OPTICA ACTA, 1983, VOL. 30, NO. 11, 1573–1585

Conditions for sub-poissonian photon statistics and squeezed states in resonance fluorescence

H. F. ARNOLDUS and G. NIENHUIS
Fysisch Laboratorium, Rijksuniversiteit Utrecht,
Postbus 80 000, 3508 TA Utrecht, The Netherlands

(*Received 29 April 1983; revision received 22 July 1983*)

Abstract. We study the conditions for sub-poissonian photon statistics and squeezed states in the field of resonance fluorescence of a two-state atom. These conditions as a function of the detuning from resonance, the linewidth and the Rabi frequency have some overlap, but they are largely complementary. Super-poissonian statistics arise for small linewidths and large detunings, irrespective of the Rabi frequency. Squeezed states require small linewidths and either low or moderate Rabi frequencies, or large detunings from resonance.

1. Introduction

The fluorescent radiation field of a two-state atom is known to display two distinct quantum features. Firstly, the fluorescent photons exhibit antibunching in time [1, 2], which makes the steady-state intensity correlation function

$$I_2(t_1, t_2) = I_1 f(t_2 - t_1) \tag{1.1}$$

disappear when the time difference $t_2 - t_1$ approaches zero. This effect, which has been observed experimentally [3, 4], is directly understood as resulting from the fact that immediately after the emission of a photon the atom is bound to be observed in its ground state, and a subsequent photon emission can occur only after a finite recovery time. For large time differences $t_2 - t_1$ the function f approaches the steady-state intensity I_1. As a result of the antibunching property, the quantity

$$Q(t) = \int_0^t dt_1 \int_0^t dt_2 \, [I_2(t_1, t_2) - I_1^2]/I_1 t \tag{1.2}$$

can easily become negative for fluorescence radiation, whereas for a classical stochastic field the quantity Q must obviously be non-negative. Whenever Q is negative, the number of detected photons during the time interval $[0, t]$ has a variance that is smaller than would correspond to a Poisson distribution with the same photon density [5]. In this sense sub-poissonian photon statistics is an essentially quantum-mechanical property of the field.

A second quantum feature of the fluorescence radiation is the occurrence of reduced quantum fluctuations in the in-phase or the out-of-phase component of the fluorescent field with respect to the incident field. As a result of the uncertainty relation for these two components, reductions of the fluctuations in one component can occur only at the expense of increased fluctuations in the other. These squeezed states in the fluorescence field have recently been discussed by Walls and Zoller [6], who pointed out that squeezed states have a negative, normally ordered variance of one of the field components, which means that they have no classical analogue.

In the present paper we evaluate the range of values of the linewidth, the detuning of the incident radiation from resonance and the Rabi frequency for which sub-poissonian photon statistics or a reduction of the quantum fluctuations occurs. We find that these two regions are rather complementary. For a large detection interval t the borderline between sub-poissonian and super-poissonian statistics becomes independent of the Rabi frequency. Squeezed states can occur only when the sum of the collisional linewidth and the bandwidth of the phase-fluctuating incident radiation is smaller than the natural linewidth of the transition. The general condition for the presence of squeezed states turns out to be equivalent to the requirement that the intensity of the coherent Rayleigh line be more than half the total intensity of the fluorescence radiation.

2. Intensity correlation functions

It is well known that the statistics of a photon-counting experiment is determined by the intensity correlation functions of the electromagnetic field [7, 8]. For fluorescent radiation from a two-level atom in a laser field, the positive-frequency part of the electric field is proportional to the atomic lowering operator [2]

$$d^{+}=|g\rangle\langle e|. \tag{2.1}$$

We define the n-fold intensity correlation function in the usual way [9, 10]:

$$I_n(t_1, t_2, \ldots, t_n)=A^n\langle d(t_1)d(t_2)\ldots d(t_n)d^{+}(t_n)\ldots d^{+}(t_1)\rangle \tag{2.2}$$

for $t_n \geqslant t_{n-1} \geqslant \ldots \geqslant t_1$. This correlation function has the physical significance that $I_n\, dt_1 \ldots dt_n$ is the probability that the atom emits a photon in the time interval $[t_1, t_1+dt_1]$, ... and a photon in the time interval $[t_n, t_n+dt_n]$, irrespective of what happens at other times. Hence the intensity is expressed as the number of photons emitted per unit time, and A is the Einstein coefficient for spontaneous emission. The time evolution of the Heisenberg operator $d(t)$ is determined by the propagator of the density matrix of the atom in the field, which is written as a Liouville operator

$$\rho(t)=T(t, t')\rho(t'). \tag{2.3}$$

We can evaluate the expression (2.2) for the correlation function

$$I_n(t_1, \ldots, t_n)=A^n \operatorname{Tr} RT(t_n, t_{n-1})R \ldots RT(t_1, 0)\rho(0). \tag{2.4}$$

The operator R is defined as

$$R\rho=d^{+}\rho d=P_g\langle e|\rho|e\rangle \tag{2.5}$$

in terms of the projector $P_g=|g\rangle\langle g|$ on the atomic ground state.

