GENERAL THEORY OF INTENSITY CORRELATION ON LIGHT SCATTERING

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ABSTRACT

We develop a general theory for spatio-temporal intensity correlations measurements for a scattered beam. A completely quantum mechanical description for both excitation and detection set up is used. Our description is essentially valid for weak incident light beams and single photon absorption processes. We describe from a unified point of view both, stationary as well as, time resolved experiments. We emphasize the interest of such experiments in the study of processes like resonance raman scattering and resonance fluorescence. Also we obtain an observable coherent contribution associated to different final levels of the target— atoms or molecules - a result which cannot be reached by intensity measurements.

I. Introduction

Much of the information of microscopic phenomena is derived from an analysis of scattering experiments for both time resolved and stationary situations. The most relevant quantities usually measured are the decay laws and the scattering cross sections respectively. These observations are obtained from the scattered intensity measurement and correspond to first order properties of the field, so that we generally measure the quantity

$$\langle \mathcal{E}^*(z,t)\, \mathcal{E}(z,t) \rangle \qquad (I.1)$$

which means the average value of the electric field at a given point. In photophysics experiments the photons are detected ordinarily by the use of any absorption process so that the field we are measuring corresponds to the photon annihilation by the detection operator Ψ. Therefore, the probability, that a photon is absorbed by the detector at point z at time t, is proportional to $\langle | \Psi^+(z,t)\Psi(z,t) | \rangle$ where $|\rangle$ means the state of the field before detection. This expression of the detection probability determines the average photon number scattered and is a particular form of a more general type of expression

$$\langle N(z,t) \rangle = \mathrm{Tr}\left[\rho\, \Psi^+(z,t)\, \Psi(z,t)\right] \qquad (I.2)$$

valid for pure, as well as, mixed states. The symbol Tr stands for the trace and ρ means the density matrix of the field. Up to now we have dealt with experiments involving one detector [1], thus, limiting ourselves to consideration of the field scattered at a single spatio-temporal point.

While the conventional methods for investigating scattered intensities are based on intensity measurements, new possibilities are arisen which are related to more general statistical descriptions of the radiation field in terms of higher order correlation functions [2,3]. These functions have been recognized as the most natural mathematical tool for describing the statistical properties of the radiation emitted by light sources. As a basic example the fundamental differences between a laser beam and a beam emitted by a thermal source only emerge when considering intensity correlations of order higher than first. Therefore, besides these experiments performed with a single detector, there are other measurements in which the observable quantities are connected with correlation functions of the scattered beam [4,5]. If we introduce a second type of measurement which consists in counting photons with two detectors located at different spatio-temporal point (z_1,t_1) and (z_2,t_2), the detection probability will be proportional to

$$\langle N(z_1,t_1)N(z_2,t_2)\rangle = \mathrm{Tr}\left[\rho\, \Psi^+(z_1,t_1)\Psi^+(z_2,t_2)\Psi(z_1,t_1)\Psi(z_2,t_2)\right] \qquad (I.3)$$

Up to now, little attention has been given to actual experiments for making measurement of spatial or temporal correlations on scattered beam. The suggestion of measuring intensity correlation which corresponds to second order properties of the scattered beam, in order to obtain information not available with a first order intensity experiment was made by Goldberger et al.[6,7]. These measurements of correlation evaluated at different spatio-temporal points can be realized by recording coincidences between counters placed at different positions.

It is the aim of this work to emphasize the interest for performing correlation measurement. We show that, with stationary excitation conditions, we can reach from correlation function similar information about the target dynamics usually obtained from time resolved situations by intensity measurements. This results from the ability of introducing the time dependence via the time delay between the detectors. Therefore, the correlation measurements can be of great interest in the study of the competitive process like resonance raman scattering and resonance fluorescence.

In this paper, we introduce a completely quantum mechanical description of the light beam and detection set-up. In the same way that the intensity measurement considers in the simplest case the single photon scattering, the correlation measurements imply just a pair of photons. This is why the light beam is described by two single photon wave packet. A more realistic description and his peculiar features will be studied in a next paper. The cases of time and energy resolved experiments are derived from a unified point of view following the same procedure described in our previous work [1]. It results from the continuous variation of the spatial localization dispersion of the photons in the incident beam. Therefore, our description will be valid essentially for weak incident light beams. Only processes involving single photon absorption and emission are considered here. The description of more complicated processes is straightforward and is not relevant in the present work. The target will be described in terms of resonances. Therefore, the expressions of the correlation functions are free from some specific model of the target. The detection set-up is constituted by two detectors. It is described by a quantum two particles detection operator. Next, we give the general expressions of the intensity correlation functions for the scattered beam in both stationary and time resolved situations. One of them has been used by us previously [8] to interpret the experimental result of Kimble et al [9].

II. Description of the free light beam

The description of the light beam has been extensively discussed in a first paper. In order to make this paper sufficiently self contained we summarize the relevant results.

The hamiltonian of the radiation field can be expressed as

$$H_F = \sum_{\underset{\sim}{k},\underset{\sim}{e}} k \; a^+_{\underset{\sim}{k}\underset{\sim}{e}} \, a_{\underset{\sim}{k}\underset{\sim}{e}} \qquad (\text{II}\cdot 1)$$

if we consider the particular units $\hbar = c = 1$. The variable k means the energy of the eigenstate $|\underset{\sim}{k},\underset{\sim}{e}>$ of the free radiation field. It describes a photon with wave vector $\underset{\sim}{k}$ and polarization vector $\underset{\sim}{e}$. The operators $a^+_{\underset{\sim}{k}\underset{\sim}{e}}$ and $a_{\underset{\sim}{k}\underset{\sim}{e}}$ are respectively the creation and annihilation operator of such photon. We omit in the following all the information resulting from the polarization. Let us recall now, the fundamental properties. The operators $a^+_{\underset{\sim}{k}}$ and $a_{\underset{\sim}{k}}$ satisfy the commutation rules

$$\begin{aligned} \left[a_{\underset{\sim}{k}} \, , \, a^+_{\underset{\sim}{k}'} \right] &= \delta(\underset{\sim}{k}-\underset{\sim}{k}') \\ \left[a_k \, , \, a_{k'} \right] &= \left[a^+_k \, , \, a^+_{k'} \right] = 0 \end{aligned} \qquad (\text{II}\cdot 2)$$

The eigenstates of H_F are built up by successive applications of $a^+_{\underset{\sim}{k}}$

$$|\underset{\sim}{k}\rangle = a_k^+ |0\rangle \qquad (II\cdot 3)$$

if $|0\rangle$ means the ground state of the field. We have also the orthonormalization property

$$\langle \underset{\sim}{k} | \underset{\sim}{k'} \rangle = \delta(\underset{\sim}{k}-\underset{\sim}{k'}) \qquad (II\cdot 4)$$

These states do not represent realistic states of the field. In fact, a real light beam contains quantum as well as classical uncertainties. The implications of the detailed features of the statistical properties of the beam will be discussed in the next paper. For the sake of convenience, we consider here, a well collimated in the Z direction and polarized beam. Therefore, the light beam can be described in terms of single photon wave packet with a spectral distribution

$$f(k) = f(k-k_o, \Gamma_k) \qquad (II\cdot 5)$$

centered at k_o with a spectral linewidth Γ_k. The distribution $f(k)$ is normalized

$$\sum_k |f(k)|^2 = 1 \qquad (II\cdot 6)$$

These single photon wave packet are localized so that we define

$$|\Phi_1(Z)\rangle = b^+(Z)|0\rangle \qquad (II\cdot 7)$$

with the operator

$$b^+(Z) = \sum_k f(k)\, e^{-ikZ}\, a_k^+ \qquad (II\cdot 8)$$

satisfying the commutations rules

$$\begin{aligned} &[b^+(Z_j), b^+(Z_i)] = [b(Z_j), b(Z_i)] = 0 \\ &[b(Z_j), b^+(Z_i)] = \sum_k |f(k-k_o,\Gamma_k)|^2\, e^{ik(Z_j-Z_i)} = v(Z_j-Z_i) \end{aligned} \qquad (II\cdot 9)$$

