Phil. Trans. R. Soc. Lond. A **293**, 223–237 (1979)
Printed in Great Britain

Atoms in strong light-fields: photon antibunching in single atom fluorescence

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Some general remarks and suggestions concerning photon antibunching in single atom fluorescence are presented. The close connection between this antibunching effect and the 'reduction of the wave packet' due to the detection process is made explicit. It is pointed out that polarization effects could considerably change the shape of the signals. A dressed atom approach to these problems reveals analogies with quantum beats and suggests the use of frequency filters at the detection, selecting one component of the fluorescence spectrum and leading to new types of photon correlation signals.

1. Introduction

In the last few years, the interest in resonance light scattering has been renewed by the development of high intensity tunable laser sources which allowed irradiation of atoms with strong light-fields in conditions such that absorption and stimulated emission predominate over spontaneous emission.

Several experimental groups have studied the fluorescence light emitted by a sodium atomic beam irradiated by a c.w. dye laser beam (the two beams and the direction of observation are mutually perpendicular so that the Doppler effect is removed). Different types of measurements have been made, including spectral distribution of the fluorescence light (Schuda *et al.* 1974; Walther 1975; Wu *et al.* 1975; Hartig *et al.* 1976; Grove *et al.* 1977) and photon correlations, more precisely the probability for detecting two photon arrivals separated by an interval τ (Kimble *et al.* 1977; Walther 1977, personal communication).

In this short theoretical paper, we will focus on the problem of 'antibunching' in single atom fluorescence, a subject which has recently attracted a lot of attention. Antibunching means a tendency of the photons to stay away from each other, in contrast with the well known bunching effect discovered by Hanbury Brown & Twiss (1956, 1957). Several theoretical papers have considered the problem of antibunching (Glauber 1963, 1964; Stoler 1974; Carmichael & Walls 1976; Kimble & Mandel 1976; Cohen-Tannoudji 1977). Here, we will not enter into any detailed calculations, but will just make a few remarks and some new suggestions based on a straightforward interpretation of the antibunching effect in terms of 'reduction of the wave packet'.

2. Interpretation of the antibunching effect

In a photon correlation experiment, the fluorescence light is monitored by two photomultipliers, and the probability $P(\varepsilon_b, t+\tau; \varepsilon_a, t)$ for detecting one photon with polarization ε_a at time t and another one with polarization ε_b a time τ later is measured. Such a probability is proportional (Glauber 1964) to the higher order correlation function $\langle E_a^-(t) E_b^-(t+\tau) E_b^+(t+\tau) E_a^+(t)\rangle$, where E_a^+, E_a^-, E_b^+, E_b^- are respectively the positive and negative frequency parts of the ε_a and ε_b polarization components of the Heisenberg electric field operator.

In experimental conditions such that the detected light is emitted by a single atom (single atom fluorescence), the electric field is proportional to the atomic dipole operator D, so that the signal reduces to

$$P(\varepsilon_b, t+\tau; \varepsilon_a, t) \approx tr[D_a^+(t)\, D_b^+(t+\tau)\, D_b^-(t+\tau)\, D_a^-(t)\, \sigma], \tag{1}$$

where σ is the density matrix of the total system, and D^+ and D^- the raising and lowering parts of D.

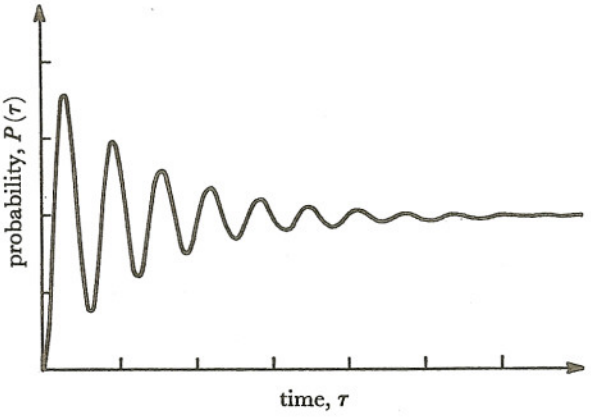

FIGURE 1. Variations with τ of the probability $P(\tau)$ of detecting two photons separated by an interval τ (two-level atom; resonant irradiation $\omega_L = \omega_0$; $\omega_1 = 10\,\Gamma$).

When the Heisenberg operators are expressed in terms of the evolution operators U, by using the invariance of a trace in a circular permutation, the above expression is transformed into

$$tr[D_b^-\, U(t+\tau, t)\, D_a^- \sigma(t)\, D_a^+ U^+(t+\tau, t)\, D_b^+] = q(\varepsilon_b, t+\tau \,|\, \varepsilon_a, t)\, p(\varepsilon_a, t), \tag{2}$$

where

$$p(\varepsilon_a, t) = tr[D_a^-\, \sigma(t)\, D_a^+], \tag{3}$$

$$q(\varepsilon_b, t+\tau \,|\, \varepsilon_a, t) = tr[D_b^-\, U(t+\tau, t)\, \Sigma_a(t)\, U^+(t+\tau, t)\, D_b^+], \tag{4}$$

$$\Sigma_a(t) = (D_a^-\, \sigma(t)\, D_a^+)/tr[D_a^-\, \sigma(t)\, D_a^+]. \tag{5}$$

The interpretation of this result is straightforward. The term $p(\varepsilon_a, t)$ is simply the probability of detecting one ε_a photon at time t. Immediately after this detection process, there is a 'reduction of the wave packet' and the state of the system is described by the (normalized) reduced density matrix $\Sigma_a(t)$. Starting from this new state $\Sigma_a(t)$ at time t, the system then evolves and the probability for detecting one ε_b photon at time $t+\tau$ is simply given by $q(\varepsilon_b, t+\tau \,|\, \varepsilon_a, t)$.

Since $\sigma(t)$ appears in (5) between the lowering component D_a^- at left and the raising component D_a^+ at right, the reduced density matrix $\Sigma_a(t)$ is restricted to the atomic ground level g. Such a result expresses the well known picture of an atom undergoing a 'quantum jump' from the excited level e to the ground level g when emitting the detected photon. In order to be able to emit a second photon, the atom must be re-excited by the laser light which requires a certain amount of time. This is why $q(\varepsilon_b, t+\tau \,|\, \varepsilon_a, t)$ starts from 0 for $\tau = 0$ (antibunching effect).