In order to proceed with the evaluation of I_n we have to consider the equation of motion of $\rho(t)$ in detail. We describe the incident laser field as a classical field with a stochastically fluctuating phase, which can account for the finite bandwidth

$$\mathbf{E}(t)=E_0 \operatorname{Re} \boldsymbol{\varepsilon}_{\mathrm{L}} \exp[-i(\omega_{\mathrm{L}} t+\psi(t))]. \tag{2.6}$$

The random phase $\psi(t)$ gives rise to a bandwidth λ around the central frequency ω_{L}.

The interaction with the atom in the dipole and the rotating-wave approximation is

$$H_{\mathrm{ar}}(t) = -\tfrac{1}{2}\hbar\Omega d \exp[-i(\omega_{\mathrm{L}} t + \psi(t))] + \text{hermitian conjugate}, \tag{2.7}$$

which contains the Rabi frequency $\Omega = E_0 \boldsymbol{\mu}_{eg} \cdot \boldsymbol{\varepsilon}_{\mathrm{L}}/\hbar$, with $\boldsymbol{\mu}_{eg}$ the dipole matrix element of the transition. Spontaneous decay and collisional relaxation in the impact limit are included by the effective operators [11]

$$\left.\begin{aligned} \Gamma\rho &= \tfrac{1}{2}A(P_e\rho + \rho P_e - 2R\rho), \\ \Phi\rho &= (\gamma + i\beta)P_e\rho P_g + (\gamma - i\beta)P_g\rho P_e, \end{aligned}\right\} \tag{2.8}$$

with γ and β the collisional width and shift. From equation (2.7), one notes that the hamiltonian is time-dependent and stochastic. We can eliminate the rapid oscillations by a unitary stochastic transformation in Liouville space [12], and introduce the transformed density matrix

$$\sigma(t) = \exp[i(\omega_{\mathrm{L}} t + \psi(t))K]\rho(t), \tag{2.9}$$

with

$$K\rho = [P_e, \rho]. \tag{2.10}$$

The transformed density matrix σ obeys the stochastic differential equation

$$i\frac{d}{dt}\sigma(t) = [L_{\mathrm{d}} - i\Gamma - i\Phi - \dot{\psi}(t)K]\sigma(t), \tag{2.11}$$

where

$$L_{\mathrm{d}}\sigma = [H_{\mathrm{d}}, \sigma]. \tag{2.12}$$

The operator H_{d} is the dressed-atom hamiltonian

$$H_{\mathrm{d}} = -\tfrac{1}{2}\hbar[\Delta(P_e - P_g) + \Omega(d + d^+)] \tag{2.13}$$

in terms of the Rabi frequency Ω and the detuning from resonance $\Delta = \omega_{\mathrm{L}} - \omega_0$.

The evolution of $\sigma(t)$ is written as

$$\sigma(t) = U(t, t')\sigma(t') \tag{2.14}$$

and the connection with the propagator of $\rho(t)$ is given by

$$T(t, t')\rho(t') = \exp[-i(\omega_{\mathrm{L}} t + \psi(t))K]U(t, t')\sigma(t'). \tag{2.15}$$

From the definition of K and R it then follows that

$$RT(t, t')\rho(t') = RU(t, t')\sigma(t'), \tag{2.16}$$

which yields the expression for the correlation function

$$I_n(t_1, \ldots, t_n) = A^n \operatorname{Tr} RU(t_n, t_{n-1})R \ldots RU(t_1, 0)\sigma(0). \tag{2.17}$$

The correlation function I_n is still a stochastic quantity. From a previous paper [12], we know how to deal with the stochastic averaging of equation (2.11) when the phase ψ can be treated as a process with independent increments [13]. The resulting equation of motion for the stochastically averaged density matrix is

$$i\frac{d}{dt}\bar{\sigma}(t) = (L_{\mathrm{d}} - i\Gamma - i\Phi - i\lambda K^2)\bar{\sigma}(t), \tag{2.18}$$

which contains an effective relaxation operator λK^2 arising from the phase fluctuations. The corresponding propagator for the averaged density matrix is given by

$$\bar{\sigma}(t) = \bar{U}(t-t')\bar{\sigma}(t'). \tag{2.19}$$

The t_n dependence of $U(t_n, t_{n-1})$ is governed by a similar equation to (2.11), but with a modified initial condition $U(t_{n-1}, t_{n-1}) = 1$. We can average the right-hand side of equation (2.17) over the phase fluctuations during the interval $[t_{n-1}, t_n]$. Then we can average the corresponding initial condition over the fluctuations during the preceding interval $[t_{n-2}, t_{n-1}]$, etc. Furthermore, we assume that the atom is in its steady state in the radiation field, which means that we can write

$$\bar{U}(t_1)\bar{\sigma}(0) = \bar{\sigma}(0) = \bar{\sigma}_0, \tag{2.20}$$

where $\bar{\sigma}_0$ is the stochastically averaged steady-state density matrix of the atom, defined by the equation

$$(L_d - i\Gamma - i\Phi - i\lambda K^2)\bar{\sigma}_0 = 0. \tag{2.21}$$

Our final result for the n-fold correlation function is

$$\bar{I}_n(t_1, \ldots, t_n) = A^n \operatorname{Tr} R\bar{U}(t_n - t_{n-1})R \ldots R\bar{U}(t_2 - t_1)R\bar{\sigma}_0. \tag{2.22}$$

