Correlation signals in resonance fluorescence : interpretation *via* photon scattering amplitudes

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Résumé. — La fluorescence de résonance est considérée ici comme une collision où des photons laser incidents sont diffusés par un atome. Les signaux de corrélation de photons sont calculés à partir du vecteur d'état décrivant le champ diffusé au second ordre. L'effet de dégroupement de photons provient d'une interférence quantique entre toutes les amplitudes de diffusion possibles. Quand les photons Rayleigh sont éliminés, certaines amplitudes disparaissent ce qui conduit à un groupement des photons détectés.

Abstract. — Resonance fluorescence is treated as a collision process where incident laser photons are scattered by an atom. Correlation signals are extracted from an expansion to the second order of the post collision field state. Photon antibunching effect appears as a quantum interference between all the possible scattering amplitudes. When Rayleigh photons are rejected, some amplitudes vanish, leading to a bunching behaviour.

1. Introduction.

In the last twenty years, a great number of experimental and theoretical works has been devoted to the study of resonance fluorescence, i.e., emission of photons by an atom irradiated by a resonant laser wave. First, it has been shown that the fluorescence spectrum has a triplet structure [1-3] (for a sufficiently intense excitation). Then, the temporal correlations between fluorescence photons have been analysed exhibiting an antibunching effect for single atom emission [4, 5]. These two types of experiments emphasize respectively the frequency or time features of photons. More recently, a mixed analysis has been performed by studying temporal correlations between fluorescence photons selected through frequency filters. This experiment has shown that the photons emitted in the two sidebands of the fluorescence triplet are strongly correlated, bunched and emitted in a given time order [6] (for a non saturating and non resonant excitation).

A powerful theoretical method initiated by Mollow [7] has been progressively developed for the investigation of resonance fluorescence. The crucial feature of this method is to relate fluorescence signals to some correlation functions of the emitting atomic dipole. The evolution of the dipole is described for example by optical Bloch equations (prediction of

the triplet by Mollow [7], of the antibunching by Cohen-Tannoudji [8] and Carmichael and Walls [9]...) or by the dressed atom approach [10] which is in particular well adapted to the study of correlations' between frequency selected photons [11]. Correlation functions are then evaluated through the quantum regression theorem [12, 13].

In this paper, we come back to the earlier treatment of resonance fluorescence based on the scattering theory [14, 15]. The « precollision » wave packet $| \psi_i \rangle$ describes the atom in presence of incident laser photons. The « post collision » wave packet $| \psi_f \rangle$ is obtained by action of the S matrix calculated through formal collision theory :

$$| \psi_f \rangle = S | \psi_i \rangle . \qquad (1.1)$$

Such a state contains all the information concerning the scattered field. We will expand it up to the second order in the laser atom interaction (perturbative treatment valid for a non saturating laser excitation. Note that it is possible to use this method for a non perturbative treatment [16-18]). In other words, we will take into account one and two laser photon scattering processes. The correlation signals will then be calculated as average values in $| \psi_f \rangle$ of some field operators (quantum detection theory [19]).

The main purpose of this paper is to justify the qualitative interpretation given in [6] of the bunching between photons selected in the two sidebands of the triplet. We also want to render a physical account of the passage from this bunching behaviour to the antibunching one appearing when frequency filters are removed [28].

2. Notations.

The Hamiltonian H is the sum of three parts :

$$H = H_A + H_R + V \qquad (2.1)$$

where H_A is the atomic Hamiltonian, H_R the radiation one and V the atom-field coupling.

The atom is considered as a two level system so that H_A can be written :

$$H_A = \hbar\omega_0\, b^+ b \qquad (2.2)$$

where ω_0 is the atomic resonance frequency and b and b^+ the lowering and raising operators :

$$b = |g\rangle\langle e| \qquad b^+ = |e\rangle\langle g| \qquad (2.3)$$

($|g\rangle$ is the atomic ground level and $|e\rangle$ the excited one).