The spatial incoherence is accounted for by $\mathcal{P}_2^S(Z_1,Z_2)$ that is, the probability to localize two photons in Z_1 and Z_2. Therefore, a two photon state is defined by

$$|\Phi_2(Z_1, Z_2)\rangle = \frac{1}{[C_2(Z_1-Z_2)]^{1/2}} \prod_{j=1}^{2} b^+(Z_j)|0\rangle \qquad (II\cdot 10)$$

where $C_2^{1/2}$ is the normalization factor

$$C_2(Z_1, Z_2) = \sum_P \prod_{j=1}^{2} v(Z_j - Z_{P_j}) \qquad (II\cdot 11)$$

if $\sum_P$ means the sum over all the permutations. The density matrix of a beam, constituted by an incoherent mixing of two single-photon-wave-packets, can be expressed by

$$\rho_2 = \sum_{\{Z\}} \mathcal{P}_2^s(\{Z\}) \, | \phi_2(\{Z\}) \rangle \langle \phi_2(\{Z\}) | \qquad (II\cdot 12)$$

For classical sources, with a good choice of the origin, we can take for the position Z_j of the photons

$$Z_j(t) = c(t - \tau_j) \quad ; \quad c = 1 \qquad (II\cdot 13)$$

and we express $\mathcal{P}_2^s(\{Z\})$ as a function of $\{t-\tau_j\}$, that is, $\mathcal{P}_2^s(\{t-\tau_j\})$. We must bear in mind that the distribution $\mathcal{P}_2^s(\{t-\tau_j\})$, which characterize the source, is independent of the observation time t_0. Then, we can express (II.12) as

$$\rho_2 = \mathcal{P}_2^s(\{t\}) * \rho_2(\{t\}) \qquad (II\cdot 14)$$

with

$$\rho_2(\{t\}) = | \phi_2(\{t\}) \rangle \langle \phi_2(\{t\}) | \qquad (II\cdot 15)$$

that is, as a double convolution product. Here the symbol $*$ means the convolution product $f * g = \int_{-\infty}^{t} f(t-\tau) g(\tau) d\tau$. Also we have assumed a continuous distribution of the parameters $\{\tau\}$.

Up to now, we have described the light beam. Following Mandel [10] we introduce the description of the detector. The detector operator in the z direction, for a light beam of extension L, is defined by

$$\Psi(z) = \frac{1}{L^{1/2}} \sum_k e^{ikz} a_k \qquad (II\cdot 16)$$

which satisfied the commutation rules

$$\begin{aligned} [\Psi(z), \Psi^+(z')] &= \frac{1}{L} \sum_k e^{ik(z-z')} \\ [\Psi(z), \Psi(z')] &= [\Psi^+(z), \Psi^+(z')] = 0 \end{aligned} \qquad (II\cdot 17)$$

Therefore, from the photon number operator

$$N_{1,1} = \int_1 dz\, \Psi^+(z)\Psi(z) \qquad (II.18)$$

we define the two photon operator needed for correlation measurements

$$N_{1_1,1_2,2} = \int_{1_1} dz_1 \int_{1_2} dz_2\, \Psi^+(z_1)\Psi(z_1)\Psi^+(z_2)\Psi(z_2) \qquad (II.19)$$

on the spatial extension 1_1 and 1_2. The intensity correlation detected will be proportional to the relation

$$\langle N_{1_1,1_2,2}\rangle = \mathrm{Tr}\left[\rho\, N_{1_1,1}\, N_{1_2,1}\right] \qquad (II.20)$$

where the trace must be evaluated over the beam states. Because of the delayed transient response of the detection set-up [7], the recorded observation is smeared over a time interval. Therefore, the expressions of the intensity correlation measurements will be modified by the response functions $\mathcal{P}_1^{d_1}$ $\mathcal{P}_1^{d_2}$ of the detectors, so that, we introduce the correlations of the counting rates by

$$\langle J_{1_1,1}(t_1)\, J_{1_2,1}(t_2)\rangle = \int dt'_1\, \mathcal{P}_1^{d_1}(t_1-t'_1)\int dt'_2\, \mathcal{P}_1^{d_2}(t_2-t'_2) \times \langle N_{1_1,1}(t'_1)\, N_{1_2,1}(t'_2)\rangle \qquad (II.21)$$

If we measure the particle flux with special detectors which have flat surfaces of area S_1 and S_2, thickness 1_1 and 1_2 and which are uniformely sensitive, the intensity correlation is given by

$$\langle I_{\Delta_1}(t_1)\, I_{\Delta_2}(t_2)\rangle = S_1 S_2\, \langle J_{1_1,1}(t_1)\, J_{1_2,1}(t_2)\rangle \qquad (II.22)$$

where $\Delta_i = 1_i S_i$ and $c = 1$ is the constant speed of the photons. With the use of relations (II.14, II.22) we obtain for the detected intensity correlation the general expression

$$\langle I_{\Delta_1}(t_1)\, I_{\Delta_2}(t_2)\rangle = S_1 S_2\, \mathcal{P}_1^{d_1}(t_1)\, \mathcal{P}_1^{d_2}(t_2) * \mathrm{Tr}\left[\rho_2(\{t\})\, N_{1_1,1}(t_1)\, N_{1_2,1}(t_2)\right] * \mathcal{P}_2^{s}(\{t\}) \qquad (II.23)$$

Therefore, the intensity correlation detected can be expressed as a double convolution products of the time responses of the detectors times the free evolution of the light beam times the excitation of the source. The general expression (II.23) can only be explicited by introducing particular models for both excitation and detection processes.

III. Detection of the free light beam

Now, we treat explicitely the detection of the free light beam constituted by two single photon wave packets. Both cases of stationary and pulsed experiments are considered. With the previous hypothesis, the correlation measurements of the photon number detected for a two photon beam is given by

$$\langle N(z_1,t+T_1)\,N(z_2,t+T_2)\rangle_2 = \mathrm{Tr}\left[\rho_2(t)\,N(z_1,T_1)\,N(z_2,T_2)\right] \qquad (\mathrm{III}\cdot 1)$$

in the particular cases of two ideal detectors located at the spatio-temporal points (z_1,T_1) and (z_2,T_2). Under these assumptions the density matrix $\rho_2(t)$ takes the form

$$\rho_2(t)=\int\prod_{j=1}^{2}d\tau_j\,\mathcal{P}_2^{s}(\{\tau\})\prod_{j=1}^{2}b^{+}(t-\tau_j)|0\rangle\langle 0|\prod_{j=1}^{2}b(t-\tau_j)\;\;C_2^{-1}(\{\tau\}) \qquad (\mathrm{III}\cdot 2)$$

because, with the relation (II.11) we find

$$C_2(\{\tau\}) = 1 + v(\tau_1-\tau_2)\,v(\tau_2-\tau_1) \qquad (\mathrm{III}\cdot 3)$$

for the normalization coefficient. The general expression for the photon density correlation is given by (II.23) and we must evaluate the quantity

$$\langle N(z_1,t+T_1)N(z_2,t+T_2)\rangle = \langle \Psi^{+}(z_1,T_1)\Psi(z_1,T_1)\Psi^{+}(z_2,T_2)\Psi(z_2,T_2)\rangle \qquad (\mathrm{III}\cdot 4)$$

With the commutation rules of the detection operators we find

$$\left[\Psi(z_1,T_1),\Psi^{+}(z_2,T_2)\right] = \sum_k e^{ik(z_1-T_1-z_2+T_2)} \qquad (\mathrm{III}\cdot 5)$$

recalling here that we have $\hbar = c = 1$. If we consider commonly encountered experimental situations so that $z_1-z_2 \neq T_1-T_2$, which correspond to the disjoint case of detection, we obtain [10]

$$[\Psi(z_1,T_1), \Psi^+(z_2,T_2)] = 0 \qquad \text{(III·6)}$$

which provides a more tractable expression for the two particle operator

$$N(z_1,T_1)N(z_2,T_2) = \Psi^+(z_1,T_1)\Psi^+(z_2,T_2)\Psi(z_1,T_1)\Psi(z_2,T_2) \qquad \text{(III·7)}$$