Let us illustrate these general considerations with the simple case of a two-level atom saturated by an intense resonant single mode laser beam. One finds in this case (omitting the polarizations ε_a and ε_b which play no role) that

$$\left.\begin{aligned} p(t) &= \tfrac{1}{2}\Gamma, \\ q(t+\tau \,|\, t) &= \tfrac{1}{2}\Gamma\,[1 - \exp(-\tfrac{3}{4}\Gamma\tau)\cos\omega_1\tau], \end{aligned}\right\} \tag{6}$$

where Γ is the natural width of e and ω_1 is the product of the atomic dipole moment by the laser electric field. In deriving (6), we have assumed $\omega_1 \gg \Gamma$ (intense field limit). The oscillatory behaviour of $P(\tau) = pq(\tau)$ (figure 1) reflects the well-known damped Rabi oscillation of a two-level system starting from the lower level and reaching a steady state where both levels are equally populated. The characteristic time of the antibunching effect (width of the 'antibunching hole') is of the order of the Rabi period, $1/\omega_1$, which is much smaller than the radiative lifetime Γ^{-1} of e.

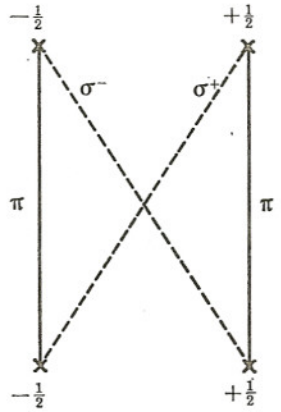

FIGURE 2. $J_g = \frac{1}{2} \leftrightarrow J_e = \frac{1}{2}$ transition.

3. How to change antibunching signals with polarization effects

In this section, we suppose that the upper and lower atomic states have a Zeeman degeneracy, which leads to the possibility of detecting fluorescence photons with different polarizations. Such a degree of freedom could be used for obtaining quite different antibunching signals.

Consider for example a $J_g = \frac{1}{2} \leftrightarrow J_e = \frac{1}{2}$ transition (figure 2) saturated by π-polarized laser light, and suppose that circular analysers are put in front of the two photodetectors. One can, for example, be interested in the probability $P(\sigma_+, \tau\,|\sigma_+)$ of detecting two σ^+ photons separated by an interval τ or in the probability $P(\sigma_-, \tau\,|\sigma_+)$ for detecting a σ^+ photon followed, a time τ later, by a σ^- one (we have suppressed the t-dependence of P which does not appear in steady state as for the two-level case: see equation (6)).

These two probabilities are calculated as explained in § 2 and one obtains

$$\left.\begin{aligned} P(\sigma_\pm, \tau; \sigma_+) &= p(\sigma_+)\, q(\sigma_\pm, \tau\,|\sigma_+), \\ p(\sigma_+) &= \tfrac{1}{6}\Gamma, \\ q(\sigma_\pm, \tau\,|\sigma_+) &= \tfrac{1}{6}\Gamma\,[(1 \mp \exp(-\tfrac{2}{3}\Gamma\tau)) - (\exp(-\tfrac{3}{4}\Gamma\tau) \mp \exp(-\tfrac{5}{12}\Gamma\tau))\cos\omega_1\tau]. \end{aligned}\right\} \quad (7)$$

The striking difference between these two results, represented in figure 3, can be simply interpreted as follows. The 'reduction of the wave packet' following the detection of the first σ^+ photon puts the atom in the $-\frac{1}{2}$ ground sublevel. Then, the π-polarized laser excitation induces a Rabi oscillation between the two $-\frac{1}{2}$ sublevels of e and g. After half a Rabi period, the atom has a great probability (of the order of 1 if $\omega_1 \gg \Gamma$) of being in the upper $-\frac{1}{2}$ sublevel, and, consequently, the probability of spontaneously emitting a σ^- (or a π) photon is large (about 4 times larger than in the steady state where all the populations have the same value $\frac{1}{4}$). This explains the rapid growth of $P(\sigma^-, \tau\,|\sigma^+)$ and the ratio between its maximum and asymptotic

values. On the other hand, for emitting a second σ^+ photon, the atom must necessarily be re-excited during the time τ from the $-\frac{1}{2}$ sublevel of g to the $+\frac{1}{2}$ sublevel of e. However, such a re-excitation requires spontaneous transitions since the π-polarized laser excitation only couples sublevels with the same magnetic quantum number m. This explains the slower growth of $P(\sigma^+, \tau|\sigma^+)$ which is determined mainly by the characteristic time Γ^{-1} of spontaneous emission, even if the Rabi oscillation (which is also visible on such a signal) has a much shorter period.

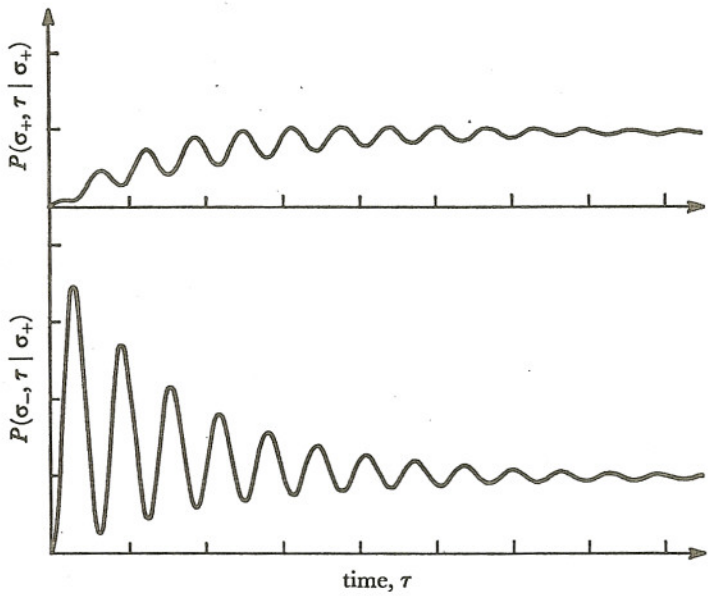

FIGURE 3. Variations with τ of the probabilities $P(\sigma_\pm, \tau \mid \sigma_+)$ for detecting one σ^+ photon followed, a time τ later, by a $\sigma^\pm$ photon ($J_g = \frac{1}{2} \leftrightarrow J_e = \frac{1}{2}$ transition; resonant irradiation $\omega_L = \omega_0$; $\omega_1 = 10\,\Gamma$).

Similar calculations could be made for higher J values. The existence of different Rabi frequencies associated with different Zeeman components, which leads to more complex fluorescence spectra than for the two level case (Cohen-Tannoudji & Reynaud 1977*a*), would give rise to beats in the photon correlation signals.

4. PHOTON CORRELATION SIGNALS IN THE DRESSED ATOM PICTURE

The dressed atom approach provides a simple interpretation of fluorescence and absorption spectra in intense laser fields (Cohen-Tannoudji & Reynaud 1977*b*, *c*, *d*). We show in this section how it can also be applied to photon correlation signals, leading to an interpretation of their modulation as quantum beat effects appearing in radiative cascades.