The electromagnetic radiation field is quantized on a complete set of orthonormal field distributions $\mathcal{E}_\lambda(\mathbf{R})$, for example the plane waves of wave vector $\mathbf{k}_\lambda$, frequency $\omega_\lambda = c\,|\mathbf{k}_\lambda|$ and polarization ε_λ. The Hamiltonian H_R of the free radiation field is thus :

$$H_R = \sum_\lambda \hbar\omega_\lambda\, a_\lambda^+ a_\lambda \qquad (2.4)$$

where a_λ and a_λ^+ are respectively the destruction and creation operators of a photon in the mode λ.

The atom field coupling V can be written in the electric dipole and rotating wave approximations

$$V = - \mathbf{d}.(b^+\, \mathbf{E}^+(0) + b\mathbf{E}^-(0)) \qquad (2.5)$$

where $\mathbf{d}$ is the atomic dipole moment (here assumed to be real) and $\mathbf{E}^+(0)$ and $\mathbf{E}^-(0)$ the positive and negative frequency components of the electric field taken at the position $\mathbf{R} = 0$ of the atom :

$$\begin{aligned}\mathbf{E}^+(\mathbf{R}) &= \sum_\lambda \mathcal{E}_\lambda(\mathbf{R})\, a_\lambda \\ \mathbf{E}^-(\mathbf{R}) &= \sum_\lambda \mathcal{E}_\lambda^*(\mathbf{R})\, a_\lambda^+ \,.\end{aligned} \qquad (2.6)$$

We will suppose that the initial state $|\psi_i\rangle$ corresponds to the atom in its ground level, N photons in the laser mode denoted λ_L and no photon in all the other radiation modes :

$$|\psi_i\rangle = |g, N\lambda_L\rangle\,. \qquad (2.7)$$

The strength of the atom laser coupling is characterized

by the Rabi frequency ω_1 :

$$\begin{aligned}\hbar\omega_1/2 &= \langle e, (N-1)\lambda_L\,|\,V\,|\,g, N\lambda_L\rangle \\ &= - \mathbf{d}.\mathcal{E}_L(0)\sqrt{N}\end{aligned} \qquad (2.8)$$

(ω_1 is related to the absorption and stimulated emission of laser photons). The characteristic constant of the coupling of the atom with the empty modes is the natural width Γ of the excited level :

$$\hbar\Gamma/2 = \pi \sum_{\omega_\lambda = \omega_0} |\langle g, \lambda\,|\,V\,|\,e\rangle|^2 \qquad (2.9)$$

(spontaneous emission rate).

3. Diagrammatic expansion of the diffracted wave packet.

We have calculated the final state vector $|\psi_f\rangle$ of the system using formal collision theory [20]. We don't detail these standard calculations [27] but rather give the results and discuss them.

Since the coupling V keeps the total number of excitations constant, the « post collision » state vector $|\psi_f\rangle$ can be expanded on the states where the atom is in its ground state in presence of N-n laser photons and n fluorescence photons in the initially empty modes. For a non saturating laser excitation, $(\omega_1 \ll \Gamma$ or $(\omega_L - \omega_0))$, such an expansion can be restricted to its first terms :

$$\begin{aligned}|\psi_f\rangle &= |g, N\lambda_L\rangle + \sum_\lambda S_1(\lambda)\,|g, (N-1)\lambda_L, \lambda\rangle + \\ &+ \sum_{(\lambda,\lambda')} S_2(\lambda, \lambda')\,|g, (N-2)\lambda_L, \lambda, \lambda'\rangle \end{aligned} \qquad (3.1)$$

where $S_1(\lambda)$ and $S_2(\lambda, \lambda')$ are the first and second order matrix elements of the S matrix corresponding respectively to the scattering of one and two laser photons. The expansion (3.1) is made more suggestive by the diagrammatic representation of figure 1. The first diagram (Fig. 1a) represents the zeroth order of (3.1) (no modification of the state). The other ones represent the first (Fig. 1b) and second (Fig. 1c) order terms.

