \frac{\xi^2}{I_\alpha} \text{Tr} \mathcal{R}_\beta e^{-i(L_d - iW - i\Gamma)\tau} \mathcal{R}_\alpha \bar{\sigma} , \quad \alpha, \beta = e \text{ or } g . \quad (38)$$

From the interpretation of the \mathcal{R} operators it then follows that $f_{\beta\alpha}(\tau)$ equals the intensity of β -photons at a time τ after the detection of an α -photon.

From the identity

$$\lim_{\tau \rightarrow \infty} e^{-i(L_d - iW - i\Gamma)\tau} \Pi = \bar{\sigma} \text{Tr} \Pi \quad , \quad (39)$$

for arbitrary Π , we then obtain

$$\lim_{\tau \rightarrow \infty} f_{\beta\alpha}(\tau) = \xi \text{Tr} R_{\beta} \bar{\sigma} = I_{\beta} \quad . \quad (40)$$

This illustrates that for a long delay time τ the detection rate of β -photons equals the uncorrelated intensity I_{β} . With Eq. (37) this gives

$$\lim_{\tau \rightarrow \infty} I_2(0, \tau) = \sum_{\alpha, \beta} I_{\beta} I_{\alpha} = (I_e + I_g)^2 = I^2 \quad , \quad (41)$$

i.e., the correlation function factorizes. For $\tau \rightarrow \infty$ we find

$$f_{ee}(0) = f_{gg}(0) = 0 \quad , \quad (42)$$

$$f_{eg}(0) = \xi > I_e \quad , \quad (43)$$

$$f_{ge}(0) = \xi |P|^2 > I_g \quad . \quad (44)$$

Equation (42) expresses that the probability for the detection of an e-photon immediately after the detection of an e-photon is zero (antibunching). This can be understood from the fact that after the emission of an e-photon the atom is in the ground state. It takes a finite time τ for the atom to make a $|g\rangle \rightarrow |e\rangle$ transition, which is necessary for the emission of a subsequent e-photon. In a similar way it follows that $f_{gg}(0)$ must be zero. Equation (43)

shows that the probability for the detection of an e-photon immediately following a g-photon is larger than the uncorrelated probability for the detection of an e-photon (bunching). This follows from the fact that after the emission of an e-photon the atom is in the ground state, rather than σ . This enhances the probability for the emission of a g-photon, which is proportional to the population of the ground state. The inequality in Eq. (44) can be explained in a similar way. From Eqs. (42)-(44) and with $\bar{n}_e + \bar{n}_g + 1$ we find

$$I_2(0,0) = \xi^2 |P|^2 \quad . \quad (45)$$

It appears that $I_2(0,0)$ does not depend on any of the parameters Ω_0 , Δ , A or λ .

The Laplace transform of Eq. (38) is

$$\bar{F}_{\beta\alpha}(s) = \frac{\xi^2}{I_\alpha} \text{Tr} R_\beta \frac{1}{s + iL_d + W + \Gamma} R_\alpha \hat{\sigma} \quad , \quad (46)$$

in terms of an operator inversion. Working out this expression then yields for the four combinations

$$\bar{F}_{ee}(s) = \frac{\xi}{sD(s)} \left(\frac{1}{2}\Omega_0^2(s + \eta) + A_g [(s + \eta)^2 + \Delta^2] \right) \quad , \quad (47)$$

$$\bar{F}_{ge}(s) = \frac{\xi |P|^2}{sD(s)} \left(\frac{1}{2}\Omega_0^2(s + \eta) + (s+A_e) [(s + \eta)^2 + \Delta^2] \right) \quad , \quad (48)$$

$$\bar{F}_{eg}(s) = \frac{\xi}{sD(s)} \left(\frac{1}{2}\Omega_0^2(s + \eta) + (s+A_g) [(s + \eta)^2 + \Delta^2] \right) \quad , \quad (49)$$

$$\bar{I}_{gg}(s) = \frac{\xi |P|^2}{sD(s)} \left(\frac{1}{2} \Omega_0^2 (s + \eta) + A_e [(s + \eta)^2 + \Delta^2] \right) , \quad (50)$$

where

$$D(s) = \Omega_0^2 (s + \eta) + (s + 2\bar{A}) [(s + \eta)^2 + \Delta^2] . \quad (51)$$

With Eq. (37) we can then construct $\bar{I}_2(0, s)$. The result, however, is not very transparent.