For the last step in the evaluation of $\bar{I}_n$, we note that

$$R\sigma = P_g \operatorname{Tr} P_e \sigma, \tag{2.23}$$

so that every operator $\bar{U}$ in equation (2.22) acts on P_g only. With the definition

$$f(t) = A \operatorname{Tr} P_e \bar{U}(t) P_g, \tag{2.24}$$

we finally have the result for the correlation functions:

$$\bar{I}_n(t_1, \ldots, t_n) = f(t_n - t_{n-1}) f(t_{n-1} - t_{n-2}) \ldots f(t_2 - t_1)\bar{I}_1 \tag{2.25}$$

for $t_n \geqslant t_{n-1} \geqslant \ldots \geqslant t_1$, and where

$$\bar{I}_1 = A n_e, \tag{2.26}$$

with $n_e = \langle e|\bar{\sigma}_0|e\rangle$ the average population of the excited state. A similar result for free atoms in a monochromatic radiation field has been obtained by several authors [5, 9, 10, 14].

From (2.24) it is obvious that $f(t)$ is the emitted intensity at time t, with the condition that there was an emission at time $t = 0$. Hence

$$f(0) = 0, \tag{2.27}$$

which causes each n-fold intensity correlation function to vanish when two time arguments approach each other. This reflects the antibunching property that two subsequently emitted photons tend to be separated in time [1].

3. Photon statistics

The statistics of the photons counted by a detector in a given time interval $[0, t]$ can be derived entirely from the distribution functions C_m of detection times. These distribution functions are defined by requiring that

$$C_m(t_1, \ldots, t_m)\, dt_1 dt_2 \ldots dt_m$$

is the probability of detecting a photon in the time interval $[t_1, t_1+dt_1], \ldots$ and a photon in the time interval $[t_m, t_m+dt_m]$. We introduce the probability $\alpha\,(0 \leqslant \alpha \leqslant 1)$ that an emitted photon is seen by the detector. Then it is rather obvious that the distribution functions C_m are related in a very simple fashion to the correlation functions $\bar{I}_n$, according to

$$C_m(t_1, \ldots, t_m) = \alpha^m \bar{I}_m(t_1, \ldots, t_m). \tag{3.1}$$

It is a standard problem in stochastic theory to derive from the distribution functions C_m the probability distribution $p_n(t)$ for detecting precisely n photons during the interval $[0, t]$. The factorial moments

$$s_m(t) = \langle n!/(n-m)! \rangle \tag{3.2}$$

are directly related to the distribution functions C_m by the integral relation [13, 5, 14]

$$s_m(t) = m! \int_0^t dt_m \int_0^{t_m} dt_{m-1} \ldots \int_0^{t_2} dt_1\, C_m(t_1, \ldots, t_m). \tag{3.3}$$

The probability distribution $p_n(t)$ is related to the factorial moments by the equations [5]

$$p_n(t) = \frac{1}{n!} \sum_{k=0}^{\infty} \frac{(-1)^k}{k!} s_{n+k}(t), \tag{3.4}$$

as can be checked by substitution of equation (3.2). When $\alpha = 1$, each emitted photon leads to a count in the detector, and then the resulting distribution $p_n(t)$ refers to the statistics of emitted photons. In reality the value of α will be diminished, as a result of both the finite aperture angle of the detector and its quantum efficiency.

Since we know the intensity correlation functions $\bar{I}_n$ explicitly from equation (2.25), we can evaluate the factorial moments s_m and the photon number probabilities p_n directly. We adopt a Laplace transformation, and we introduce

$$\tilde{p}_n(v) = \int_0^{\infty} dt \exp(-vt) p_n(t), \tag{3.5}$$

with $\mathrm{Re}\, v > 0$, and likewise we define the Laplace transform $\tilde{s}_m(v)$ of $s_m(t)$. Explicit expressions for $\tilde{s}_m$ result from equation (2.25), and we find that

$$\begin{aligned} &\tilde{s}_0(v) = 1/v \\ &\tilde{s}_m(v) = m!(\alpha \tilde{f}(v))^{m-1} \alpha A n_e / v^2 \quad \text{if} \quad m \geqslant 1. \end{aligned} \tag{3.6}$$

Substituting this result in equation (3.4), we obtain

$$\begin{aligned} &\tilde{p}_0(v) = 1/v - \alpha A n_e / v^2 (1 + \alpha \tilde{f}(v)), \\ &\tilde{p}_n(v) = \alpha A n_e [\alpha \tilde{f}(v)]^{n-1} / v^2 (1 + \alpha \tilde{f}(v))^{n+1} \quad \text{if} \quad n \geqslant 1. \end{aligned} \tag{3.7}$$

An explicit expression for the Laplace transform $\tilde{f}(v)$ results from equations (2.24), (2.18) and (2.19). The formal result

$$\tilde{f}(v) = A \,\mathrm{Tr}\, P_e (v + \Gamma + \Phi + \lambda K^2 + i L_d)^{-1} P_g \tag{3.8}$$

allows direct evaluation, with the result

$$\bar{f}(v)=A\Omega^2(v+\tfrac{1}{2}A+\gamma+\lambda)/2vD(v), \tag{3.9}$$

with

$$D(v)=(v+A)[(v+\tfrac{1}{2}A+\gamma+\lambda)^2+(\Delta-\beta)^2]+\Omega^2(v+\tfrac{1}{2}A+\gamma+\lambda). \tag{3.10}$$