In the dressed atom picture, one first considers the system formed by the atom and the laser photons. Figure 4(*a*) shows some unperturbed states of such a system in the simple case of a two-level atom: the two states $|e, n\rangle$ and $|g, n+1\rangle$ (atom in e or g in the presence of n or $n+1$ laser photons) are separated by the detuning $\delta = \omega_0 - \omega_L$ between the atomic and laser frequencies. The atom–laser interaction introduces between these two states a coupling $\frac{1}{2}\omega_1$ which leads to the dressed atom energy levels of figure 4(*b*). The two states $|1, n\rangle$ and $|2, n\rangle$, which are some linear combinations of $|e, n\rangle$ and $|g, n+1\rangle$, are separated by a splitting:

$$\bar{\omega} = (\omega_1^2 + \delta^2)^{\frac{1}{2}}. \tag{8}$$

Similar considerations apply to $|e, n-1\rangle$, $|g, n\rangle$, $|1, n-1\rangle$ and $|2, n-1\rangle$ which are at distance ω_L below, and to all other multiplicities (not represented on figure 4).

The coupling of the atom with the empty modes of the electromagnetic field is described by a master equation, which can be written in the dressed atom basis $\{|i, n\rangle\}$. Such a basis is particularly convenient when the dressed atom levels are well separated ($\bar{\omega} \gg \Gamma$), which implies either intense fields ($\omega_1 \gg \Gamma$) or large detunings ($|\delta| \gg \Gamma$). Resonance fluorescence can then be described as being due to spontaneous transitions between the dressed atom levels. The allowed transitions, which correspond to the non-zero matrix elements of the atomic dipole moment D, connect adjacent multiplicities and are represented by the wavy arrows of figure 4 (b). This provides a straightforward interpretation of the triplet structure of the fluorescence spectrum, first predicted by Mollow from a different approach (Mollow 1969): the frequencies $\omega_L + \bar{\omega}$, $\omega_L - \bar{\omega}$, ω_L are associated respectively with the transitions $|1, n\rangle \rightarrow |2, n-1\rangle$; $|2, n\rangle \rightarrow |1, n-1\rangle$, $|i, n\rangle \rightarrow |i, n-1\rangle$ $(i = 1, 2)$.

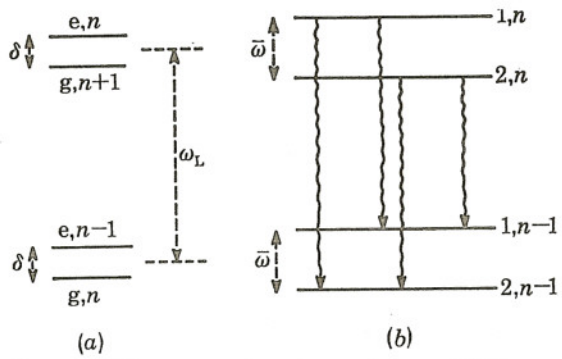

FIGURE 4. (a) Some unperturbed states of the system atom–laser photons. (b) Corresponding dressed atom states. The wavy arrows represent the allowed spontaneous transitions between these states.

To return to the photon correlation signal given in (1), this can now be expressed in the dressed atom basis rather than in the bare atom one. The corresponding calculations are given in the appendix. Here we will just outline some simple physical pictures emerging from these calculations.

The photon correlation signal appears (in the steady state régime and for $\bar{\omega} \gg \Gamma$) as a product of three terms which are associated with the processes represented on figure 5. The first process (figure 5 (a)) corresponds to the detection of the first photon which puts the atom in g: the dressed atom, starting from one of its energy levels, is projected into a linear superposition of the two sublevels of the adjacent lower multiplicity ($|g, n\rangle$ is a linear superposition of $|1, n-1\rangle$ and $|2, n-1\rangle$) described by a projected density matrix having diagonal as well as off-diagonal elements (respectively left and right parts of figure 5 (a)). Then, the dressed atom evolves during the time τ, which corresponds to a spontaneous radiative cascade (figure 5 (b)). During such a cascade, a redistribution of the populations π_1 and π_2 occurs as well as a damped oscillation of the off-diagonal elements. Finally, we have the second detection process, represented in figure 5 (c). Since the density matrix before this detection process has diagonal as well as off-diagonal elements, the signal now contains static and modulated components (respectively left and right parts of figure 5 (c)). Note the difference from the first detection process, where we start from the purely diagonal steady state density matrix.

The previous analysis clearly shows that the modulation of the photon correlation signal is due to the 'coherence' between the two sublevels, $|1, n-1\rangle$ and $|2, n-1\rangle$, introduced by the first detection process. Such a process plays the same role as the percussional excitation which, in a quantum beat experiment, prepares the atom in a coherent superposition of two excited sublevels. Note however that the situation represented in figure 5 corresponds to a radiative cascade so that it would be more judicious to compare it with perturbed correlations in atomic or nuclear radiative cascades (Steffen & Frauenfelder 1964).

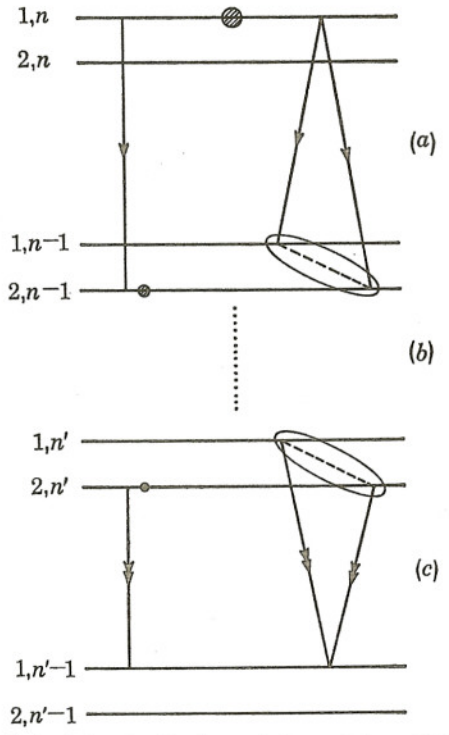

FIGURE 5. (*a*) Detection of the first photon in the dressed atom picture. (*b*) Evolution of the system during the time τ. (*c*) Detection of the second photon.

5. PHOTON CORRELATION SIGNALS WITH FREQUENCY SELECTION

Up to now, we have implicitly supposed that the photodetectors are broad band detectors with extremely short response time. The discussion of the previous section shows that the two detection processes can be considered as instantaneous as soon as the bandwidth $\Delta\nu$ of the photomultipliers is large in comparison with the beat frequency $\overline{\omega}$. Note the analogy with quantum beat experiments where the spectral width of the exciting pulse must be larger than the atomic structure giving rise to the beats (broad band condition).