6. Photon statistics

From $\bar{I}_2(0, s)$ and with Eq. (4) we can calculate $\bar{h}(s)$. Then the Q-factor follows from Eq. (8), with result

$$Q(\infty) = \xi \frac{\Omega_0^2 (1 + |P|^2) \{ (\bar{A} - \lambda) (\eta^2 + \Delta^2) - 4\eta^2 \bar{A} \} + A |P|^2 (3 + |P|^2) \{ \Omega_0^2 (\eta^2 - \Delta^2) - (\eta^2 + \Delta^2)^2 \}}{(\Omega_0^2 \eta + 2\bar{A} (\eta^2 + \Delta^2))^2} + \xi \frac{4 |P|^2 (\eta^2 + \Delta^2)}{\Omega_0^2 \eta (1 + |P|^2) + A |P|^2 (\eta^2 + \Delta^2) (3 + |P|^2)} . \quad (52)$$

For $|P|^2 \rightarrow 0$ this reduces to the Q-factor of a free atom [6].

Close to resonance ($\Delta \rightarrow 0$) and for a small laser linewidth ($\lambda \rightarrow 0$) the Q-factor can be written as

$$Q(\infty) = \frac{\xi}{A} q(x, y) , \quad (53)$$

which depends basically only on the two parameters

$$x = |P|^2, \quad y = \Omega_0^2/A^2. \quad (54)$$

The function $q(x,y)$ is

$$q(x,y) = \frac{x(3+x)(y - \frac{1}{4}(1+x)^2) - \frac{3}{2}y(1+x)^2}{(y + \frac{1}{2}(1+x)^2)^2} + \frac{2x}{y + \frac{1}{2}x(3+x)}. \quad (55)$$

The sign of $q(x,y)$ then determines the regions of sub- and super-poissonian statistics, and this is shown in Fig. 1. It can be checked by inspection that $q(x,y)$ has a factor $1-x$, and therefore we have $q(x,y) = 0$ for $x = 1$, all y . This is the vertical line in Fig. 1. The curve in Fig. 1 gives the second solution of $q(x,y) = 0$. On the y -axis (PC absent) the statistics is always sub-poissonian, and $q(0,y)$ has a minimum of $-\frac{3}{4}$ at $y = \frac{1}{2}$. In absence of the laser (x -axis), $q(x,0)$ decreases monotonically from $\frac{4}{3}$ at $x = 0$ to -1 at $x \rightarrow \infty$. The function $q(x,y)$ is discontinuous at $(x,y) = (0,0)$ and has a saddle point at $(x,y) = (1,2)$.

7. Unit reflectivity

The reflection coefficient $|P|^2$ is proportional to the square of the intensity of the pump lasers of the four-wave mixer, and can therefore have any value. In particular, reflectivities larger than unity have been obtained experimentally [13-15]. An interesting special case is unit reflectivity, for which $|P|^2 = 1$. With Eqs. (47) - (51) we then obtain

$$f_{ee}(\tau) + f_{ge}(\tau) = \xi, \quad (56)$$

$$f_{eg}(\tau) + f_{gg}(\tau) = \xi. \quad (57)$$

These two combinations of correlation functions turn out to be independent of τ . The intensity, Eq. (35), reduces to

$$I = \xi \quad , \quad (58)$$

and the two-photon correlation factors as

$$I_2(0, \tau) = I^2 \quad , \quad (59)$$

for all τ . With Eq. (4) this yields $h(\tau) = 0$, and therefore

$$Q(\tau) = 0 \quad , \quad (60)$$

for all τ and any combination of parameters. In general, it is not necessary that $h(\tau)$ is identically zero in order for the statistics to be poissonian; for $\tau \rightarrow \infty$, only the average of $h(\tau)$, in the sense of Eq. (6), has to vanish. Furthermore, the functions $f_{\beta\alpha}(\tau)$ separately have a non-trivial τ -dependence, as illustrated in Figs. (2) and (3).

8. Conclusions

We have studied the photon correlations and statistics of resonance fluorescence radiation, emitted by an atom near the surface of a PC. The two-time intensity correlation appeared to have four distinct terms, each of which is proportional to a function $f_{\beta\alpha}(\tau)$. These $f_{\beta\alpha}(\tau)$'s were shown to have the significance of the detection rates of β -photons at time τ , after the detection of an α -photon at time zero. From these correlation functions we constructed the long-time Q-factor, as given by Eq. (52). For $\Delta = \lambda = 0$, $Q(\infty)$

could be expressed in a two-parameter function $q(x,y)$, and the conditions for sub-poissonian statistics ($q < 0$) were represented pictorially in Fig. 1. For small reflectivities $|P|^2$ the statistics is always sub-poissonian; for $|P|^2 \rightarrow 1$ the photons have Poisson statistics for any value of the optical parameters; and for large values of $|P|^2$ the statistics becomes again sub-poissonian. It follows from Eq. (52) that for $|P|^2 \rightarrow \infty$ the Q-factor reaches its ultimate lower limit of $Q(\infty) = -\xi/A$.