One notes that $f(t)$ is simply proportional to the twofold intensity correlation function $\bar{I}_2(t_0, t_0+t)$, which has been studied by several workers [1, 2, 9, 15]. The long-time limit of $f(t)$ obeys the equality

$$\lim_{t\to\infty} f(t)=\lim_{v\downarrow 0} v\bar{f}(v)=An_e, \tag{3.11}$$

so that

$$n_e=\Omega^2(\tfrac{1}{2}A+\gamma+\lambda)/2D(0). \tag{3.12}$$

In figures 1 and 2 $f(t)$ is plotted for some representative examples. In the special case of free atoms in a monochromatic radiation field at resonance ($\gamma=\lambda=\Delta=0$), equation (3.9) and its inverse Laplace transform coincide with the result obtained by Mandel [5].

Following Mandel [5], we introduce the normalized correlation function $\kappa(t)$ by the defining relation

$$f(t)=An_e(1-\kappa(t)), \tag{3.13}$$

so that $\kappa(0)=1$ and $\kappa(\infty)=0$. When κ would be zero for all times, the distribution functions C_m would factorize into C_1^m, indicating that subsequent photons would arrive in a totally uncorrelated fashion. Likewise the factorial moments s_m would factorize according to the relation $s_m=s_1^m$, and the photon statistics would be

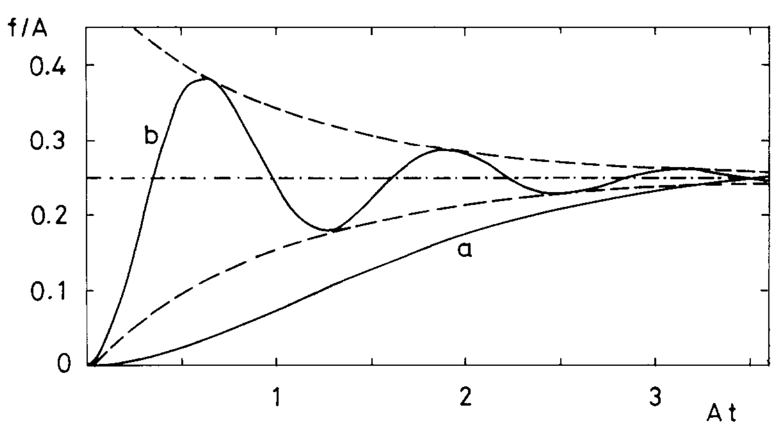

Figure 1. Behaviour of the intensity correlation function as expressed by the function $f(t)$, defined by equation (2.24). Curve (a) corresponds to the values $\eta=\frac{1}{2}$, $\delta=0$ and $\zeta^2=\frac{1}{2}$, and gives rise to the minimum value of $\bar{Q}$ (equation (3.18)), and thereby to the strongest sub-poissonian statistics. Curve (b) corresponds to the values $\eta=1$, $\delta^2=12$ and $\zeta^2=13$, which gives rise to the same asymptotic value $f(\infty)=\frac{1}{4}$.

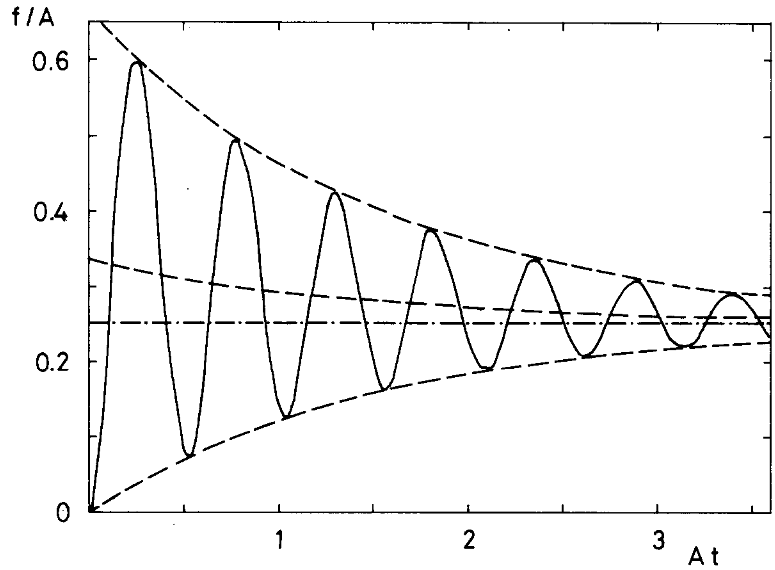

Figure 2. Behaviour of the function $f(t)$ (equation (2.24)) for the values $\delta^2=48$, $\zeta^2=96$ and $\eta=\frac{1}{2}$. The average of the upper and lower envelopes of the rapid oscillations lies above the asymptotic value $f(\infty)=\frac{1}{4}$, which gives rise to a positive value of $\bar{Q}$, and hence to super-poissonian statistics.