If the previous assumptions are accounted for, we have for the photon density correlation

$$\langle N(z_1,t_1)N(z_2,t_2)\rangle = \int \prod_{j=1}^{2} d\tau_j \, \mathcal{P}_2^s(\{\tau\}) \;\; B \;\; C_2^{-1}(\{\tau\}) \qquad \text{(III·8)}$$

where $t_j = t + T_j$ and B means the quantity

$$B = \langle 0| \prod_{j=1}^{2} b(t-\tau_j)\Psi^+(z_1,T_1)\Psi^+(z_2,T_2)\Psi(z_1,T_1)\Psi(z_2,T_2)\prod_{j=1}^{2} b^+(t-\tau_j)|0\rangle \qquad \text{(III·9)}$$

that we have to evaluate. This can be done by calculating

$$\Psi(z_2,T_2)\prod_{j=1}^{2} b^+(t-\tau_j)|0\rangle = \sum_{i=1}^{2} \phi(z_2-t_2+\tau_i)\prod_{j\neq i} b^+(t-\tau_j)|0\rangle \qquad \text{(III·10)}$$

Let

$$\phi(z_2-t_2+\tau_i) = [\Psi(z_2,T_2), b^+(t-\tau_i)] = \frac{1}{L^{1/2}}\sum_k e^{ik(z_2-t_2+\tau_i)} f(k) \qquad \text{(III·11)}$$

with the help of this notation we can write

$$\Psi(z_1,T_1)\Psi(z_2,T_2)\prod_{j=1}^{2} b^+(t-\tau_j)|0\rangle$$
$$= \sum_{i=1}^{2} \phi(z_2-t_2+\tau_i)\sum_{j\neq i} \phi(z_1-t_1+\tau_j)|0\rangle \qquad \text{(III·12)}$$

In order to obtain the expression of B, we multiply (III.12) by its complexe conjugate. Therefore, B can be expressed into the form

$$B = \sum_{i=1}^{2}\sum_{j\neq i}\left[\langle z_1,t_1 ; z_2,t_2\rangle_{di} + \langle z_1,t_1 ; z_2,t_2\rangle_{ex}\right] \qquad \text{(III·13)}$$

that is, like a sum of a direct term

$$\langle z_1,t_1 ; z_2,t_2\rangle_{di} = |\phi(z_2-t_2+\tau_i)\,\phi(z_1-t_1+\tau_j)|^2 \qquad \text{(III·14)}$$

and an exchange term

$$\langle z_1,t_1\,;\,z_2,t_2\rangle_{ex} = \phi^*(z_2-t_2+\tau_j)\,\phi(z_1-t_1+\tau_j)\,\phi^*(z_1-t_1+\tau_i)\,\phi(z_2-t_2+\tau_i) \qquad (\mathrm{III}\cdot 15)$$

The direct term represents just the probability of detecting two non correlated photons by two detectors, that is to say, one photon per detector. The possibility to detect both photons with the same detector is obviously rejected because in this case one of the probability cancels it. On the contrary the exchange term corresponds to a product of two factors. Each one represents an exchange probability and corresponds to crossed terms of the detection probability amplitude that a photon be detected either by detector 1 or 2. The existence for such terms has been extensively discussed in the literature in relation with the studies of the statistical properties of the light beams.

In the following, we assume that we can factorize the distribution probability, so that

$$\mathcal{P}_2^s(\{\tau\}) = \prod_{j=1}^{2} \mathcal{P}_1^s(\tau_j) \qquad (\mathrm{III}\cdot 16)$$

and that the one photon distribution are normalized

$$\int d\tau_j\, \mathcal{P}_1^s(\tau_j) = 1 \qquad (\mathrm{III}\cdot 17)$$

With these two assumptions, the expression of the photon density correlation is given by

$$\langle N(z_1,t_1)N(z_2,t_2)\rangle_2 = \sum_{i=1}^{2}\sum_{j\neq i}\iint d\tau_i\, d\tau_j\, C_2^{-1}(\{\tau\})\,\mathcal{P}_1^s(\tau_i)\,\mathcal{P}_1^s(\tau_j) \times\left[\langle z_1,t_1\,;\,z_2,t_2\rangle_{di} + \langle z_1,t_1\,;\,z_2,t_2\rangle_{ex}\right] \qquad (\mathrm{III}\cdot 18)$$

In fact, it results from the integration over the current subscripts i and j that all the summation are equivalent, so that, the photon density correlation can be written

$$\langle N(z_1,t_1)N(z_2,t_2)\rangle_2 = 2\left[\langle N(z_1,t_1)N(z_2,t_2)\rangle_{2,di} + \langle N(z_1,t_1)N(z_2,t_2)\rangle_{2,ex}\right] \qquad (\mathrm{III}\cdot 19)$$

where we have separated the contributions of the direct term

$$\langle N(z_1,t_1)\,N(z_2,t_2)\rangle_{2,di} = \int d\tau_i\, \mathcal{P}^s_1(\tau_i)\,|\,\phi(z_2-t_2+\tau_i)|^2$$
$$\times \int d\tau_j\, \mathcal{P}^s_1(\tau_j)\,|\,\phi(z_1-t_1+\tau_j)|^2\ C_2^{-1}(\{\tau\}) \qquad (III\cdot 20)$$

and exchange term

$$\langle N(z_1,t_1)\,N(z_2,t_2)\rangle_{2,ex} = \int d\tau_i\, \mathcal{P}^s_1(\tau_i)\ \phi^*(z_1-t_1+\tau_i)\ \phi(z_2-t_2+\tau_i) \qquad (III\cdot 21)$$
$$\times \int d\tau_j\, \mathcal{P}^s_1(\tau_j)\ \phi^*(z_2-t_2+\tau_j)\,\phi(z_1-t_1+\tau_j)\ C_2^{-1}(\{\tau\})$$

When the normalization coefficient is not τ dependent, we must note that the direct term can be expressed as a product of the photon densities at the spatio-temporal points (z_1,t_1) and (z_2,t_2) for the single photon wave packet beams, that is

$$\langle N(z_1,t_1)\,N(z_2,t_2)\rangle_{2,di} = \langle N(z_1,t_1)\rangle_1\ \langle N(z_2,t_2)\rangle_1 \qquad (III\cdot 22)$$

The exchange term is responsible for correlation resulting of the partial overlapping of the photon wave packets. In effect, the photons behave as indistinguishable particules and this implies a tendency toward bunching [11-13].

Let us analyze now two extreme experimental situations and we begin by the stationary case. This situation is depicted by considering a random spatial or temporal photon distribution and can be characterized by

$$\mathcal{P}^s_1(\tau) = \frac{1}{L} \qquad (III\cdot 23)$$

In this case, the normalization coefficient is not τ-dependent. Moreover, if the normalization condition (III.18) is accounted for, we obtain for the direct term

$$\langle N(z_1,t_1)\,N(z_2,t_2)\rangle_{2,di} = \frac{1}{L^2} \qquad (III\cdot 24)$$

while for the exchange term we have

$$\langle N(z_1,t_1)\,N(z_2,t_2)\rangle_{2,ex} = \frac{1}{L^2}\int d\tau_1\,\phi^*(z_1-t_1+\tau_1)\,\phi(z_2-t_2+\tau_1)$$
$$\times\int d\tau_2\,\phi^*(z_2-t_2+\tau_2)\,\phi(z_1-t_1+\tau_2) \qquad \text{(III·25)}$$

The opposite situation corresponding to a time resolved experiment needs a good localization of the photons so that we take

$$\mathcal{P}_1^{6}(\tau) = \delta(\tau) \qquad \text{(III·26)}$$

The direct and exchange terms are given now by

$$\langle N(z_1,t_1)N(z_2,t_2)\rangle_{2,di} = |\phi(z_1-t_1)|^2\,|\phi(z_2-t_2)|^2$$
$$\langle N(z_1,t_1)\,N(z_2,t_2)\rangle_{2,ex} = \phi^*(z_1-t_1)\,\phi(z_2-t_2)\,\phi^*(z_2-t_2)\,\phi(z_1-t_1) \qquad \text{(III·27)}$$

The relations (III.24,25) and (III.22) have been extensively discussed in relation with the study of the bunching effect, a phenomenon which has not been completely explained till now [14].