3.1 FIRST ORDER TERMS. — We give now the expression of $S_1(\lambda)$ (obtained from the S matrix theory) and discuss its structure. $S_1(\lambda)$ is a product of four factors :

$$\begin{aligned}S_1(\lambda) &= [- \delta(\omega_\lambda - \omega_L)\, 2\,i\pi/\hbar]\,[\hbar\omega_1/2] \times \\ &\times [1/\hbar(\omega_L - \omega_0 + i\Gamma/2)]\,[- \mathbf{d}\mathcal{E}_\lambda^*(0)]\,. \end{aligned} \qquad (3.2)$$

The first factor expresses the energy conservation : it is proportional to $\delta(E_f - E_i)$ where E_i is the initial energy (here $E_i = E_g + N\hbar\omega_L$) and E_f the final one (here $E_f = E_g + (N-1)\hbar\omega_L + \hbar\omega_\lambda$).

The second factor is the matrix element of V between the initial state and the intermediate state in the first order diagram : it describes a laser photon absorption
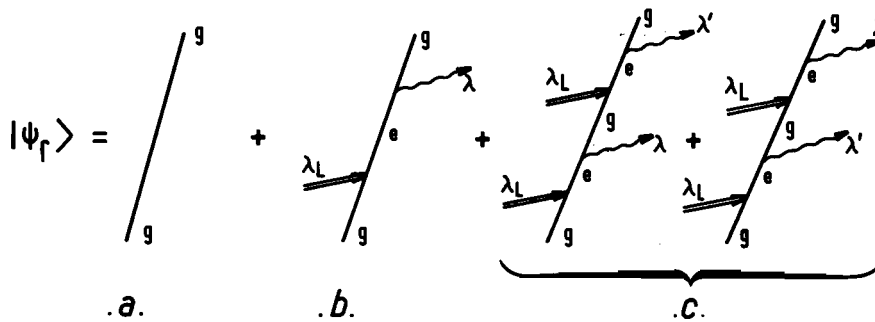
Fig. 1. — Diagrammatic expansion of the diffracted wave packet.

(see Fig. 1b). The third factor is the inverse of the energy difference between the initial state and the intermediate one : it describes the propagation of the system in this intermediate state. The last factor is the matrix element of V between the intermediate state and the final one and describes the spontaneous emission of a fluorescence photon in the mode λ (see Fig. 1b).

Note that a complex term $(-i\hbar\Gamma/2)$ has been added to the energy of the excited level e (see the third factor in (3.2)). Such a term stands for all the diagrams (such as the one of Fig. 2) where the atom in its excited state emits and reabsorbs fluorescence photons [8, 14, 15].

Remarks.

(i) These diagrams are known to produce also shifts of the atomic levels (Lamb shift) which are here included in the value of the resonance frequency ω_0. Note that the right value for the Lamb shift is obtained only if the two level and RWA approximations are dropped.

(ii) The term $S_1(\lambda_L)$ must be treated separately. The expression 3.2 is not correct for it (it must indeed be multiplied by $\sqrt{N}$). On the other hand, the corresponding contribution to $|\psi_f\rangle$ can be written $S_1(\lambda_L)|g, N\lambda_L\rangle$. When added to the zeroth order term, it ensures the normalization of $|\psi_f\rangle$ and describes absorption of laser photons. In the following, we will study pure fluorescence signals. Consequently, we will ignore this term and similar ones $S_2(\lambda, \lambda_L)$, $S_2(\lambda_L, \lambda_L)$...

3.2 Second order terms.

— For the calculation of $S_2(\lambda, \lambda')$, let us first emphasize that the two diagrams of figure 1c which correspond to the same initial and final states have to be taken into account.
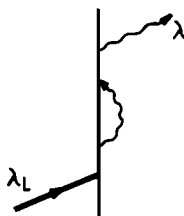

Fig. 2. — One-photon scattering process involving emission and reabsorption of a virtual fluorescence photon.