poissonian. As a measure of the deviation from poissonian statistics, Mandel [5] introduced the quantity

$$Q(t)=(s_2(t)-s_1(t)^2)/s_1(t), \tag{3.14}$$

which is equivalent to equation (1.2). If we substitute equation (3.3) in equation (3.14), we obtain

$$Q(t)=-2\alpha A n_e \int_0^t d\tau\,(t-\tau)\,\kappa(\tau)/t. \tag{3.15}$$

Mandel [5] has demonstrated that Q is a negative quantity for monochromatic radiation on resonance, which means that the variance of the detected number of photons is smaller than it would be for Poisson statistics with the same photon density. Here we wish to point out that sub-poissonian statistics is not a necessary consequence of the antibunching property of fluorescence radiation. In fact, for large detection times t the quantity Q approaches the value

$$\bar{Q}=-2\alpha A n_e \tilde{\kappa}(0). \tag{3.16}$$

For convenience we express the linewidth, the Rabi frequency and the detuning as dimensionless quantities by introducing

$$\eta=(\tfrac{1}{2}A+\gamma+\lambda)/A, \quad \zeta=\Omega/A, \quad \delta=(\Delta-\beta)/A. \tag{3.17}$$

From the explicit result (3.9) for $\tilde{f}(v)$, we derive

$$\bar{Q}=\alpha\zeta^2\,\frac{\delta^2(1-\eta)-\eta^2(1+\eta)}{(\eta^2+\eta\zeta^2+\delta^2)^2}. \tag{3.18}$$

For $\delta=0$, this is always a negative quantity. Its minimal value occurs at $\eta=\frac{1}{2}$, $\delta=0$, $\zeta^2=\frac{1}{2}$, and it takes the value $\bar{Q}_{\min}=-3\alpha/4$. Naturally, a small detection efficiency tends to diminish the correlation between the few photons that are detected, and the distribution becomes more poissonian. For values of the detuning obeying

$$(1-\eta)\delta^2>\eta^2(1+\eta), \tag{3.19}$$

$\bar{Q}$ is larger than corresponding to a Poisson distribution, and we obtain super-poissonian statistics, even though the antibunching property of the photons is preserved. This may be understood by noting that for large detunings and not too large linewidths the behaviour of $f(t)$ for small times is governed by rapid optical nutations [16], which tend to make f larger, on average, than its large-time limit An_e. For these intermediate times the function κ is negative, which leads to an effective bunching of subsequent photons. This behaviour of $f(t)$ is illustrated in figure 2. The regions of sub-poissonian and super-poissonian statistics in the η–δ plane are indicated in figure 3. Super-poissonian statistics can only occur when $\eta<1$, which means that the sum of the collisional linewidth γ and the laser bandwidth λ is smaller than the natural linewidth $\frac{1}{2}A$. It is remarkable that these regions do not depend on the Rabi frequency.

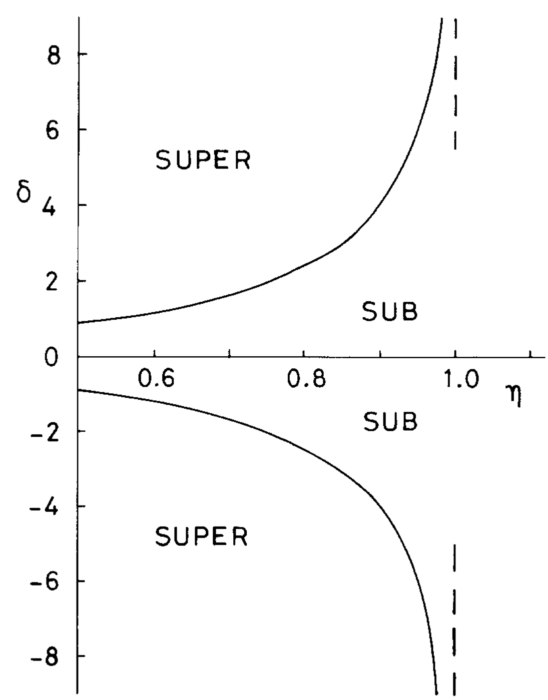

Figure 3. Regions in the η–δ plane where $\bar{Q}$ (equation (3.18)) is positive or negative, corresponding to super- or sub-poissonian photon statistics. The regions are found to be independent of the reduced Rabi frequency ζ.

Recently, Singh [17] has shown that super-poissonian photon statistics can occur for zero detuning ($\delta=0$) in a limited range of observation times t when the atom is prepared in its excited state at the beginning of the observation interval. He also demonstrated that a finite bandwidth can make the photon-number distribution narrower. The latter conclusion can also be drawn from our results, which are valid in the steady state and in the limit of a large observation interval, and which allow a detuning from resonance.