Photon correlations and statistics of phase-conjugated resonance fluorescence should be amenable to experimental observation. When an atomic beam and a laser beam are projected along the surface of the PC, above which they intersect, then the fluorescence can be detected in a direction perpendicular to the surface. A complication, however, might be the background radiation which is emitted spontaneously in all directions by the nonlinear medium.

Acknowledgments

This research was supported by the Office of Naval Research and the National Science Foundation under Grant CHE-9016789.

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Figure Captions

Fig. 1 Regions of sub-poissonian (-) and super-poissonian (+) statistics for $\Delta = \lambda = 0$ as a function of the phase-conjugate reflectivity x and the relative laser power y . The indicated signs are the signs of the function $q(x,y)$. At $(x,y) = (1,2)$, q has a saddle point. The curved line approaches the asymptotic value of $x = 3$ for $y \rightarrow \infty$, and $q(x,y)$ is negative for all $x > 3$.

Fig. 2 The two curves represent $f_{ee}(\tau)/\xi$ and $f_{eg}(\tau)/\xi$ as a function of $A\tau$ for $\Delta = \lambda = 0$, $|P|^2 = 1$ and $\Omega_0^2 = 2A^2$. These parameters correspond to the saddle point in Fig. 1. The dashed line indicates the value for $A\tau \rightarrow \infty$, which is $\frac{3}{8}$ for both curves. The overshoot in the curve of $f_{eg}(\tau)/\xi$ below the asymptotic limit is a remnant of Rabi oscillations. The Rabi frequency for these values of the parameters is $\frac{1}{2}\sqrt{7}A$.

Fig. 3 Functions $f_{ge}(\tau)/\xi$ and $f_{gg}(\tau)/\xi$ for the same parameters as in Fig. 2. The asymptotic limit here is $\frac{5}{8}$, as shown by the dashed line.