The numerator $\mathcal{N}$ is the same for both diagrams. It is the product of an energy conservation term and of four matrix elements.

$$\mathcal{N} = \left[- \delta(\omega_\lambda + \omega_{\lambda'} - 2\,\omega_L)\,2\,i\pi/\hbar\right] \times$$
$$\times\,(\hbar\omega_1/2)^2(d.\mathcal{E}_\lambda^*(0))\,(d.\mathcal{E}_{\lambda'}^*(0)). \quad (3.3)$$

For the first diagram, the denominator $\mathfrak{D}$ is :

$$\mathfrak{D} = \hbar^3(\omega_L - \omega_0 + i\Gamma/2)(\omega_L - \omega_\lambda + i\eta) \times$$
$$\times\,(2\,\omega_L - \omega_0 - \omega_\lambda + i\Gamma/2). \quad (3.4)$$

For the second diagram, the same expression holds with $\omega_{\lambda'}$ replacing ω_λ. Note the term $i\hbar\eta$ ($\eta \to 0^+$), added to the initial energy which gives sense to the second denominator :

$$1/(\omega_L - \omega_\lambda + i\eta) = -i\pi\delta(\omega_L - \omega_\lambda) +$$
$$+ PP(1/\omega_L - \omega_\lambda). \quad (3.5)$$

Using the energy conservation $(\omega_\lambda + \omega_{\lambda'} = 2\,\omega_L)$ and summing the two diagrams, one gets :

$$S_2(\lambda, \lambda') = S_1(\lambda)\,S_1(\lambda') + T_2(\lambda, \lambda') \quad (3.6)$$

with :

$$T_2(\lambda, \lambda') = \left[\delta(\omega_\lambda + \omega_{\lambda'} - 2\,\omega_L)\,i\pi/2\,\hbar^2\right] \times$$
$$\times\,(d.\mathcal{E}_\lambda^*(0))\,(d.\mathcal{E}_{\lambda'}^*(0))\,(\omega_1/(\omega_L - \omega_0 + i\Gamma/2))^2$$
$$\times\,\left[1/(\omega_\lambda - \omega_0 + i\Gamma/2) + 1/(\omega_{\lambda'} - \omega_0 + i\Gamma/2)\right]. \quad (3.7)$$

Remark.

Taking into account the reabsorption of virtual fluorescence photons seems now more complicated than for first order terms ; more complex processes indeed appear (see Fig. 3). Actually diagrams where emission and reabsorption of virtual photons are separated by other vertices (Fig. 3a) have a negligible contribution : one can show this general result by a summation on the virtual photon frequency. One is then left with figure 3b-type diagrams ; they are taken into account as previously, by adding the complex term $i\Gamma/2$ to the upper level energy.
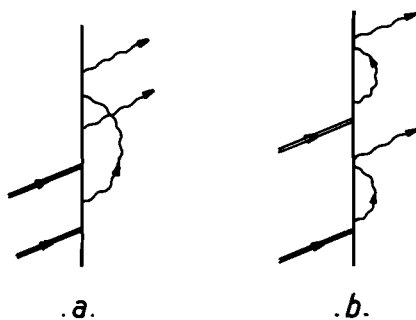
Fig. 3. — Two types of second order scattering process involving emission and reabsorption of virtual fluorescence photons.

4. Spectrum of the fluorescence light.

We now discuss the expressions of S_1 and S_2 obtained in section 3 and their respective contributions to the fluorescence spectrum.

Scattering of a single photon is described by the expression 3.2 of $S_1(\lambda)$. Energy conservation implies that the emitted photon has the same frequency as the laser ones ($\delta(\omega_\lambda - \omega_L)$ function in (3.2)). This clearly appears when the first order diagram of figure 1b is sketched in the way of figure 4. In other terms, there is no broadening due to the diffusion process (elastic Rayleigh scattering). The one photon scattering is linear ($S_1(\lambda)$ is proportional to ω_1, i.e. to the incident field amplitude) and has a resonant behaviour when the laser is tuned through the atomic frequency.