4. Squeezed states

As pointed out in § 2, the statistical properties of the fluorescent radiation field are directly related to the statistical properties of the atomic dipole operator. Therefore the occurrence of squeezed states in the fluorescence can be studied by examining squeezed states in the atom in the driving field [6, 18]. We introduce the Pauli operators, with their usual definition

$$S_x=d+d^+, \quad S_y=-id+id^+, \quad S_z=P_e-P_g. \tag{4.1}$$

In the stochastically rotating frame, where the density matrix σ obeys the Liouville equation (2.11), the operator S_x is proportional to the in-phase component of the fluorescence field, whereas S_y is proportional to the out-of-phase component. These two operators obey the relations

$$[S_x, S_y]=2iS_z, \quad S_x^2=S_y^2=1. \tag{4.2}$$

Hence their variances are

$$\langle \Delta S_x^2\rangle = 1-P_x^2, \quad \langle \Delta S_y^2\rangle = 1-P_y^2, \tag{4.3}$$

where we use the expansion

$$\bar{\sigma}_0=\tfrac{1}{2}[1+\mathbf{P}\cdot\mathbf{S}] \tag{4.4}$$

of the stochastically averaged steady-state density matrix. This expansion is equivalent to the vector equality

$$\langle \mathbf{S}\rangle = \mathbf{P}. \tag{4.5}$$

From the uncertainty relation

$$\langle \Delta S_x^2\rangle\langle \Delta S_y^2\rangle \geqslant \langle S_z\rangle^2, \tag{4.6}$$

it follows that squeezing occurs in S_x when [6, 17]

$$1-P_x^2<|P_z|, \tag{4.7}$$

and then squeezing is also present in the in-phase component of the fluorescence field. Likewise, squeezing in the out-of-phase (absorptive) component requires that

$$1-P_y^2<|P_z|. \tag{4.8}$$

The vector $\mathbf{P}$ can be regarded as the polarization vector of a fictitious spin $\frac{1}{2}$ that models the two-state atom; the magnitude of this vector cannot be greater than 1.

The conditions (4.7) and (4.8) are readily generalized to arbitrary linear combinations

$$S=aS_x+bS_y, \tag{4.9}$$

with a and b two real numbers obeying the equality $a^2+b^2=1$. The condition for squeezing in this component (4.9), or, equivalently, in the corresponding phase component of the fluorescence, is found directly to be

$$1-(aP_x+bP_y)^2<|P_z|. \tag{4.10}$$

As a quantitative measure determining the amount of squeezing, we introduce the quantity

$$R=[1-(aP_x+bP_y)^2-|P_z|]/|P_z|. \tag{4.11}$$

Like the quantity Q introduced in equation (3.14), R can never be smaller than -1. The condition for squeezing is that R must be negative. For a given density matrix $\bar{\sigma}_0$, or for a given polarization vector $\mathbf{P}$, the minimum value of R occurs in the phase component obeying $a/b=P_x/P_y$, so that the unit vector (a,b) is directed along the projection of $\mathbf{P}$ on to the x–y plane. For this phase component, R is given by the relation

$$R=(1-P_x^2-P_y^2-|P_z|)/|P_z|. \tag{4.12}$$

For a given density matrix $\bar{\sigma}_0$, this quantity is negative when $|P_z|<1-P_x^2-P_y^2$. The minimal value $R=-1$ is attained when the polarization vector $\mathbf{P}$ has its maximum length 1 (so that the atom is in a pure state) and lies in the x–y plane.

In our special case of the atom in a radiation field, the steady-state density matrix is determined by equation (2.21). The resulting expression for the polarization vector is

$$\mathbf{P}=\frac{-1}{\eta^2+\delta^2+\eta\zeta^2}\begin{pmatrix}\zeta\delta\\ \zeta\eta\\ \eta^2+\delta^2\end{pmatrix}. \tag{4.13}$$

By substituting equation (4.13) into equations (4.7) and (4.8), one recovers the conditions of Walls and Zoller [6] in the special case of free atoms in a monochromatic field ($\eta=\frac{1}{2}$). If we substitute equation (4.13) into equation (4.11), we obtain

$$R=\zeta^2[\eta^2\zeta^2+(\eta-1)(\eta^2+\delta^2)]/(\eta^2+\delta^2)(\eta^2+\delta^2+\eta\zeta^2). \tag{4.14}$$

Squeezed states occur when this quantity is negative, which happens when

$$(1-\eta)\delta^2>\eta^2(\zeta^2-1+\eta). \tag{4.15}$$

The regions in the η–δ plane where squeezed states are possible are indicated in figure 4 for various values of ζ. For $\zeta^2=2$, this region coincides exactly with the region indicated by equation (3.19), where super-poissonian statistics prevail. One notes that the regions of sub-poissonian statistics and the regions of squeezing are rather complementary, even though both phenomena are recognized as essentially quantum-mechanical.

The strongest reduction of quantum fluctuations occurs at the minimum value of R,

$$R_{\min}=-(\sqrt{2}-1)^2=-0{\cdot}1716, \tag{4.16}$$

and it is attained for

$$\eta=\tfrac{1}{2},\quad \delta=0,\quad \zeta^2=\tfrac{1}{2}(\sqrt{2}-1). \tag{4.17}$$