IV. Formal description of the target dynamics

The detection of the free light beam has been described for correlation measurements. It is the goal of the quantum optics to analyze the dependence of these functions in order to understand the nature and the behavior of the light beams. As we are interested by the knowledge of the target dynamics, we must study these corresponding second order correlation function for the scattered beam. It brings part of the information that we can expect to extract from a photophysics experiment. The correlation function for a two single photon wave packet state can be written here as

$$\langle N(z_1,t_1)\,N(z_2,t_2)\rangle_2 = \mathrm{Tr}\left[\rho_2^{sc}(t)\,N(z_1,t_1)N(z_2,t_2)\right] \qquad \text{(IV·1)}$$

where $\rho_2^{SC}(t)$ is the density matrix of the scattered beam. It is given by

$$\rho_2^{sc}(t) = e^{-iH(t-t_0)}\left[\rho_2(t_0)\,\rho_t(t_0)\right]e^{iH(t-t_0)} \qquad \text{(IV·2)}$$

if we assume at the initial time t_o the factorization of the density matrix $\rho_2(t_o)$ of the beam alone by the one $\rho_t(t_o)$ of the target. Here H means the total hamiltonian of the system beam-target. If we note $|g>$ the ground states of the target, that is, states which cannot be reached by the excitation, the ground states of the total system are given by $|g,0>$. Therefore, $\rho_2^{sc}(t)$ can be expressed as

$$\rho_2^{sc}(t) = \int \prod_{j=1}^{2} d\tau_j \, \mathcal{P}_2^{s}(\{\tau\}) \, C_2^{-1}(\{\tau\}) \times \prod_{j=1}^{2} \overset{+}{b}(t-\tau_j) \, e^{-iH(t-t_o)} \, |g,0><g,0| \, e^{iH(t-t_o)} \prod_{j=1}^{2} b(t-\tau_j) \qquad (IV\cdot3)$$

where

$$\overset{+}{b}(t-\tau_j) = e^{-iH(t-t_o)} \, \overset{+}{b}(t_o-\tau_j) \, e^{iH(t-t_o)} \qquad (IV\cdot4)$$

All the dynamics is contained in the total hamiltonian H. In order to obtain completely general expressions for the correlation functions, we characterize the target by its resonances. When H is not time dependent, we introduce the integral representation of the evolution operator

$$U_+(t-t_o) = \frac{i}{2\pi} \int_{-\infty}^{+\infty} dE \, e^{-iE(t-t_o)} \, G_+(E) \qquad (IV\cdot5)$$

where

$$G_+(E) = \lim_{\epsilon \to 0^+} [E - H + i\epsilon] \qquad (IV\cdot6)$$

The spectral decomposition of G(E) is obtained by the introduction of some convenient partitioning technique [15]. Let us consider P and Q two appropriate projection operators so that

$$P + Q = 1 \qquad (IV\cdot7)$$

G(E) can be expressed in terms of the reduced resolvents whose expression have been given by Mower [15]

$$\begin{aligned} PG(E)P &= [E - PH_oP - PR(E)P]^{-1} P \\ QG(E)Q &= [E - QHQ]^{-1} [1 + QH'P \, PG(E)P \, PH'Q \, (E - QHQ)^{-1}] \, Q \end{aligned} \qquad (IV\cdot8)$$

where the level shift operator R(E) is defined by

$$R(E) = H' + H'Q[E-QHQ]^{-1}QH' \qquad (IV.9)$$

if we consider the partition of H in model hamiltonian H_o and residual interaction H', that is

$$H = H_o + H' \qquad (IV.10)$$

All the reduced resolvents are depending on PG(E)P which can be written as

$$PG(E)P = P \frac{1}{E - H_{PP}} P \qquad (IV.11)$$

where

$$H_{PP} = P[H_o + R(E)]P \qquad (IV.12)$$

acts like an effective hamiltonian in the subspace spanned by P. We introduce also the biorthogonal basis set [15] built up from the eigenstates of H_{PP}

$$H_{PP}|\beta\rangle = E_\beta|\beta\rangle \qquad (IV.13)$$

so that

$$\langle\tilde{\beta}|\beta'\rangle = \delta_{\beta\beta'}$$

$$\sum_\beta |\beta\rangle\langle\tilde{\beta}| = 1 \qquad (IV.14)$$

Therefore, the matrix elements of PG(E)P will be expressed by a Mittag Leffler expansion given by

$$\langle s|PG(E)P|s'\rangle = \sum_\beta \frac{\langle s|\beta\rangle\langle\tilde{\beta}|s'\rangle}{E - E_\beta} \qquad (IV.15)$$

$$E_\beta = \Delta_\beta - i\,\Gamma_\beta/2$$

which shows that the discrete eigenvalues E_β of the effective hamiltonian are the poles of $\langle s|PG(E)P|s'\rangle$ with the corresponding residues $\langle s|\beta\rangle\langle\tilde{\beta}|s'\rangle$. We obtain a very practical sum rule

$$\sum_{\beta} \langle s|\beta\rangle\langle\tilde{\beta}|s'\rangle = \delta_{ss'} \qquad (\text{IV}\cdot 16)$$

It results that each pole defines a resonance whose contribution to $\langle s|PG(E)P|s'\rangle$ is proportional to the corresponding residue.

V. Correlation of the scattered light beam

Up to now, we have dealt with measurements on the free light beam. We wish to describe here the intensity correlation measurements on the scattered beam. As we have seen before, the main quantity is the correlation of the photon density at different spatio-temporal points and we must evaluate it. We express the photon density correlation as

$$\langle N(z_1,t_1)N(z_2,t_2)\rangle_2 = \int \prod_{j=1}^{2} d\tau_j \, \mathcal{P}_2^{s}(\{\tau\})\, C_2^{-1}(\{\tau\})\; B \qquad (\text{V}\cdot 1)$$

where B means

$$B = \langle g,0| \prod_{j=1}^{2} b(t_0-\tau_j)\, e^{iH(t-t_0)}\, \psi^{+}(z_1,T_1)\,\psi^{+}(z_2,T_2)$$
$$\times\, \psi(z_1,T_1)\,\psi(z_2,T_2)\, e^{-iH(t-t_0)} \prod_{j=1}^{2} b^{+}(t_0-\tau_j)\,|g,0\rangle \qquad (\text{V}\cdot 2)$$

If we introduce the notation

$$[j(1)] = [\psi(z_1,T_1),\, b^{+}(t-\tau_j)] \qquad (\text{V}\cdot 3)$$

we obtain by successive applications of the detection operator

$$\psi(z_2,T_2)\, e^{-iH(t-t_0)} \prod_{j=1}^{2} b^{+}(t_0-\tau_j)\,|g,0\rangle$$
$$= \sum_{i=1}^{2} [i(2)]\, e^{-iH(t-t_0)} \prod_{j\neq i} b^{+}(t_0-\tau_j)\,|g,0\rangle \qquad (\text{V}\cdot 4)$$

and

$$\psi(z_1,T_1)\,\psi(z_2,T_2)\, e^{-iH(t-t_0)} \prod_{j=1}^{2} b^{+}(t_0-\tau_j)\,|g,0\rangle$$
$$= \sum_{i=1}^{2} [i(2)] \sum_{j\neq i} [j(1)]\, e^{-iH(t-t_0)}\,|g,0\rangle \qquad (\text{V}\cdot 5)$$