The two photon scattering amplitude $S_2(\lambda, \lambda')$ contains two terms (see Eq. 3.6). The first one, $S_1(\lambda) S_1(\lambda')$, describes the diffusion of two Rayleigh photons (Fig. 5a : both emitted photons have the laser frequency). The second term of (3.6) corresponds to the emission of a pair of photons such as :

$$\omega_\lambda + \omega_{\lambda'} = 2\,\omega_L \qquad (4.1)$$

(δ function in (3.7)). These photons are distributed in two lines of width Γ centred at ω_0 and $2\,\omega_L - \omega_0$ (function $1/(\omega_\lambda - \omega_0 + i\Gamma/2) + 1/(\omega_{\lambda'} - \omega_0 + i\Gamma/2)$ in (3.7)). Such an inelastic two photon scattering is sketched in figure 5b.

Recombining these results, one finds that the fluorescence spectrum is a symmetrical triplet consisting of an elastic line at ω_L and two sidebands of width Γ at ω_0 and $2\,\omega_L - \omega_0$. Taking into account third order diagrams such as the one of figure 6, one would also find an inelastic component of width $2\,\Gamma$ to the central line.
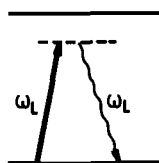

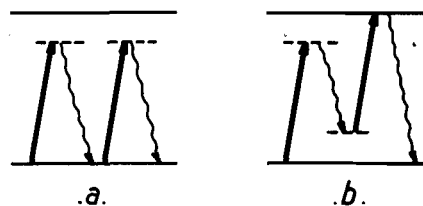Fig. 4. — First order elastic scattering process (Rayleigh diffusion).

Fig. 5. — Two photon scattering processes : (a) double Rayleigh scattering ; (b) non linear scattering process giving rise to the two sidebands of the fluorescence triplet.
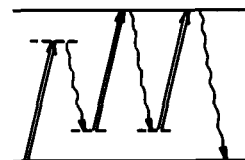

Fig. 6. — Third order scattering process leading to an inelastic central component.

5. Temporal correlation between fluorescence photons.

The signal given by a correlation set up is the probability $P(\mathbf{R}_A, \varepsilon_A, t_A ; \mathbf{R}_B, \varepsilon_B, t_B)\,dt_A\,dt_B$ for detecting a photon of polarization ε_A at point $\mathbf{R}_A$ between time t_A and $t_A + dt_A$ and a photon of polarization ε_B at point $\mathbf{R}_B$ between t_B and $t_B + dt_B$. One shows [19] that :

$$P = C \langle \psi_f \mid E_A^-(\mathbf{R}_A, t_A)\, E_B^-(\mathbf{R}_B, t_B)\, E_B^+(\mathbf{R}_B, t_B) \times$$
$$\times E_A^+(\mathbf{R}_A, t_A) \mid \psi_f \rangle \qquad (5.1)$$

where C is some multiplicative constant and $E_A^-(\mathbf{R}, t)$, $E_A^+(\mathbf{R}, t)$ (resp. E_B^-, E_B^+) the ε_A (resp. ε_B) polarization component of the negative and positive frequency parts of the electric field operator now taken in the Heisenberg point of view :

$$E_A^+(\mathbf{R}, t) = \sum_{\lambda \neq \lambda_L} \varepsilon_A . \mathcal{E}_\lambda(\mathbf{R})\, a_\lambda \exp(- i\omega_\lambda t) \qquad (5.2)$$

(compare with (2.6)). Note that we have excluded the laser mode in the sum 5.2 since we suppose that the detectors only see the fluorescence field but not the laser one.