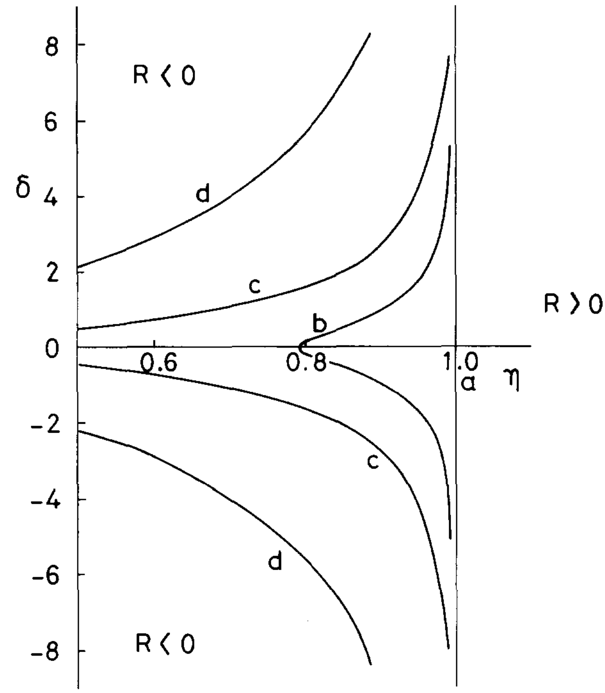

Figure 4. Regions in the η–δ plane where R (equation (4.14)) is positive or negative, for various values of the reduced Rabi frequency ζ. Only the boundaries between the regions are indicated. The relevant ζ values are (*a*) $\zeta^2 \downarrow 0$, (*b*) $\zeta^2 = 0{\cdot}207$, (*c*) $\zeta^2 = 1$ and (*d*) $\zeta^2 = 10$. Curve (*b*) corresponds to the case where the absolute minimum value of R occurs, as indicated by equations (4.16) and (4.17).

There is a direct physical significance in the components of the polarization vector $\mathbf{P}$ which occurs in equation (4.12). The total fluorescent intensity as given in equation (2.26) obeys the relation

$$\bar{I}_1 = \tfrac{1}{2}A(1+P_z). \tag{4.18}$$

The coherent Rayleigh line has a strength [19, 12]

$$\bar{I}_{\mathrm{R,c}} = \tfrac{1}{4}A(P_x^2 + P_y^2). \tag{4.19}$$

Since P_z will always be negative for a two-state atom, the quantity R as given by equation (4.12) (or (4.14)) can be put in the form

$$R = (2\bar{I}_1 - 4\bar{I}_{\mathrm{R,c}})/(A - 2\bar{I}_1). \tag{4.20}$$

Hence the condition for squeezed states in resonance fluorescence is summarized by the requirement that the coherent Rayleigh line contain more than half the total intensity.

5. Conclusions

We have studied the quantity Q, defined by equation (3.14), which is a quantitative measure of the deviation from Poisson statistics of the number of fluorescent photons detected in a time interval. An explicit expression for Q in the limit of large counting intervals is given by equation (3.18). The region of sub-poissonian statistics is found to be independent of the value of the reduced Rabi frequency ζ. Super-poissonian statistics can occur for small collisional linewidths and a small bandwidth of the incident radiation ($\eta < 1$), and large detunings, as indicated in figure 3. Hence sub-poissonian statistics is not a necessary consequence of the antibunching property of the photons for not too small detection intervals. In the limit of very small detection intervals, the antibunching property always makes Q non-positive, which gives sub-poissonian statistics. Figure 2 shows an example of a reduced intensity correlation function, which exhibits antibunching as expected. However, when the rapid oscillations are smeared out, the resulting average value lies above the long-time limit. In this sense the photons show here an average bunching behaviour.

The quantity Q can never be smaller than -1 for any radiation field, and negative Q values, or equivalently sub-poissonian statistics, cannot be produced by a classical field [5]. We have introduced an analogous quantity R (equation (4.12)) as a quantitative measure of the minimum quantum fluctuations in the components of the oscillating dipole or the fluorescence field. Negative R values correspond to squeezed states, which do not have a classical analogue [6, 17], and R can never be smaller than -1 for any state of the radiating atom. For an atom in a driving field, R is given by equation (4.14). The regions where R is negative are illustrated in figure 4; the minimum value is $-0{\cdot}1716$. In general, the regions where squeezed states are possible are quite different from the regions where the photon statistics have a sub-poissonian character. For $\zeta^2 = 2$, the two regions in the η–δ plane are exactly complementary.

We conclude that antibunching, sub-poissonian photon statistics and squeezing reflect quite different properties of the fluorescence field, even though each of these characteristics is essentially quantum-mechanical in origin.

Appendix

In equation (3.7) we give an expression for (the Laplace transform of) the photon-number probability distribution p_n as a function of the detection probability α. The probabilities p_n are not independent, and we can obtain p_n with $n \geqslant 1$ from the probability p_0 as a function of α. Moreover, this relation is not restricted to fluorescent photons. We shall prove these statements in this appendix.