This leads us to investigate the modification of the photon correlation signals which would result from the insertion of frequency filters in front of the photodetectors. More precisely, we will suppose that these filters, centred on one of the three components of the fluorescence spectrum ($\omega_L+\overline{\omega}$, $\omega_L-\overline{\omega}$ or ω_L), have a bandwidth $\Delta\nu$ that is small compared to $\overline{\omega}$ (only one component is selected) but large compared to Γ (the filtered component is not distorted):

$$\Gamma \ll \Delta\nu \ll \overline{\omega}. \tag{9}$$

The analogy with quantum beats discussed above suggests that the modulation at $\bar{\omega}$ in the photon correlation signal should disappear in such a case. This is what actually occurs: each frequency filter selects one particular frequency component of the dipole moment D so that the modulated terms sketched on the right part of figure 5 vanish, since two different frequency components of D simultaneously appear in every detection process.

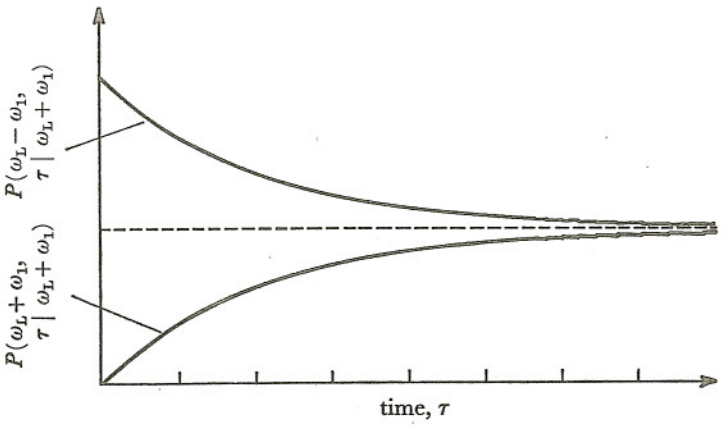

FIGURE 6. Variations with τ of the probabilities $P(\omega_L \mp \omega_1, \tau \mid \omega_L + \omega_1)$ for detecting one $\omega_L + \omega_1$ photon followed, a time τ later, by a $\omega_L \mp \omega_1$ photon (two-level atom; resonant irradiation $\omega_L = \omega_0$; $\omega_1 \gg \Gamma$).

The signal can now be entirely described in terms of populations (as shown in the left part of figure 5). Suppose, for example, that the first filter is centred at $\omega_L + \bar{\omega}$. The first detector is then only sensitive to transitions of the type $|1, n+1\rangle \rightarrow |2, n\rangle$. After this first detection, the dressed atom state is projected into $|2, n\rangle$. Consequently, it cannot emit a second $\omega_L + \bar{\omega}$ photon immediately afterwards since no $\omega_L + \bar{\omega}$ transition starts from this level $|2, n\rangle$ (figure 4(*b*)). The probability $P(\omega_L + \bar{\omega}, \tau \mid \omega_L + \bar{\omega})$ for detecting two $\omega_L + \bar{\omega}$ photons separated by a time τ exhibits therefore an antibunching behaviour. On the other hand, one $\omega_L - \bar{\omega}$ photon can be emitted from the $|2, n\rangle$ state (figure 4(*b*)). Immediately after the detection of the first $\omega_L + \bar{\omega}$ photon, the population π_2 of level $|2, n\rangle$ has a value 1, larger than the steady state value (reached for large values of τ), so that $P(\omega_L - \bar{\omega}, \tau \mid \omega_L + \bar{\omega})$ exhibits a bunching behaviour (the apparent contradiction with the general discussion of § 2 which excludes bunching effects in single atom fluorescence is removed by noting that the filtering devices store the emitted photons during a time $(\Delta\nu)^{-1}$ much larger than $(\bar{\omega})^{-1}$). We have illustrated these considerations in figure 6, which gives the two probabilities $P(\omega_L \pm \omega_1, \tau \mid \omega_L + \omega_1)$ calculated in the simple case of a resonant irradiation ($\delta = 0$; $\bar{\omega} = \omega_1$).

In the off-resonant case ($|\delta| \gg \omega_1$; $\bar{\omega} \approx |\omega_0 - \omega_L|$), the fluorescence spectrum can be interpreted perturbatively: the central component ω_L corresponds to the Rayleigh process of figure 7(*a*) while the two sidebands at frequencies $\omega_A = 2\omega_L - \omega_0$ and $\omega_B = \omega_0$ are associated with the second order nonlinear scattering process of figure 7(*b*). We have represented on figure 8 the two frequency filtered correlation signals that appear at lowest order (i.e. $(\omega_1/\delta)^4$, all others being of higher order). The variation of these two signals can be analysed from the same perturbative approach. The probability $P(\omega_L, \tau \mid \omega_L)$ for detecting two Rayleigh photons separated by an interval τ does not vary with τ. This is due to the fact that the two corresponding Rayleigh scattering events are uncorrelated. Although the diagram of figure 7(*b*) is of higher order than the one of figure 7(*a*), it can give rise to a photon correlation signal of the

same order. After the absorption of two laser photons and the emission of the ω_A one, the atom reaches level e (figure 7 (*b*)) from which it has a great probability for emitting one ω_B photon. This explains also the exponential decrease of $P(\omega_B, \tau | \omega_A)$ with the radiative lifetime Γ^{-1}. If one calculates the probability $P(\omega_A, \tau | \omega_B)$ of emission of the two photons ω_A and ω_B in the reverse order, one finds a much smaller quantity (in $(\omega_1/\delta)^8$) independent of τ. This shows that, for each nonlinear scattering process, the two photons ω_A and ω_B appear in a certain order with a delay of the order of Γ^{-1} and that, for detecting them in the reverse order, one requires two independent scattering processes.

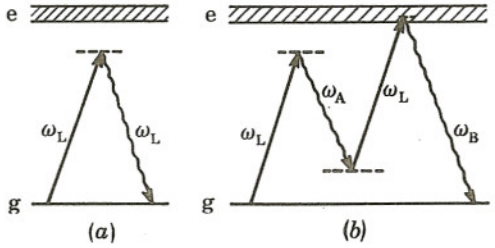

FIGURE 7. Perturbative interpretation of the three components of the fluorescence spectrum for large detunings: (*a*) Rayleigh component. (*b*) Sidebands $\omega_A = 2\omega_L - \omega_0$ and $\omega_B = \omega_0$.