In order to calculate P, one first remarks that it can be expressed as the norm of some vector $\mid \varphi \rangle$:

$$P = C \langle \varphi \mid \varphi \rangle \qquad (5.3)$$

with

$$\mid \varphi \rangle = \sum_{\lambda'} \alpha_B(\lambda')\, a_{\lambda'} \sum_\lambda \alpha_A(\lambda)\, a_\lambda \mid \psi_f \rangle \qquad (5.4)$$

where

$$\alpha_A(\lambda) = \varepsilon_A . \mathcal{E}_\lambda(\mathbf{R}_A) \exp(- i\omega_\lambda t_A) \qquad (5.5)$$

is the amplitude for detecting the λ photon on the detector A (polarization ε_A, position $\mathbf{R}_A$, time t_A).

Similar expression holds for $\alpha_B(\lambda')$, amplitude for detecting the λ' photon on the detector B. Coming back to (3.1), one remarks that :

$$a_\lambda \, a_{\lambda'} \, | \, \psi_f \, \rangle = S_2(\lambda, \lambda') \, | \, g, (N - 2) \, \lambda_L \, \rangle \, . \qquad (5.6)$$

The correlation signal can therefore be written :

$$P = C \left| \sum_\lambda \sum_{\lambda'} S_2(\lambda, \lambda') \, \alpha_A(\lambda) \, \alpha_B(\lambda') \right|^2 . \qquad (5.7)$$

This expression 5.7 has a simple interpretation. All the processes consisting in the scattering of two laser photons followed by the absorption of the two fluorescence photons by two detectors A and B have

indeed the same initial $| \, g, \, N\lambda_L, \, g_A, \, g_B \, \rangle$ and final states $| \, g, \, (N - 2) \, \lambda_L, \, e_A, \, e_B \, \rangle \, (g_A, \, g_B, \, e_A, \, e_B$ for the ground and excited states of the detecting atoms). So the correlation signal P is obtained by summing the amplitudes of all the quantum paths leading from this initial state to the final one.

To go further, one must now perform the angular integration of (5.7) (summation over shells of modes λ and λ' of frequencies ω_λ and $\omega_{\lambda'}$). As usually, retardation effects appear at this stage : the detection times t_A and t_B are replaced by the emission times :

$$\tau_A = t_A - R_A/c \, ; \quad \tau_B = t_B - R_B/c \, . \qquad (5.8)$$

More precisely, one gets :

$$P = C' \left| \iint d\omega_\lambda \, d\omega_{\lambda'} \, s_2(\omega_\lambda, \omega_{\lambda'}) \, e^{-i\omega_\lambda \tau_A} \, e^{-i\omega_{\lambda'} \tau_B} \right|^2 \qquad (5.9)$$

where $s_2(\omega_\lambda, \omega_{\lambda'})$ contains the frequency dependence of $S_2(\lambda, \lambda')$ (see Eqs. 3.6, 3.7 and 3.2) :

$$s_2(\omega_\lambda, \omega_{\lambda'}) = \delta(\omega_\lambda - \omega_L) \, \delta(\omega_{\lambda'} - \omega_L) \, +$$

$$\delta(\omega_\lambda + \omega_{\lambda'} - 2 \, \omega_L) \, (1/2 \, i\pi) \, (1/(\omega_\lambda - \omega_0 + i\Gamma/2) + 1/(\omega_{\lambda'} - \omega_0 + i\Gamma/2)) \, . \qquad (5.10)$$

The new constant C' is in particular proportional to $(\omega_1^2/((\omega_L - \omega_0)^2 + \Gamma^2/4))^2$.

A straightforward integration then gives :

$$P = C' \, | \, e^{-i\omega_L \tau} - \theta(-\tau) \, e^{-i(2\omega_L - \omega_0)\tau} \, e^{+\Gamma\tau/2} - \theta(\tau) \, e^{-i\omega_0\tau} \, e^{-\Gamma\tau/2} \, |^2 \qquad (5.11)$$

with :

$$\tau = \tau_B - \tau_A \qquad (5.12)$$

(θ is the Heaviside function). It is worth noting for the forthcoming discussion (sect. 7) that the three terms in (5.11) correspond respectively to the three contributions to s_2 in (5.10). So the first one is related to the Rayleigh scattering while the two others are associated with the inelastic two photon scattering (see sects. 3 and 4).