Therefore, the expression of B can be written as

$$B = \sum_{i=1}^{2} \sum_{j\neq i} \left[B_{2,di} + B_{2,ex} \right] \tag{V·6}$$

with

$$\begin{aligned} B_{2,di} &= \langle g,0| e^{iH(t-t_0)} [j(1)]^+ [i(2)]^+ [i(2)] [j(1)] e^{-iH(t-t_0)} |g,0\rangle \\ B_{2,ex} &= \langle g,0| e^{iH(t-t_0)} [i(1)]^+ [j(2)]^+ [i(2)] [j(1)] e^{-iH(t-t_0)} |g,0\rangle \end{aligned} \tag{V·7}$$

which are responsible for the direct and exchange contributions to the intensity correlation. We transform these last expressions by introducing the identity operator

$$1 = e^{-iH(t-t_0)} e^{iH(t-t_0)} \tag{V·8}$$

between the commutators and next, the completeness relation. When there is not coupling between the ground states in the material subspace. The evolution operator acts on the ground state like a phase factor, so that, we have the property

$$\Psi(z_1,T_1)\, e^{-iH(t-t_0)} |g,0\rangle = 0 \tag{V·9}$$

Therefore, we express the direct term as

$$\begin{aligned} B_{2,di} = \sum_{g_1 g_2 g_3} &\langle g,0| e^{iH(t-t_0)}\, b(t-\tau_j)\, \Psi^+(z_1,T_1)\, e^{-iH(t-t_0)} |g_1,0\rangle \\ \times &\langle g_1,0| e^{iH(t-t_0)}\, b(t-\tau_i)\, \Psi^+(z_2,T_2) |g_2,0\rangle \langle g_2,0| \Psi(z_2,T_2)\, b^+(t-\tau_i)\, e^{-iH(t-t_0)} |g_3,0\rangle \\ \times &\langle g_3,0| e^{iH(t-t_0)}\, \Psi(z_1,T_1)\, b^+(t-\tau_j)\, e^{-iH(t-t_0)} |g,0\rangle \end{aligned} \tag{V·10}$$

and the exchange term as

$$\begin{aligned} B_{2,ex} = \sum_{g_1 g_2 g_3} &\langle g,0| e^{iH(t-t_0)}\, b(t-\tau_i)\, \Psi^+(z_1,T_1)\, e^{-iH(t-t_0)} |g_1,0\rangle \\ \times &\langle g_1,0| e^{iH(t-t_0)}\, b(t-\tau_j)\, \Psi^+(z_2,T_2) |g_2,0\rangle \langle g_2,0| \Psi(z_2,T_2)\, b^+(t-\tau_i)\, e^{-iH(t-t_0)} |g_3,0\rangle \end{aligned}$$

$$\times \langle q_3, 0| e^{iH(t-t_0)} \psi(z_1,T_1)\, b^+(t-\tau_j)\, e^{-iH(t-t_0)} |g,0\rangle \qquad (V.11)$$

The final expressions for the direct and exchange terms of the photon density correlation include integrations over the parameters $\{\tau\}$. With the use of factorization and normalisation properties of the probability distribution given by the relations (III.16-17), all the terms in the sum (V.6) are identical. Therefore, the photon density correlation takes the form

$$\langle N(z_1,T_1)N(z_2,T_2)\rangle = 2\left[\langle N(z_1,T_1)\,N(z_2,T_2)\rangle_{2,di} + \langle N(z_1,t_1)\,N(z_2,T_2)\rangle_{2,ex}\right] \qquad (V.12)$$

whose direct and exchange terms are obtained from (V.10) and (V.11) and given by

$$\begin{aligned}\langle N(z_1,t_1)N(z_2,t_2)\rangle_{2,di} &= \int d\tau_i\, \mathcal{P}^s_1(\tau_i)\int d\tau_j\, \mathcal{P}^*_1(\tau_j)\, C_2^{-1}(\{\tau\})\, B_{2,di} \\ \langle N(z_1,t_1)\, N(z_2,t_2)\rangle_{2,ex} &= \int d\tau_i\, \mathcal{P}^s_1(\tau_i)\int d\tau_j\, \mathcal{P}^s_1(\tau_j)\, C_2^{-1}(\{\tau\})\, B_{2,ex}\end{aligned} \qquad (V.13)$$

In the general case we cannot decouple the integrations over τ_i and τ_j because the $\{\tau\}$-dependence of the normalization factor. The definitions of the operators $\psi(z_1,T_1)$, $b^+(t-\tau_j)$ and the integral representation of the evolution operator (IV.5), we obtain

$$\begin{aligned}&\langle g_r,0| e^{iH(t-t_0)} \psi(z_1,T_1)\, b^+(t-\tau_j)\, e^{-iH(t-t_0)} |g,0\rangle \qquad (V.14)\\ &= \frac{1}{L^{1/2}}\, e^{iE_{g_r}(t-t_0)} \sum_{k k'} e^{ik'(z_1-T_1)}\, e^{-ik(t_0-\tau_j)}\, f(k)\, \frac{i}{2\pi}\int_{-\infty}^{+\infty} dE\, e^{-iE(t-t_0)} \langle g_r,k'|G(E)|g,k\rangle\end{aligned}$$

For the sake of simplicity, we consider the detection in a distinct direction from the incident one. Therefore, we can omit in the projection $QG(E)Q$ the first term $Q(E-QHQ)^{-1}$ since it does not imply interaction with the target. We introduce also the definition for the projector

$$P = \sum_m |m\rangle\langle m| \qquad (V.15)$$

and we assume in the following that the interaction radiation field-target is a slowly varying function of the photon energy, so that, we can extract it from the integration

$$\int dk \langle g', k|H'|m\rangle [\cdots] = \langle g', \bar{k}|H'|m\rangle \int dk [\cdots] \qquad (V.16)$$

Therefore, with the spectral decomposition (IV.15), we obtain

$$\begin{aligned} &\langle g_r, 0| e^{iH(t-t_0)} \psi(z_1,T_1)\, b^+(t-\tau_j)\, e^{-iH(t-t_0)} |g, 0\rangle \\ &= \mathrm{Sign}(z_1-T_1) \frac{(-i)}{L^{1/2}} e^{iE_{g_r}(t-t_0-z_1+T_1)} \sum_\beta \sum_m \sum_n \langle g_r, k|H'|m\rangle\langle m|\beta\rangle\langle\tilde{\beta}|n\rangle \qquad (V.17) \\ &\times \langle n|H'|g, k\rangle \sum_k f(k) e^{-ik(t_0-\tau_j)} \frac{1}{E_\beta - E_g - k}\left[e^{-iE_\beta(t-t_0-z_1+T_1)} - e^{-i(k+E_g)(t-t_0-z_1+T_1)}\right] \end{aligned}$$

Now, we wish to consider the extreme situations of energy and time resolved experiments. In the following, we consider a normalized lorentzian lineshape for the spectral distribution

$$f(k) = \frac{\sqrt{\Gamma_k}}{k - E_k} \quad ; \quad E_k = \Delta_k - i\,\Gamma_k/2 \qquad (V.18)$$

and we define

$$\bar{f}(k) = \frac{\sqrt{\Gamma_k}}{k - \bar{E}_k}$$

We stress that all the expressions established in the following will be correct as long as it will be possible to characterize the spectral distribution $f(k)$ by a simple pole in the negative imaginary part of the complex plane. It will be also rigorously true for any material system.