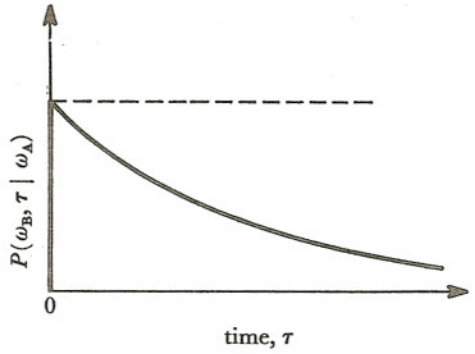

FIGURE 8. Variations with τ of the probability $P(\omega_B, \tau \mid \omega_A)$ for detecting one photon ω_A followed, a time τ later, by a ω_B photon. The probability $P(\omega_L, \tau \mid \omega_L)$, denoted by the broken line, for detecting two Rayleigh photons ω_L does not depend on the interval τ.

APPENDIX

In this appendix, we find explicitly the photon correlation signal in the dressed atom basis $|i, n\rangle$. The notation is the same as in Cohen-Tannoudji & Reynaud (1977*b*).

Because of the quasi-classical character of the laser field, the various elements of the density matrix $\sigma(t)$ may be written as

$$\langle i, n|\sigma(t)|j, n-p\rangle = \rho_{ij}^{p}(t)\, p_0(n), \tag{A 1}$$

where $p_0(n)$ is the normalized distribution of the number of photons in the laser and the $\rho_{ij}^{p}(t)$ are some reduced density matrix elements. We will omit the symbol p for the elements corresponding to $p = 0$: i.e., for the populations $\pi_i = \rho_{ii}^{0}$ of the dressed atom states and for the coherences' $\rho_{ij} = \rho_{ij}^{0}$ between states of the same multiplicity.

The only non-zero matrix elements of the dipole moment D between the dressed atom states are

$$\left.\begin{aligned} d_{ij}^{-} &= \langle i, n-1\,|D|\, j, n\rangle, \\ d_{ij}^{+} &= \langle i, n+1\,|D|\, j, n\rangle = (d_{ji}^{-})^{*} \end{aligned}\right\} \tag{A 2}$$

(we therefore ignore the vectorial character of D). In order to get a simple expression for the correlation signal, we decompose the lowering and raising parts of D in their frequency components:

$$D^- = \sum_{ij} D_{ij}^-; \quad D^+ = \sum_{ij} D_{ij}^+, \tag{A 3}$$

$$\left.\begin{aligned} D_{ij}^- &= d_{ij}^- \sum_n |i, n-1\rangle \langle j, n| \\ D_{ij}^+ &= d_{ij}^+ \sum_n |i, n+1\rangle \langle j, n| \end{aligned}\right\}. \tag{A 4}$$

By introducing this decomposition (A 3) of D into the expression (1) of the photon correlation signal, one obtains:

$$P(t+\tau|t) = \sum_{ij} \sum_{kl} \sum_{pq} \sum_{rs} P_{ij\,kl\,pq\,rs}(t+\tau|t), \tag{A 5}$$

where

$$P_{ij\,kl\,pq\,rs}(t+\tau|t) = \langle D_{ij}^+(t)\, D_{kl}^+(t+\tau)\, D_{pq}^-(t+\tau)\, D_{rs}^-(t)\rangle. \tag{A 6}$$

By using the quantum regression theorem (Lax 1968), this expression is transformed into

$$P_{ij\,kl\,pq\,rs}(t+\tau|t) = [\delta_{lp}\, d_{kl}^+\, d_{pq}^-\, \rho(qk, \tau|rj)]\, [d_{ij}^+\, d_{rs}^-\, \rho_{si}(t)], \tag{A 7}$$

where $\rho(qk, \tau|rj)$ is a Green function of the master equation; $\rho(qk, \tau|rj)$ is the value of $\rho_{qk}(\tau)$ corresponding to an initial state where only $\rho_{rj}(0)$ is non-zero ($\rho_{rj}(0) = 1$). Because of the secular approximation, the elements $\rho_{ij}(t)$ evolving at different frequencies are not coupled; the off-diagonal elements ($i \neq j$) are only coupled to themselves and they tend to zero after a transient damped oscillation. The populations are coupled together and they reach a steady state régime after a time of the order of Γ^{-1}. This allows some important simplifications of expression (A 7): first, if we neglect the transient contribution of atoms entering the laser beam, only the steady state populations contribute to the second factor of (A 7) (which implies $s = i$); then the only non-zero Green functions correspond either to the damped oscillation of a 'coherence' ($q = r, k = j, r \neq j$), or to the redistribution of the populations ($r = j, q = k$). The corresponding contributions to $P(t+\tau|t)$ can be written (respectively for the two types of terms):

$$\delta_{lp}\, \delta_{si}\, \delta_{qr}\, \delta_{kj} [d_{jl}^+ d_{lr}^-]\, \rho(rj, \tau|rj)\, [d_{ij}^+ d_{ri}^- \Pi_i(\infty)] \tag{A 8}$$

with $r \neq j$

$$\delta_{lp}\, \delta_{si}\, \delta_{qk}\, \delta_{rj}\, \Gamma_{lk}\, \Pi(k, \tau|j)\, [\Gamma_{ji} \Pi_i(\infty)], \tag{A 9}$$

where Γ_{ji} is the transition rate from level $|i, n\rangle$ to level $|j, n-1\rangle$ ($\Gamma_{ji} = d_{ij}^+ d_{ji}^- = |d_{ji}^-|^2$) and $\Pi(k, \tau|j)$ is the population $\Pi_k(\tau)$ corresponding to an initial state where only $\Pi_j(0)$ is different from 0 and equal to 1. Note that $\rho(rj, \tau|rj)$ is simply equal to $\exp(-L_{rj}\tau) \exp(-i\omega_{rj}\tau)$ where ω_{rj} is the energy difference between levels $|r, n\rangle$ and $|j, n\rangle$ and L_{rj} the width of the $\omega_L + \omega_{rj}$ component of the fluorescence spectrum. The right and left parts of figures 5 (*a*), 5 (*b*) and 5 (*c*) correspond respectively in the algebraic expressions (A 8) and (A 9) to the terms $d_{ij}^+ d_{ri}^- \Pi_i(\infty)$ and $\Gamma_{ji}\Pi_i(\infty)$, $\rho(rj, \tau|rj)$ and $\Pi(k, \tau|j)$, $d_{jl}^+ d_{lr}^-$ and Γ_{lk}.