The correlation signal can finally be written from (5.11) :

$$P(\tau) = C'[1 + \exp(-\Gamma \, | \, \tau \, |) - 2 \cos((\omega_L - \omega_0) \, \tau) \exp(-\Gamma \, | \, \tau \, |/2)] \, . \qquad (5.13)$$

This result is identical to the perturbative limit $(\omega_1 \ll (\omega_L - \omega_0)$ or $\Gamma)$ of the well known expression obtained by the correlation function method [8, 9]. In particular, $P(\tau)$ is zero for $\tau = 0$ which indicates an antibunching of the fluorescence photons.

6. Temporal correlation between frequency filtered fluorescence photons.

One now supposes that frequency filters F_A and F_B are interposed between the emitting atom and the two detectors A and B. The expression of the correlation signal as a function of the electric field is the same as previously (see Eq. 5.1). But the electric field itself is modified by the filters :

$$E_A(R_A, t_A) = \sum_{\lambda \neq \lambda_L} G_A(\lambda) \, \alpha_A(\lambda) \, a_\lambda \qquad (6.1)$$

(compare with 5.2 and 5.5). The functions $G_A(\lambda)$ and $G_B(\lambda)$ characterize the two filters F_A and F_B.

The same demonstration as in section 5 leads to :

$$P(F_A, F_B) = C \left| \sum_\lambda \sum_{\lambda'} S_2(\lambda, \lambda') \, \alpha_A(\lambda) \, G_A(\lambda) \, \alpha_B(\lambda') \, G_B(\lambda') \right|^2 . \qquad (6.2)$$

This means that the amplitudes of all the quantum paths appearing in (5.7) are multiplied by a filtering amplitude

$G_A(\lambda)\, G_B(\lambda')$. The angular integration now gives :

$$P(F_A, F_B) = C' \left| \iint d\omega_\lambda \, d\omega_{\lambda'} \, s_2(\omega_\lambda, \omega_{\lambda'}) \, g_A(\omega_\lambda) \, g_B(\omega_{\lambda'}) \, e^{-i\omega_\lambda \tau_A} \, e^{-i\omega_{\lambda'}\tau_B} \right|^2 \qquad (6.3)$$

where $g_A(\omega_\lambda)$ is the filtering amplitude $G_A(\lambda)$ for the modes λ of frequency ω_λ directed from the emitter to the detector A.

From now on, we will consider a particular situation, where the three components of the spectrum are well separated, which occurs in the perturbative limit when :

$$\Gamma, \omega_1 \ll (\omega_L - \omega_0). \qquad (6.4)$$

We will furthermore suppose the spectral width Δv of the filters much larger than the width Γ of the sidebands :

$$\Gamma \ll \Delta v. \qquad (6.5)$$

Equation 6.3 can thus be integrated to :

$$P(F_A, F_B, \tau) = C' \left| e^{-i\omega_L \tau} \, g_A(\omega_L)\, g_B(\omega_L) - \theta(-\tau)\, e^{-i(2\omega_L - \omega_0)\tau} \, e^{\Gamma\tau/2} \, g_A(\omega_0)\, g_B(2\,\omega_L - \omega_0) \right.$$

$$\left. - \theta(\tau)\, e^{-i\omega_0\tau} \, e^{-\Gamma\tau/2} \, g_A(2\,\omega_L - \omega_0)\, g_B(\omega_0) \right|^2. \qquad (6.6)$$

When the bandwidth Δv of the filters is larger than the line splitting $(\omega_L - \omega_0)$, g_A and g_B have the same values for $\omega_L, \omega_0, (2\,\omega_L - \omega_0)$ and one gets the non filtered signal (see Eqs. 5.11 or 5.13). But, when :

$$\Delta v \ll (\omega_L - \omega_0) \qquad (6.7)$$

one can study temporal correlations between photons emitted in given lines of the spectrum [6].