1. Stationary experiment

In such experiment, the distribution probability is constant and given by (III.23). As the experiment starts at a finite time t_0, the observation of the stationary regimen can only be obtained for times

$$t > \Delta T + t_0 + z_1 - T_1 \tag{V.20}$$

where ΔT means the time delay resulting from the scattering by the target and corresponds to $\Delta T = \Gamma_\beta^{-1}$. Therefore, we omit in the expression (V.17) the terms decaying with the lifetime of the material system. We perform the integration over τ and use the property of the resulting delta function to integrate over k. We remember here that the normalization coefficient can be evaluated directly from intensity measurements [1], by extending the volume of the detector onto the spatial extension of the light beam. It gives a multiplicative constant that we consider normalized to unity. At last, we obtain the final expression of the photon density for a stationary experiment as

$$\langle N(z_1,T_1)N(z_2,T_2)\rangle = 2\left[\langle N(z_1,T_1)N(z_2,T_2)\rangle_{2,di} + \langle N(z_1,T_1)N(z_2,T_2)\rangle_{2,ex}\right] \tag{V.21}$$

where the direct contribution corresponds to

$$\begin{aligned}
\langle N(z_1,T_1)N(z_2,T_2)\rangle_{2,di} &= \sum_{g_1 g_2 g_3} \frac{1}{L^2}\, e^{-i(E_{g_3}-E_{g_1})(z_1-T_1-z_2+T_2)} \\
&\times \Big\{ \sum_{\beta m n} \sum_{\beta' m' n'} \langle g_2,k'|H'|m\rangle\langle m|\beta\rangle\langle\tilde{\beta}|n\rangle\langle n|H'|g_3,k\rangle\langle g_2,k'|H'|m'\rangle^*\langle m'|\beta'\rangle^* \\
&\times \langle\tilde{\beta}'|n'\rangle^*\langle n'|H'|g_1,k\rangle^* \Big[\frac{1}{\Gamma_k} f(E_\beta-E_{g_3}) f(E^*_{\beta'}-E_{g_1}) - \frac{i}{E_\beta-E_{g_3}-E^*_{\beta'}+E_{g_1}} f(E_\beta-E_{g_3}) \\
&\times \bar{f}(E_\beta-E_{g_3}) \Big]\Big\} \Big\{ \sum_{\beta m n} \sum_{\beta' m' n'} \langle g_3,k'|H'|m\rangle\langle m|\beta\rangle\langle\tilde{\beta}|n\rangle\langle n|H'|g,k\rangle \\
&\times \langle g_1,k'|H'|m'\rangle^*\langle m'|\beta'\rangle^*\langle\tilde{\beta}'|n'\rangle^*\langle n'|H'|g,k\rangle^* \Big[\frac{1}{\Gamma_k} f(E_\beta-E_g) f(E^*_{\beta'}-E_g) \\
&- \frac{i}{E_\beta-E^*_{\beta'}} f(E_\beta-E_g)\bar{f}(E_\beta-E_g) \Big]\Big\}
\end{aligned} \tag{V.22}$$

and the exchange contribution to

$$\langle N(z_1,T_1)N(z_2,T_2)\rangle_{2,ex} = \sum_{g_1g_2g_3} e^{-i(E_{g_3}-E_{g_1})(z_1-T_1-z_2+T_2)}$$
$$\times\Big\{\sum_{\beta mn}\sum_{\beta'm'n'}\langle g_2,k|H'|m\rangle\langle m|\beta\rangle\langle\tilde\beta|n\rangle\langle n|H'|g_3,k\rangle\langle g_1,k|H'|m'\rangle^*\langle m'|\beta'\rangle^*$$
$$\times\langle\tilde\beta'|n'\rangle^*\langle n'|H'|g,k\rangle^*\Big[\frac{1}{\Gamma_k}f(E_\beta-E_{g_3})f(E^*_{\beta'}-E_g)\,e^{-iE_k(z_1-T_1-z_2+T_2)}-\frac{i}{E_\beta-E_{g_3}-E^*_{\beta'}+E_g}$$
$$\times f(E_\beta-E_{g_3})\bar f(E_\beta-E_{g_3})\,e^{-i(E_\beta-E_{g_3})(z_1-T_1-z_2+T_2)}\Big]\Big\}\Big\{\sum_{\beta mn}\sum_{\beta'm'n'}\langle g_3,k|H'|m\rangle$$
$$\times\langle m|\beta\rangle\langle\tilde\beta|n\rangle\langle n|H'|g,k\rangle\langle g_2,k|H'|m'\rangle^*\langle m'|\beta'\rangle^*\langle\tilde\beta'|n'\rangle^*\langle n'|H'|g_1,k\rangle^*$$
$$\times\Big[\frac{1}{\Gamma_k}\bar f(E_\beta-E_g)\bar f(E^*_{\beta'}-E_{g_1})\,e^{iE^*_k(z_1-T_1-z_2+T_2)}-\frac{i}{E_\beta-E_g-E^*_{\beta'}+E_{g_1}}f(E^*_{\beta'}-E_{g_1})$$
$$\times\bar f(E^*_{\beta'}-E_{g_1})\,e^{i(E^*_{\beta'}-E_{g_1})(z_1-T_1-z_2+T_2)}\Big]\Big\} \qquad (V\cdot 23)$$

if we assume $z_1-T_1-z_2+T_2>0$. Numerical calculations and discussions of the correlation function of the scattered beam will be presented in the next section.

2. Time resolved experiment

In such experimental situation, we need a precise time resolution, so we introduce a complete localization of the photons. Therefore, the probability distribution is given by the relation (III.26). From the relations (V.13, V.17) we obtain for the direct contribution

$$\langle N(z_1,t_1)N(z_2,t_2)\rangle_{2,di} = \sum_{g_1g_2g_3} e^{-i(E_{g_1}-E_{g_3})(t-t_2-z_1+T_1)}\int d\tau_i\,\frac{\delta(\tau_i)}{L}\sum_{\beta mn}\sum_{\beta'm'n'}\langle g_2,k|H'|m\rangle$$
$$\times\langle m|\beta\rangle\langle\tilde\beta|n\rangle\langle n|H'|g_3,k\rangle\langle g_2,k|H'|m\rangle^*\langle m'|\beta'\rangle^*\langle\tilde\beta'|n'\rangle^*\langle n'|H'|g_1,k_1\rangle^*\sum_{k\,k_1}f(k)f^*(k_1)$$

$$\times e^{-i(k-k_1)(t_0-\tau_i)} \frac{1}{(E_\beta-E_{g_3}-k)(E^*_{\beta'}-E_{g_1}-k)} \left[e^{-iE_\beta(t-t_0-z_2+T_2)} - e^{-i(k+E_{g_3})(t-t_0-z_2+T_2)}\right]$$

$$\times\left[e^{iE^*_{\beta'}(t-t_0-z_2+T_2)} - e^{i(k_1+E_{g_1})(t-t_0-z_2+T_2)}\right]\Big\}\Big\{\int d\tau_j \frac{\delta(\tau_j)}{L} \sum_{\beta mn}\sum_{\beta'm'n'} \langle g_3,k'|H'|m\rangle\langle m|\beta\rangle$$

$$\times\langle\tilde\beta|n\rangle\langle n|H'|g_1k\rangle\langle g_1,k'|H'|m'\rangle^*\langle m'|\beta'\rangle^*\langle\tilde\beta'|n'\rangle^*\langle n'|H'|g_1k_1\rangle^* \sum_{kk_1} f(k)f^*(k_1)$$

$$\times e^{-i(k-k_1)(t_0-\tau_j)} \frac{1}{(E_\beta-E_g-k)(E^*_{\beta'}-E_g-k_1)} \left[e^{-iE_\beta(t-t_0-z_1+T_1)} - e^{-i(k+E_g)(t-t_0-z_1+T_1)}\right]$$

$$\times\left[e^{iE^*_{\beta'}(t-t_0-z_1+T_1)} - e^{i(k_1+E_g)(t-t_0-z_1+T_1)}\right]\Big\} \qquad (V\cdot 24)$$

and for the exchange contribution

$$\langle N(z_1,t_1)N(z_2,t_2)\rangle_{2,ex} = \sum_{g_1g_2g_3} e^{-i(E_{g_1}-E_{g_3})(t-t_0-z_1+T_1)} \Big\{\int d\tau_i \frac{\delta(\tau_i)}{L}$$

$$\times\sum_{\beta mn}\sum_{\beta'm'n'} \langle g_2,k'|H'|m\rangle\langle m|\beta\rangle\langle\tilde\beta|n\rangle\langle n|H'|g_3,k\rangle\langle g_1,k'|H'|m'\rangle^*\langle m'|\beta'\rangle^*\langle\tilde\beta'|n'\rangle^*\langle n'|H'|g_1k_1\rangle^*$$

$$\times\sum_{kk_1} f(k)f^*(k_1)\, e^{-i(k-k_1)(t_0-\tau_i)} \frac{1}{(E_\beta-E_{g_3}-k)(E^*_{\beta'}-E_g-k_1)} \left[e^{-iE_\beta(t-t_0-z_2+T_2)} - e^{-i(k+E_{g_3})(t-t_0-z_2+T_2)}\right]$$