It is often convenient to describe number statistics by a generating function [13, 14]

$$G(\mu, \alpha) = \langle (1-\mu)^n \rangle = \sum_{n=0}^{\infty} p_n(\alpha)(1-\mu)^n. \tag{A 1}$$

(We suppress the dependence on the time interval t in this appendix, but indicate explicitly the dependence on the parameter α.) Hence the probabilities p_n are directly proportional to the coefficients in the Taylor expansion of $G(\mu, \alpha)$ around $\mu = 1$. On the other hand, by differentiating equation (A 1) m times and setting $\mu = 0$, we obtain the factorial moments $s_m(\alpha)$, apart from a sign. This leads to the expansion

$$G(\mu, \alpha) = \sum_{m=0}^{\infty} \frac{(-\mu)^m}{m!} s_m(\alpha). \tag{A 2}$$

Since $s_m(\alpha)$ is proportional to α^m, according to equations (3.3) and (3.1), we find that $G(\mu, \alpha)$ depends only on the product $\alpha\mu$, but not on μ and α separately. Hence we may define a function g that obeys the relation

$$G(\mu, \alpha) = g(\alpha\mu). \tag{A 3}$$

If we set $\mu = 1$ in equation (A 1), we find that

$$g(\alpha) = p_0(\alpha), \tag{A 4}$$

so that

$$G(\mu, \alpha) = p_0(\alpha\mu). \tag{A 5}$$

We conclude that the function $g(\alpha)$, and thereby the generating function $G(\mu, \alpha)$ can be determined experimentally, simply by measuring the probability for detecting zero photons, as a function of the detection probability α. According to equation (A 1), the probabilities p_n are found by taking the nth derivative of $G(\mu, \alpha)$, and then setting $\mu = 1$. If we substitute equation (A 5), we obtain the general relation

$$p_n(\alpha) = \frac{(-\alpha)^n}{n!}\left(\frac{\partial}{\partial \alpha}\right)^n p_0(\alpha), \tag{A 6}$$

which allows one to evaluate $p_n(\alpha)$ as soon as $p_0(\alpha)$ is known. It will be obvious that the validity of the result (A 6) does not depend on the particular shape of the correlation functions $\bar{I}_n$, but only on the fact that the distribution functions C_m—and thereby the factorial moments—are simply proportional to α^m.

In our special case of fluorescent photons, the validity of equation (A 6) can be checked directly for equation (3.7).

On étudie les conditions pour que les statistiques de photons soient sous-poissonniennes et les états serrés dans le champ de la fluorescence résonnante d'un atome à deux états. Ces conditions en fonction du désaccord par rapport à la résonance, de la largeur de raie et de la fréquence de Rabi se recouvrent partiellement, mais elles sont largement complémentaires. Les statistiques superpoissonniennes se produisent pour de faibles largeurs de raie et de grands désaccords, quelque soit la fréquence de Rabi. Les états serrés requièrent de faibles largeurs de raie et des fréquences de Rabi basses ou modérées, ou encore de grands désaccords par rapport à la résonnance.

Wir untersuchen die Bedingungen für Sub-Poisson-Photonenstatistik und für 'squeezed' Zustände im Resonanzfluoreszenzfeld eines Atoms mit zwei Zuständen. Diese Bedingungen haben als Funktion der Verstimmung aus der Resonanz, der Linienbreite und der Rabi-Frequenz einen gewissen Überlapp, sind aber sonst weitgehend komplementär. Super-Poisson-Statistik tritt auf bei kleinen Linienbreiten und starken Verstimmungen, ungeachtet der Rabi-Frequenz 'Squeezed' Zustände erfordern kleine Linienbreiten und entweder niedrige bis mäßige Rabi-Frequenzen oder große Verstimmungen aus der Resonanz.

References

[1] Carmichael, H. J., and Walls, D. F., 1976, *J. Phys.* B, **9**, 1199.
[2] Kimble, H. J., and Mandel, L., 1976, *Phys. Rev.* A, **13**, 2123.
[3] Kimble, H. J., Dagenais, M., and Mandel, L., 1978, *Phys. Rev.* A, **18**, 201.
[4] Dagenais, M., and Mandel, L., 1978, *Phys. Rev.* A, **18**, 2217.
[5] Mandel, L., 1979, *Optics Lett.*, **4**, 205.
[6] Walls, D. F., and Zoller, P., 1981, *Phys. Rev. Lett.*, **47**, 709.
[7] Glauber, R. J., 1965, *Quantum Optics and Electronics*, edited by C. DeWitt, A. Blandin and C. Cohen-Tannoudji (New York: Gordon & Breach), p. 63.

[8] KELLEY, P. L., and KLEINER, W. H., 1964, *Phys. Rev.*, **136,** A316.
[9] GEORGES, A. T., 1981, *Optics Commun.*, **38,** 274.
[10] AGARWAL, G. S., 1977, *Phys. Rev.* A, **15,** 814.
[11] BREENE, R. G., JR., 1981, *Theories of Spectral Line Shape* (New York: Wiley–Interscience).
[12] ARNOLDUS, H. F., and NIENHUIS, G., 1983, *J. Phys.* B, **16,** 2325.
[13] VAN KAMPEN, N. G., 1981, *Stochastic Processes in Physics and Chemistry* (Amsterdam: North-Holland).
[14] LENSTRA, D., 1982, *Phys. Rev.* A, **26,** 3369.
[15] WÓDKIEWICZ, K., 1980, *Physics Lett.* A, **77,** 315.
[16] ALLEN, L., and EBERLY, J. H., 1975, *Optical Resonance and Two-Level Atoms* (New York: Wiley).
[17] SINGH, S., 1983, *Optics Commun.*, **44,** 254.
[18] MANDEL, L., 1982, *Phys. Rev. Lett.*, **49,** 136.
[19] NIENHUIS, G., 1982, *J. Phys.* B, **15,** 535.