One can for example study the correlation between Rayleigh photons $P(\omega_L, \omega_L, \tau)$ by considering two filters F_A and F_B selecting the Rayleigh component and rejecting the sidebands

$$g_A(\omega_L) = g_B(\omega_L) = 1$$

$$g_A(\omega_0) = g_A(2\,\omega_L - \omega_0) = g_B(\omega_0) = g_B(2\,\omega_L - \omega_0) = 0.$$

One gets from (6.6) :

$$P(\omega_L, \omega_L, \tau) = C' \qquad (6.8)$$

which means that the Rayleigh emissions are uncorrelated. Considering now two filters F_A and F_B selecting the two sidebands :

$$g_A(2\,\omega_L - \omega_0) = g_B(\omega_0) = 1 \,; \qquad g_A(\omega_L) = g_B(\omega_L) = g_A(\omega_0) = g_B(2\,\omega_L - \omega_0) = 0$$

one gets the correlation signal :

$$P(2\,\omega_L - \omega_0, \omega_0, \tau) = C'\, \theta(\tau)\, e^{-\Gamma\tau}. \qquad (6.9)$$

This means that the photons emitted in the two sidebands are strongly bunched, emitted in a given order (the $(2\,\omega_L - \omega_0)$ one before the ω_0 one) with an exponential decay law. These results are in complete agreement with the qualitative predictions deduced from the observation of figure 5b (see the discussion in [6]). As well, the absence of correlation between Rayleigh photons corresponds with the diagram of figure 5a.

One checks that there is no correlation between the central component and the sidebands (at the second order in the calculation of $|\psi_f\rangle$; the diagram of figure 6 suggests that such correlations appear at the third order).

7. Discussion.

When interested in photon correlation signals, one has to develop the post collision state $|\psi_f\rangle$ at least up to the second order (measurement of this signal involves destruction of two fluorescence photons). The corresponding diagrams (Fig. 1c) present two resonances. The first one consists in the emission of two Rayleigh photons (Fig. 5a). The second resonance is a non linear one : absorption of two laser photons and emission of a pair of fluorescence photons whose frequencies are distributed around $2\,\omega_L - \omega_0$ and ω_0 respectively (Fig. 5b).

When studying time correlations between frequency filtered photons, one has to keep only the diagram corresponding to the detected photons. For Rayleigh-Rayleigh correlations, one keeps only diagram 5a, and finds $P(\tau) = C'$ (formula 6.7). For correlations between the two sidebands of the triplet, the relevant diagram is that of figure 5b leading to $P(\tau) = C' \theta(\tau) \times \exp(-\Gamma\tau)$ (formula 6.8).

When frequency filters are removed, all the diagrams contribute to the signal. The correlation function then contains three terms (see (5.12)); the first one corresponds to Rayleigh-Rayleigh correlation, the second one to sidebands correlation, and the third one appears as an interference between the two previous terms, cancelling exactly $P(\tau)$ for $\tau = 0$ (antibunching effect).

In conclusion of this paper, we will risk some remarks concerning non classical features that resonance fluorescence is known to reveal. The antibunch-ing of unselected photons and the bunching of photons emitted in the two sidebands are such features [21-25]. It has sometimes been argued that these quantum properties can be entirely attributed to the emitting atom and give no evidence for the quantum nature of the field [26]. However, in this paper, these properties are calculated by averaging some pure field operator in a pure field state (the « post collision » state $|\psi_f\rangle$ can be factorized as a product of a field state and of the atomic state $|g\rangle$). In our opinion, such an approach strongly supports their interpretation as manifestations of the quantum nature of the electromagnetic field.

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