$$\times\left[e^{iE^*_{\beta'}(t-t_0-z_1+T_1)} - e^{i(k_1+E_g)(t-t_0-z_1+T_1)}\right]\Big\}\Big\{\int d\tau_j \frac{\delta(\tau_j)}{L} \sum_{\beta mn}\sum_{\beta'm'n'} \langle g_3,k'|H'|m\rangle\langle m|\beta\rangle$$

$$\times\langle\tilde\beta|n\rangle\langle n|H'|g_1k\rangle\langle g_2,k'|H'|m'\rangle^*\langle m'|\beta'\rangle^*\langle\tilde\beta'|n'\rangle^*\langle n'|H'|g_1,k_1\rangle^* \sum_{kk_1} f(k)f^*(k_1)$$

$$\times e^{-i(k-k_1)(t_0-\tau_j)} \frac{1}{(E_\beta-E_g-k)(E^*_{\beta'}-E_{g_1}-k_1)} \left[e^{-iE_\beta(t-t_0-z_1+T_1)} - e^{-i(k+E_g)(t-t_0-z_1+T_1)}\right]$$

$$\times\left[e^{iE^*_{\beta'}(t-t_0-z_2+T_2)} - e^{i(k_1+E_{g_1})(t-t_0-z_2+T_2)}\right]\Big\} \qquad (V\cdot 25)$$

With the property of the delta function, we eliminate the integration over τ. Also we assume that the spectral distribution $f(k)$ is defined like in the case of the free light beam, so that, we have after integration

$$\sum_k f(k)\, e^{-ikt_0} \frac{1}{E_\beta-E_{g_3}-k} \left[e^{-iE_\beta(t-t_0-z_2+T_2)} - e^{-i(k+E_{g_3})(t-t_0-z_2+T_2)}\right]$$

$$= i f(E_\beta-E_{g_3})\, e^{-iE_k t_0}\left[e^{-i(E_{g_3}+E_k)(t-t_0-z_2+T_2)} - e^{-iE_\beta(t-t_0-z_2+T_2)}\right] \qquad (V\cdot 26)$$

Now, it is possible to express the final expression of the total photon density correlation (V.12), in the time resolved situation, by the general form

$$\langle N(z_1,t_1)N(z_2,t_2)\rangle = 2\left[\langle N(z_1,t_1)N(z_2,t_2)\rangle_{2,di} + \langle N(z_1,t_1)N(z_2,t_2)\rangle_{2,ex}\right] \quad (V.27)$$

whose direct term is given by

$$\langle N(z_1,t_1)N(z_2,t_2)\rangle_{2,di} = \sum_{g_1 g_2 g_3} \frac{1}{L^2}\, e^{-i(E_{g_1}-E_{g_3})(t-t_0-z_1+T_1)}\, I_2(z_2,t_2)\, I_1(z_1,t_1) \quad (V.28)$$

and exchange term by

$$\langle N(z_1,t_1)N(z_2,t_2)\rangle_{2,ex} = \sum_{g_1 g_2 g_3} \frac{1}{L^2}\, e^{-i(E_{g_1}-E_{g_3})(t-t_0-z_1+T_1)}\, I_2(z_1,t_1;z_2,t_2)\, I_1(z_1,t_1;z_2,t_2) \quad (V.29)$$

We have introduced the definition for the intensities

$$\begin{aligned}
I_2(z_2,t_2) = {} & \sum_{\beta m n}\sum_{\beta' m' n'} \langle g_2,k'|H'|m\rangle\langle m|\beta\rangle\langle\tilde\beta|n\rangle\langle n|H'|g_3,k\rangle\langle g_2,k'|H'|m'\rangle^*\langle m'|\beta'\rangle^* \\
& \times \langle\tilde\beta'|n'\rangle^*\langle n'|H'|g_1,k_1\rangle^*\, f(E_\beta-E_{g_3})\, f^*(E_{\beta'}-E_{g_1})\, e^{-\Gamma_k t_0}\Big[e^{-i(E_{g_3}-E_{g_1})(t-t_0-z_2+T_2)} \\
& \times e^{-\Gamma_k(t-t_0-z_2+T_2)} + e^{-i(\Delta_\beta-\Delta_{\beta'})(t-t_0-z_2+T_2)}\, e^{-\frac{1}{2}(\Gamma_\beta+\Gamma_{\beta'})(t-t_0-z_2+T_2)} - e^{-i(E_{g_3}+\Delta_k-\Delta_{\beta'})(t-t_0-z_2+T_2)} \\
& \times e^{-\frac{1}{2}(\Gamma_k+\Gamma_{\beta'})(t-t_0-z_2+T_2)} - e^{i(E_{g_1}+\Delta_k-\Delta_\beta)(t-t_0-z_2+T_2)}\, e^{-\frac{1}{2}(\Gamma_k+\Gamma_\beta)(t-t_0-z_2+T_2)}\Big]
\end{aligned} \quad (V.30)$$

$$\begin{aligned}
I_1(z_1,t_1) = {} & \sum_{\beta m n}\sum_{\beta' m' n'} \langle g_3,k'|H'|m\rangle\langle m|\beta\rangle\langle\tilde\beta|n\rangle\langle n|H'|g,k\rangle\langle g_1,k'|H'|m'\rangle^*\langle m'|\beta'\rangle^* \\
& \times \langle\tilde\beta'|n'\rangle\langle n'|H'|g,k_1\rangle^*\, f(E_\beta-E_g)\, f^*(E_{\beta'}-E_g)\, e^{-\Gamma_k t_0}\Big[e^{-\Gamma_k(t-t_0-z_1+T_1)} + e^{-i(\Delta_\beta-\Delta_{\beta'})(t-t_0-z_1+T_1)} \\
& \times e^{-\frac{1}{2}(\Gamma_\beta+\Gamma_{\beta'})(t-t_0-z_1+T_1)} - e^{-i(E_g+\Delta_k-\Delta_{\beta'})(t-t_0-z_1+T_1)}\, e^{-\frac{1}{2}(\Gamma_k+\Gamma_{\beta'})(t-t_0-z_1+T_1)} \\
& - e^{i(E_g+\Delta_k-\Delta_\beta)(t-t_0-z_1+T_1)}\, e^{-\frac{1}{2}(\Gamma_k+\Gamma_\beta)(t-t_0-z_1+T_1)}\Big]
\end{aligned}$$

and also for the exchange intensities

$$
\begin{aligned}
I_2(z_1,t_1;z_2,t_2) = {} & \sum_{\beta m n} \sum_{\beta' m' n'} \langle g_2,k'|H'|m\rangle\langle m|\beta\rangle\langle\tilde{\beta}|n\rangle\langle n|H'|g_3,k\rangle\langle g_1,k'|H'|m'\rangle^*\langle m'|\beta'\rangle^* \\
& \times \langle\tilde{\beta}'|n'\rangle^*\langle n'|H'|g,k_1\rangle^* f(E_\beta-E_{g_3}) f^*(E_{\beta'}-E_g)\, e^{-\Gamma_k t_0} \Big[e^{-i(E_{g_3}+\Delta_k)(t-t_0-z_2+T_2)}\, e^{i(E_g+\Delta_k)(t-t_0-z_1+T_1)} \\
& \times e^{-\frac{1}{2}\Gamma_k(2t-2t_0-z_1+T_1-z_2+T_2)} + e^{-i(\Delta_\beta-\Delta_{\beta'})(t-t_0)}\, e^{-i[\Delta_\beta(-z_2+T_2)-\Delta_{\beta'}(-z_1+T_1)]}\, e^{-\frac{1}{2}(\Gamma_\beta+\Gamma_{\beta'})(t-t_0)} \\
& \times e^{-\frac{1}{2}[\Gamma_\beta(-z_2+T_2)+\Gamma_{\beta'}(-z_1+T_1)]} - e^{-i(E_{g_3}+\Delta_k)(t-t_0-z_2+T_2)}\, e^{i\Delta_{\beta'}(t-t_0-z_1+T_1)}\, e^{-\frac{1}{2}(\Gamma_k+\Gamma_{\beta'})(t-t_0)} \\
& \times e^{-\frac{1}{2}[\Gamma_k(-z_2+T_2)+\Gamma_{\beta'}(-z_1+T_1)]} - e^{i(E_g+\Delta_k)(t-t_0-z_1+T_1)}\, e^{-i\Delta_\beta(t-t_0-z_2+T_2)}\, e^{-\frac{1}{2}(\Gamma_k+\Gamma_\beta)(t-t_0)} \\
& \times e^{-\frac{1}{2}[\Gamma_k(-z_1+T_1)+\Gamma_\beta(-z_2+T_2)]} \Big]
\end{aligned}
\tag{V-31}
$$