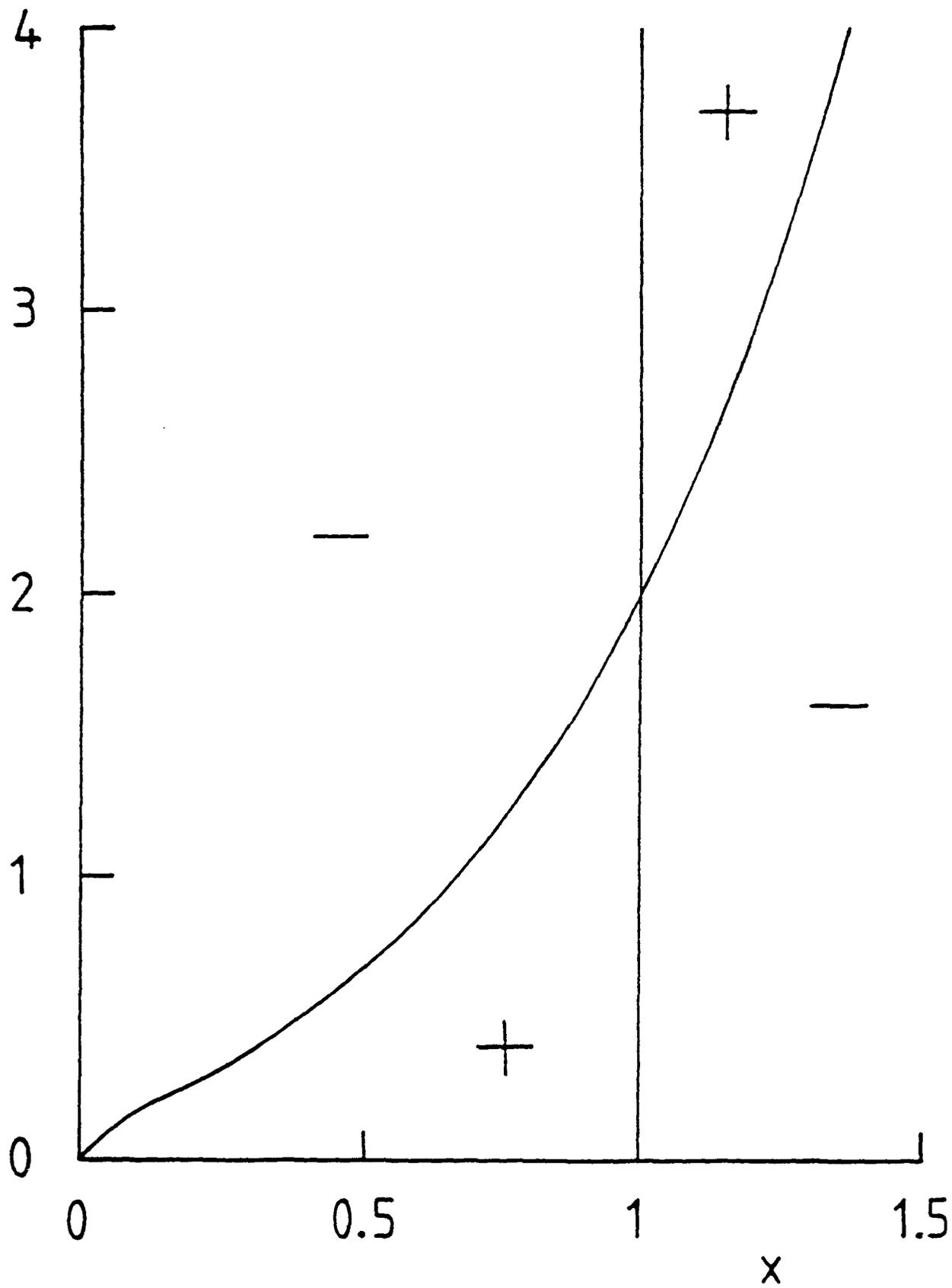


Fig. 1

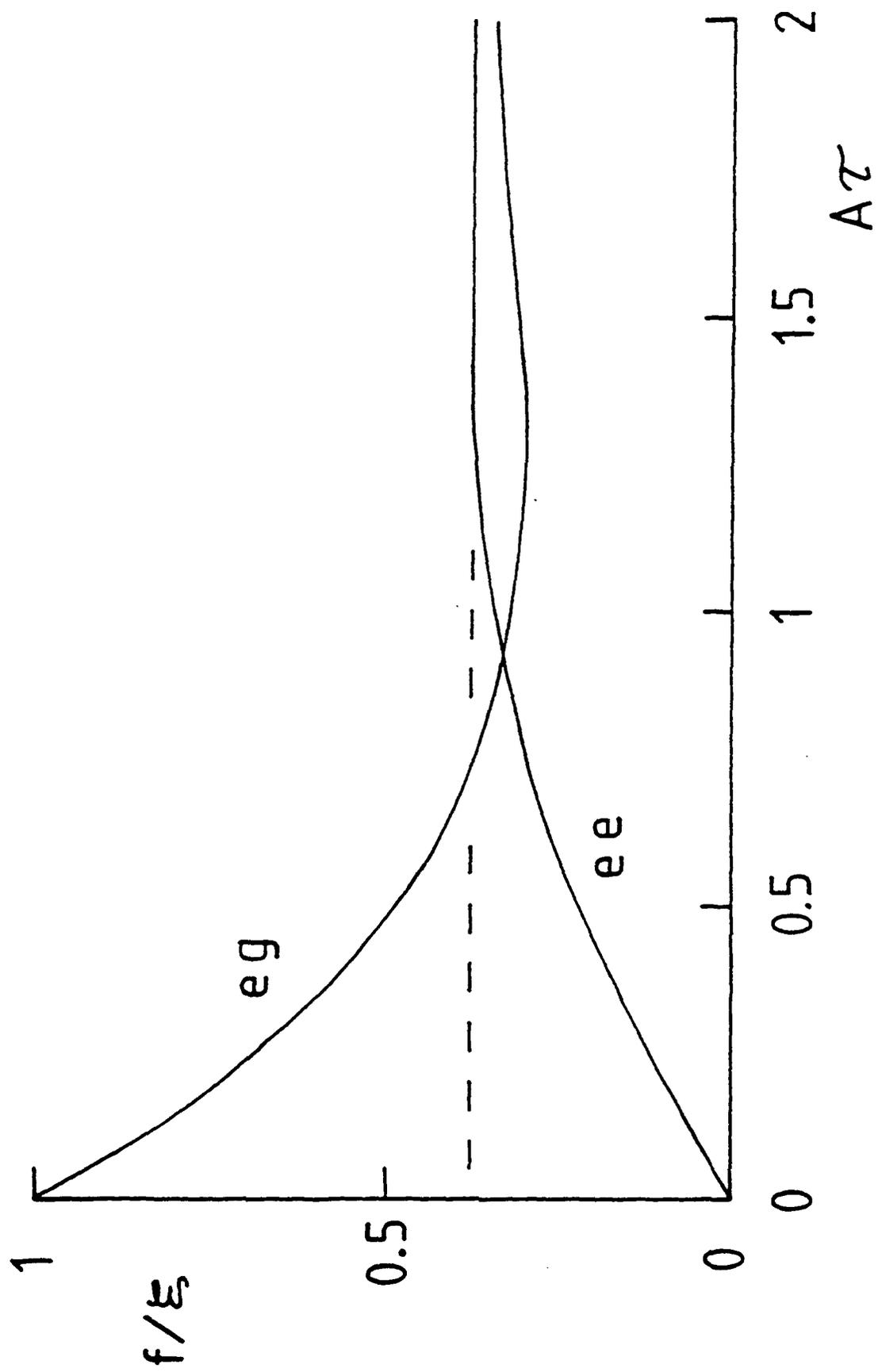