If one uses frequency filters satisfying condition (9), only certain terms of the type (A 9) contribute to the signal. These are the ones for which Γ_{ji} and Γ_{lk} correspond to the mean frequencies of the two filters. For example, the various signals represented on figures 6 and 8 are easily found to be

$$P(\omega_L \pm \omega_1, \tau; \omega_L + \omega_1) = (\tfrac{1}{4}\Gamma)^2\, \tfrac{1}{2}(1 \mp \exp(-\Gamma\tau/2)), \tag{A 10}$$

$$P(\omega_L, \tau; \omega_L) = (\Gamma\omega_1^2/4\delta^2)^2, \tag{A 11}$$

$$P(\omega_B, \tau; \omega_A) = (\Gamma\omega_1^2/4\delta^2)^2 \exp(-\Gamma\tau). \tag{A 12}$$

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Discussion

R. K. Bullough (*Department of Mathematics, U.M.I.S.T., P.O. Box* 88, *Manchester M*60 1*QD, U.K.*). I should like to raise the question of how far the antibunching of photons in resonance fluorescence is concerned with photons at all. It is fair to say that the argument for considering the intensity–intensity correlation function $G^{(2)}(\tau)$ in the form $G^{(2)}(\tau) \equiv \langle D^+(t)\,D^+(t+\tau)\,D^-(t+\tau)\,D^-(t)\rangle$ considered by Dr Cohen-Tannoudji goes most easily in terms of quantized field operators. But, given this form of the correlation function, the antibunching becomes solely a property of the atom rather than the field. For a 2-level atom at $\tau = 0$ and $t = 0$, as Cohen-Tannoudji has mentioned, D^- cannot lower the atom twice – in photon language, it cannot emit a photon and then another (correlated) photon without the atom first returning to its excited state before emitting the second photon. However, it is the transitions of the atom we are concerned with in this description and not the photons. In the steady state $t = \infty$, and all finite t, one also has the operator property for 2-level atoms $(D^+(t))^2(D^-(t))^2 = 0$ for $\tau = 0$. Again this is strictly a property of the atom.

My colleague Dr S. S. Hassan and I have calculated the value of $G^{(2)}(\tau)$ for a 2-level atom driven by a single mode coherent state field and bathed in a multimode broad band chaotic field. I shall quote the results in a number of limiting cases in one moment, and they certainly indicate the significance of the atom to this sort of antibunching. First of all I would like to make a comment on the corresponding resonance fluorescence spectrum.

We have calculated this exactly (Hassan & Bullough 1977)†. Its general features are a light-

† Our 'exact solution' assumes rotating wave approximation for the coherent field but not for the chaotic, but involves a decorrelation of the chaotic field from the atomic inversion. The result is then obtained in closed form as a closed expression for the Laplace transform on τ.

shifted asymmetric (even on resonance) spectrum consisting of three peaks in the case when the Rabi frequency $2|A| \gg n_{k_s}\Gamma_0$ (n_{k_s} is the mean occupation number of the chaotic mode on resonance with the vacuum shifted atomic frequency; Γ_0 is the A-coefficient). In the weak coherent field case, $2|A| \ll \Gamma_0$, $n_{k_s}\Gamma_0$, the inelastic spectrum is essentially a light-shifted Lorentzian characteristic of the broad band chaotic field and the Einstein rate equation régime. Superimposed on this are two Lorentzians, neither being light-shifted at resonance, but having relative order $|A|^2\Gamma_0^{-2}$. Then there is the elastic scattering δ-function of strength $\langle D^+(\infty)\rangle\langle D^-(\infty)\rangle$. This classical result obtained from the exact quantum theory is the reason for believing that atoms scatter through the electric dipole moment induced in them by the incident electric field. It is the source of Professor Buckingham's analysis (paper 4 following) and indeed of all the work presented at this meeting for discussion! Far enough off resonance it will always dominate the single atom scattering process in $S(\boldsymbol{k}, \omega)$ but for resonance scattering with incident fields of about 1 mW cm^{-2} or above it is necessary to consider dynamical Stark effects of this kind as well as the classical dipole scattering. No many-body theory as comprehensive as this has yet been constructed.

We have also calculated $G^{(2)}(\tau)$ exactly. Our method of calculation differs from the dressed atom method of Dr Cohen-Tannoudji and involves the use of operator reaction field theory. I have been particularly delighted by his demonstration both here and on previous occasions, that a driven atom problem can, by changing to the dressed atom basis, be treated as a spontaneous emission problem in which the dressed atom cascades down its own sequence of Bohr energy levels emitting fluorescence photons as it does so. The method is particularly effective in yielding positions, weights and widths of peaks in the strong single mode coherent field limit for quite complicated multilevel atoms. We have indeed been so delighted by the dressed atom picture that my colleague Mr E. Abraham has derived the master equation Dr Cohen-Tannoudji quotes from reaction field theory by transforming to the dressed atom basis. It can be derived exactly.

Outside the strong field single mode régime, however, we find the dressed atom picture somewhat less effective. In our opinion this very elegant transformation is not well adapted to the *exact* solution of the atom–mixed coherent–chaotic field problem. (Dr Cohen-Tannoudji may not agree however?)

I quote our results for $G^{(2)}(\tau)$. Notice that each result takes the form $p(t)\Pi(\tau)$ quoted by Dr Cohen-Tannoudji. Furthermore $p(t) \equiv \frac{1}{2}(1+\bar{R}_3(t))$, and $\bar{R}_3(t)$ is the atomic inversion so that $p(t)$ is indeed the probability of the atom being in its upper state. The general form is an aspect of an extended form of the quantum regression theorem, although we nowhere appeal to this theorem. If the field is quantized it amounts to commuting free field and matter operators but there are arguments why this can be done even in this quantized case and so there may be no evidence of photons here either.

The results for $G^{(2)}(\tau)$ are:

(i) Exact resonance, strong coherent field ($2|A| \gg n_{k_s}\Gamma_0$):

$$G^{(2)}(\tau) = \tfrac{1}{4}(1+\bar{R}_3(t))\,[1-\exp\{-(\tfrac{3}{4}+n_{k_s})\Gamma_0\tau\}\{\cos(2|A|\tau)+3(\tfrac{1}{4}+n_{k_s})(\Gamma_0/2|A|)\sin(2|A|\tau\}].$$

The result is that quoted by Dr Cohen-Tannoudji except that the A-coefficient is power broadened by the chaotic field (but not in the $(1+2n_{k_s})\Gamma_0$ Einstein rate equation form) and we include Rabi oscillations of order $\Gamma_0|A|^{-1}$ in phase quadrature as the leading correction from our exact solution.

This solution both bunches and antibunches but $G^{(2)}(0) = 0$.

(ii) Exact resonance, weak coherent field, ($n_{k_s'}\Gamma_0$, $\Gamma_0 \gg 2|A|$ or $\frac{1}{2}\Gamma_0 + 2\Gamma_0 n_{k_s'} > 4|A|$):

$$G^{(2)}(\tau) = \tfrac{1}{4}(1+\bar{R}_3(t))\,[g_1 - g_2 \exp(-\tfrac{1}{2}\Gamma_0\tau) - g_3 \exp\{-\Gamma_0(1+2k_i)\tau\}],$$

$$g_1 = \frac{2(\Gamma_0^2 n_{k_s'} + 4|A|^2)}{\Gamma_0^2(1+2n_{k_s'})}, \quad g_2 = \frac{8|A|^2}{\Gamma_0^2(\frac{1}{2}+2n_{k_s'})},$$

$$g_3 = \frac{2\Gamma_0^2 n_{k_s'}(2n_{k_s'}+\frac{1}{2}) - 4|A|^2}{\Gamma_0^2(1+2n_{k_s'})\,(2n_{k_s'}+\frac{1}{2})}.$$

This result only antibunches and $G^{(2)}(0) = 0$. This weak coherent field case contains three further cases of interest within it.

(iii) Weak coherent field, no chaotic field ($n_{k_s'} = 0$):

$$G^{(2)}(\tau) = 2|A|^2\Gamma_0^{-2}(1+\bar{R}_3(t))\,[1-\exp(-\tfrac{1}{2}\Gamma_0\tau)]^2.$$

This result agrees with that of Carmichael & Walls (1976).† It only antibunches, and $G^{(2)}(0) = 0$.

(iv) Pure chaotic field ($|A| = 0$):

$$G^{(2)}(\tau) = \tfrac{1}{2}(1+\bar{R}_3(t))\,n_{k_s'}(2n_{k_s'}+1)^{-1}[1-\exp\{-\Gamma_0(1+2n_{k_s'})\tau\}]$$
$$= [n_{k_s'}(2n_{k_s'}+1)^{-1}]^2[1-\exp\{-\Gamma_0(1+2n_{k_s'})\tau\}],$$

for $t = \infty$. This $G^{(2)}(\tau)$ only antibunches, and $G^{(2)}(0) = 0$.

(v) Pure spontaneous emission ($n_{k_s'} = |A| = 0$ – our initial condition is that the atom starts in its upper state at $t = 0$):

$$G^{(2)}(\tau) = 0 \text{ for all } \tau > 0.$$

It is obvious that the spontaneously emitting atom is the best of all antibunchers; once it has emitted its photon (fallen to its ground state) it cannot emit a second one and so it remains in that ground state).

The case (iv) illustrates the key role of the atom, since the $G^{(2)}(\tau)$ for the pure chaotic field without the atom is bunched with $G^{(2)}(0) = 2\times$ intensity squared (so that $G^{(2)}(0) = 2$: the Hanbury Brown–Twiss situation). Indeed, all five results illustrate the point that the antibunching feature is particularly associated with the atom. Thus the photons must at best be associated with the measuring process for it is this which suggests we calculate $G^{(2)}(\tau)$ in the form Dr Cohen-Tannoudji assumed.

In final comment it might be helpful actually to show the form of the spectrum for a 2-level atom in the mixed coherent–chaotic field. The chaotic field can be characterized for present purposes by two numbers, $n_{k_s'}$ and n_3. In the absence of the coherent field the light shift $\Delta_1 \equiv n_3\Gamma_0\pi^{-1}$. In an additional strong resonant coherent field the central peak of the three-peaked spectrum shifts $\frac{1}{2}\Delta_1$: the side bands each shift $\frac{1}{4}\Delta_1$. The number n_3 can be about 5 for a broad band chaotic field of 5 mW cm^{-2} per MHz and for sodium D_2 transitions ($\Gamma_0 \approx$ 50 MHz) $\Delta_1 \approx$ 80 MHz. Figure 1 shows the resonant spectrum for $|A| = 5\Gamma_0$, $n_{k_s'} = 1$ and $n_3 = 0$, 1, 2, 5 and 10. The different curves may be identified by the movement of their central peaks as n_3 increases. The abscissa is $(\omega - \omega_k)\Gamma_0^{-1}$: ω_k is the laser frequency on exact resonance

† Result (i) for $n_{k_s'} = 0$ was also first reported by Carmichael & Walls. We reported results (i) (for $n_{k_s'} = 0$) and (iii) at the one-day International Conference on resonant light scattering, at M.I.T. (30 April 1976), and I believe Dr Cohen-Tannoudji had similar results then.

with the vacuum shifted atomic frequency ck'_s. As the laser is detuned from resonance a single Lorentzian centred near the vacuum shifted frequency, but still Stark shifted, emerges, so that off resonance the light shifted asymmetry is crushed by the asymmetry due to the chaotic field

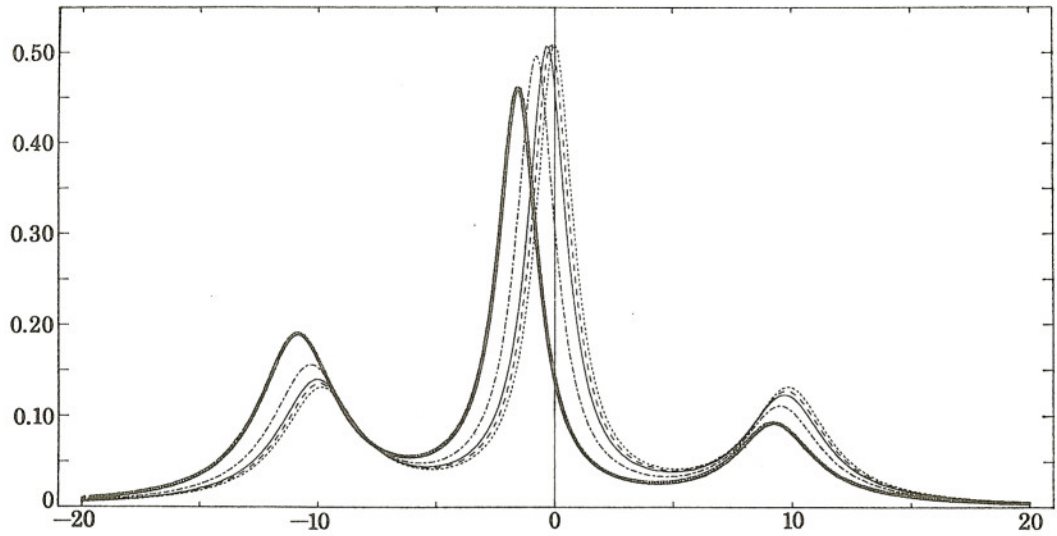

FIGURE 1. Asymmetrically light shifted three peaked resonance fluorescence spectrum. Exact resonance.