Fig. 2

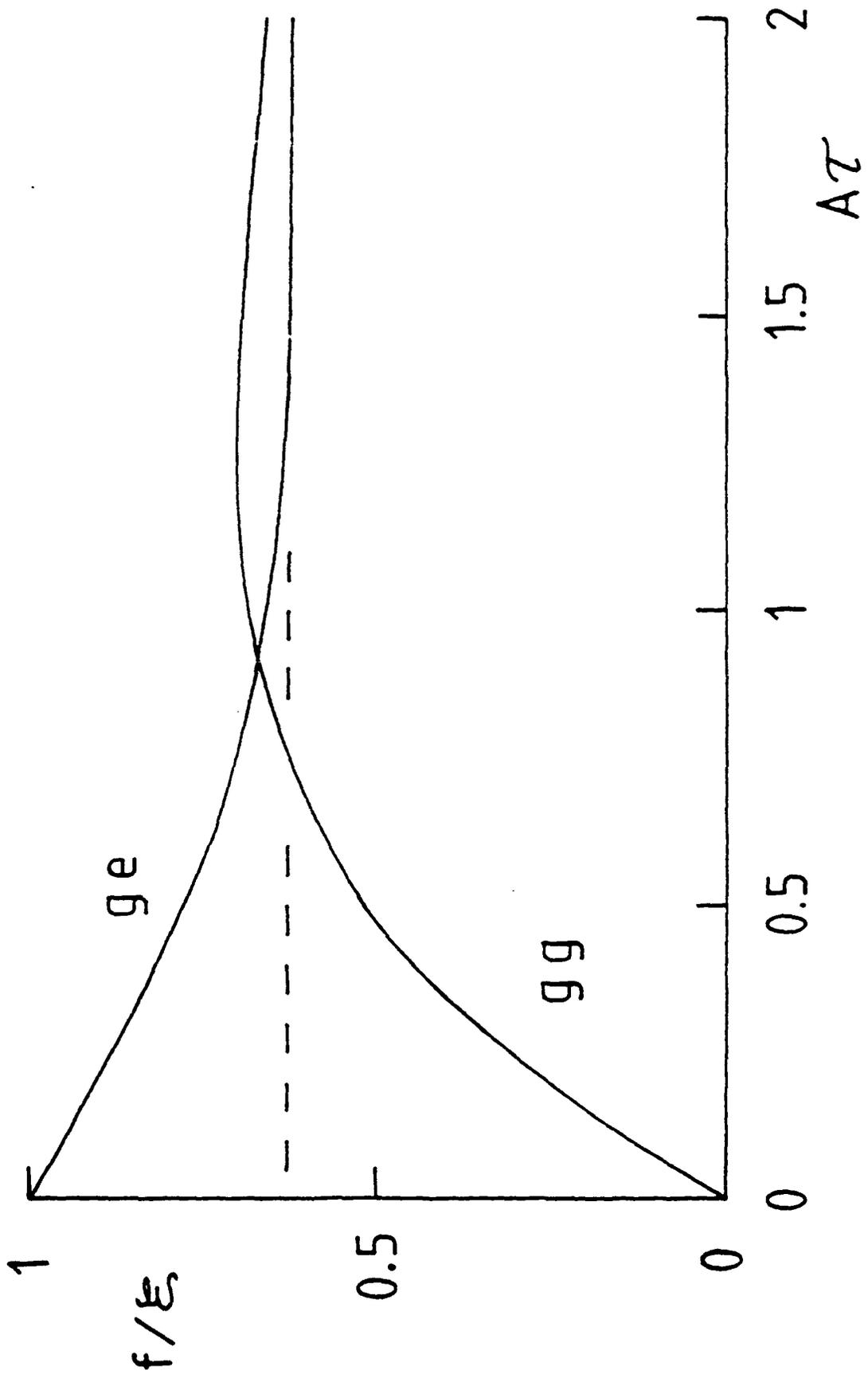


Fig. 3