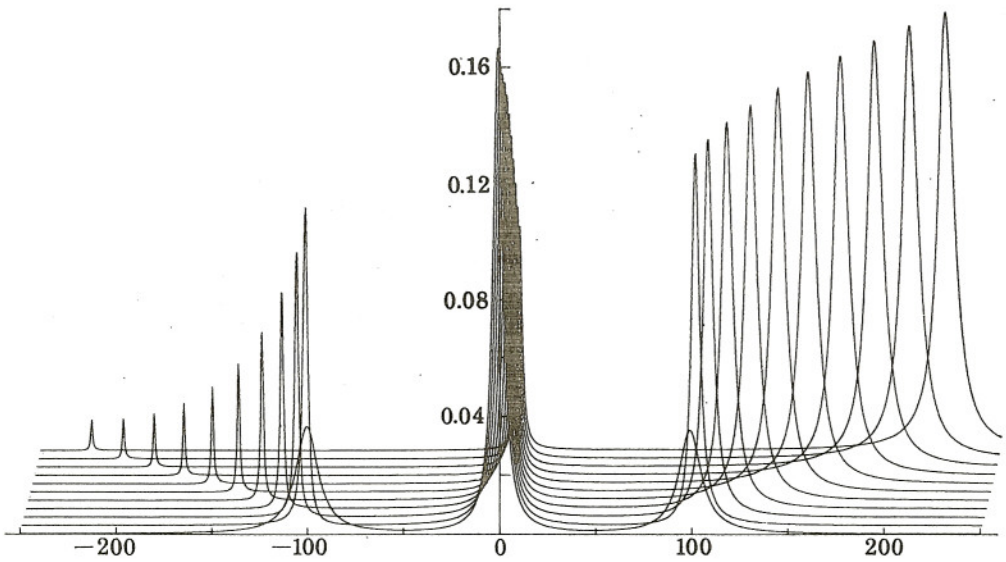

FIGURE 2. Strong opposite asymmetry developed by increasing detuning of the laser.

spectrum. Figure 2 shows the spectrum for $|A| = 50\Gamma_0$, $n_{k'_s} = n_3 = 5$ and detuning $0, -20, \ldots, -200$ in units of Γ_0. The emergence of the chaotic field peak associated with the Einstein rate equation régime is quite spectacular and shows how this régime dominates the non-resonant scattering process. The classical elastic scattering δ-function is not plotted on the Figures but is of course present in each case though is relatively weak on resonance.

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C. COHEN-TANNOUDJI. For a fermion field, the second order correlation function $\langle \phi^-(r, t)\ \phi^-(r, t+\tau)\ \phi^+(r, t+\tau)\ \phi^+(r, t)\rangle$ always vanishes for $\tau = 0$. This antibunching effect is certainly a property of the fermion field. On the other hand, for a photon field, Glauber has shown that, depending on the state of the field, the second order correlation function $\langle E^-(r, t)\ E^-(r, t+\tau)\ E^+(r, t+\tau)\ E^+(r, t)\rangle$, may exhibit either bunching or antibunching behaviour. For a field emitted by a source, such behaviour obviously depends on the atomic properties of the source. One predicts a continuous transition between antibunching (single atom source) and bunching (ensemble of many independent atoms). Coming back to the single atom case, it is clear that the emitted field is related to the atomic dipole. Since this atomic operator has a quantum nature [$D(t)$ and $D(t')$ do not commute when $t \neq t'$] one cannot ignore the quantum nature of the field, and this explains why one cannot construct a classical random field leading to the same result as the full quantum theory. On the other hand, for a many-atom source, crossed terms between the fields emitted by different atoms become predominant. Since different dipole moment operators generally commute (uncorrelated atoms), it becomes possible in this case to simulate the results with a classical chaotic field.

Mathematically, the dressed atom approach consists in choosing a particular basis of states (the eigenstates of the atom-laser mode subsystem) for writing equations of motion including the effect of the coupling with the empty modes. Such a basis is particularly convenient in the strong field régime since, in such a case, the non-secular terms associated with spontaneous emission (coupling with the empty modes) have a negligible contribution in comparison with the secular ones. Neglecting these non-secular terms leads to simple equations having a simple physical interpretation. However, if one keeps the non-secular terms, one gets exact equations which are also valid in the weak field regime and which can be shown to be strictly equivalent with the semiclassical equations.

We think that the dressed atom approach can be easily extended to the situation of an atom + mixed coherent chaotic field. One has to introduce first the energy levels of the single mode laser-atom system (dressed atom). The effect of the broad band chaotic field can then be described in this basis by a master equation quite analogous to the one describing spontaneous emission. One obtains new terms describing absorption and stimulated emission processes induced by the chaotic field between the dressed atom energy levels. Such an approach may be shown to lead to simple physical interpretation for the asymmetry of the fluorescence spectrum (analogy with collision induced fluorescence).

J. M. VAUGHAN (*Royal Signals and Radar Establishment* (*S*), *Great Malvern, Worcs., U.K.*). After the interesting paper of Dr Cohen-Tannoudji I think it is worth commenting on the difficulty that is likely to be experienced in observing true photon antibunching. We have been interested in this problem at R.S.R.E. and in a recent letter due to Jakeman *et al.* (1977) we point to the problem when a randomly fluctuating number of atoms is observed. We comment that in the recent experiment of Kimble *et al.* (1977) antibunching, with a non-classical intercept of the intensity correlation function less than unity, is attained only after making a heterodyne correction.

This correction factor may not be physically realistic, and if it is neglected the experimental intercept is in fact unity within experimental error. Our calculation derives the expected intercept when the source contains a fluctuating number of atoms each emitting antibunched radiation. In the case of a Poisson distribution of atoms, the extra degree of randomness leads to a predicted value of the intercept of exactly unity. According to this view, in present experiments the antibunched character of radiation from a single atom may be inferred but has not been observed.

Similar considerations are likely to apply to the ingenious possibilities outlined by Dr Cohen-Tannoudji, and it would seem that antibunching will only be observed when a fixed, small number of atoms or molecules, preferably only one, is examined.

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C. COHEN-TANNOUDJI. In the present paper we are interested in the fact that, in single atom fluorescence, the distribution of relative arrival times of photons, $P(\tau)$, is an increasing function of τ around $\tau = 0$, contrary to what is observed in Hanbury Brown & Twiss's experiment (where it is a decreasing function). Such a behaviour is not destroyed by fluctuations in the number of atoms, provided that the mean number of atoms in the observation volume is sufficiently small: $P(\tau)$ remains an increasing function of τ around $\tau = 0$, even if $P(0)$ is no longer equal to zero. Similarly, the different schemes proposed here for increasing the width of the 'hole' of $P(\tau)$ around $\tau = 0$ remain valid in presence of fluctuations of the number of atoms.