$$
\begin{aligned}
I_1(z_1,t_1;z_2,t_2) = {} & \sum_{\beta m n} \sum_{\beta' m' n'} \langle g_3,k'|H'|m\rangle\langle m|\beta\rangle\langle\tilde{\beta}|n\rangle\langle n|H'|g,k\rangle\langle g_2,k'|H'|m'\rangle^*\langle m'|\beta'\rangle^* \\
& \times \langle\tilde{\beta}'|n'\rangle\langle n'|H'|g_4,k_1\rangle^* f(E_\beta-E_0) f^*(E_{\beta'}-E_{g_4})\, e^{-\Gamma_k t_0} \Big[e^{-i(E_g+\Delta_k)(t-t_0-z_1+T_1)}\, e^{i(E_g+\Delta_k)(t-t_0-z_2+T_2)} \\
& \times e^{-\frac{1}{2}\Gamma_k(2t-2t_0-z_1+T_1-z_2+T_2)} + e^{-i(\Delta_\beta-\Delta_{\beta'})(t-t_0)}\, e^{-i[\Delta_\beta(-z_1+T_1)-\Delta_{\beta'}(-z_2+T_2)]}\, e^{-\frac{1}{2}[\Gamma_\beta(-z_1+T_1)+\Gamma_{\beta'}(-z_2+T_2)]} \\
& \times e^{-\frac{1}{2}(\Gamma_\beta+\Gamma_{\beta'})(t-t_0)} - e^{-i(E_g+\Delta_k)(t-t_0-z_1+T_1)}\, e^{i\Delta_{\beta'}(t-t_0-z_2+T_2)}\, e^{-\frac{1}{2}(\Gamma_{\beta'}\ \Gamma_k)(t-t_0)} \\
& \times e^{-\frac{1}{2}[\Gamma_k(-z_1+T_1)+\Gamma_{\beta'}(-z_2+T_2)]} - e^{i(E_{g_3}+\Delta_k)(t-t_0-z_2+T_2)}\, e^{-i\Delta_\beta(t-t_0-z_1+T_1)}\, e^{-\frac{1}{2}(\Gamma_k+\Gamma_\beta)(t-t_0)} \\
& \times e^{-\frac{1}{2}[\Gamma_k(-z_2+T_2)+\Gamma_\beta(-z_1+T_1)]} \Big]
\end{aligned}
$$

From these two general expressions of the scattered beam correlation functions, describing stationary and time resolved situation, we will discuss in the following the influence of the various physical parameters. Besides, we must note that the contribution to the direct and exchange terms is expressed in terms of single photon amplitudes of absorption or emission processes, even if we need two incident photons for performing correlation measurements. The exchange contribution results from the indistinguishability of the scattered photons. It will be discussed in more details in the simplest case of one resonance model considered for the target model.

VI. Numerical calculations and discussions

Up to now, we have dealt with general expressions corresponding to typical experimental situations. In fact, correlations measurement offer much more possibilities of measurements than the intensity ones. It is the object of this section to discuss these various possibilities and to analyze what best or new informations arise from these measurements. For practical purposes, all the discussions and numerical calculations will be performed on the simplest target system - atoms or molecules - characterized by just one resonance. Obviously, these measurements will be doubly so interesting as the target spectra will be complicated. More sophisticated material models can be treated, but their internal complexity is not relevant here.

We begin by the case of stationary experimental situation. From relations (V.21-23) we have described [8] a particular case of single resonance target models, assuming also the existence of a single level in the fundamental configuration. Then, the time delay dependence of the correlation function is provided by the exchange term alone and corresponds to

$$\begin{aligned}\langle N(z_1,T_1)\,N(z_2,T_2)\rangle_{ex} &= 2\,|\langle g,k|H'|\beta\rangle|^8\,\Gamma_k^2\left[(\Delta_k-\Delta_\beta+E_g)^2+\tfrac{1}{4}(\Gamma_\beta-\Gamma_k)^2\right]^{-1}\\ &\times\left[(\Delta_k-\Delta_\beta+E_g)^2+\tfrac{1}{4}(\Gamma_\beta+\Gamma_k)^2\right]^{-1}\Big\{\frac{1}{\Gamma_k^2}\,e^{-\Gamma_k(z_1-T_1-z_2+T_2)}+\frac{1}{\Gamma_\beta^2}\,e^{-\Gamma_\beta(z_1-T_1-z_2+T_2)}\\ &+\frac{2}{\Gamma_k\Gamma_\beta}\,e^{-\frac{1}{2}(\Gamma_k+\Gamma_\beta)(z_1-T_1-z_2+T_2)}\,\sin\left[(\Delta_k-\Delta_\beta+E_g)(z_1-T_1-z_2+T_2)+\lambda\right]\Big\}\end{aligned}\qquad \text{(VI.1)}$$

with

$$\sin\lambda = \left[(\Delta_k - \Delta_\beta + E_g)^2 - \frac{1}{4}(\Gamma_\beta + \Gamma_k)^2\right]\left[(\Delta_k - \Delta_\beta + E_g)^2 + \frac{1}{4}(\Gamma_\beta + \Gamma_k)^2\right]^{-1} \qquad (\mathrm{VI}\cdot 2)$$

In figures I we represent the dependence of the correlation as a function of the time delay. The various contribution to $\langle N(z_1,T_1)\,N(z_2,T_2)\rangle$ have been discussed previously [8]. The contribution arising from resonance fluorescence decays rapidly because for a stationary experiment Γ_k is smaller than Γ_β. Also its contribution is very small because of the factors Γ_k^{-2} and Γ_β^{-2}. Therefore, in common experimental situations, the decay is essentially governed by the resonance raman scattering except near the origin. Also, we note a stronger influence of the excitation wave packet structure than for intensity measurements [1]. The overlapping between both target and field resonances varies with the relative importance of Γ_β and Γ_k so, it is responsible for the diminution of the correlation function. Figures (Ia) and(Ib) show the influence of the resonance parameter, which modify too the overlapping of the resonances. We note, in figure (Ic), an inversion of the relative importance of the correlation function. This phenomenon has yet been discussed by myself in the case of intensity measurements. It depends on the analytical dependence of the resonance overlapping. On the contrary, figures II show the same dependence like in figures I, but here, we have superimposed the curves corresponding to different off-resonance parameters. For small values of Γ_k, we observe a global diminution of the correlation associated to the overlapping resonance, but the curves have the same characteristics. With higher values of Γ_k we have too a variation of the characteristics of the curves. This last one, results from the mixed contribution of resonance fluorescence and resonance raman. Its contribution becomes more important with the increase of Γ_k. We cannot observe the oscillation provided by the sines function because the period is longer than the decay time $(\Gamma_\beta+\Gamma_k)^{-1}$. Also we note a small oscillation in all the curves. This phenomenon will be discussed now extensively and results from the existence of two ground levels in the fundamental configuration. From relations (V.22-23) we note the presence of a purely oscillating term $\exp\left[-i(E_{g_2}-E_{g_1})(z_1-T_1-z_2+T_2)\right]$ whose frequency corresponds to the energy gap between the ground levels and gives rise to small oscillations in figures II. This factor indicates the ability to observe a coherent contribution of the scattered photons, a result which cannot be reached by intensity measurements, at least, in the framework of the theory of quantum electrodynamics [17]. In pictures III we have represented the time delay dependence of the scattered intensity correlation for various energy gap between two ground levels. We observe the more important contribution, when the two levels are degenerated. If the energy gap is different from zero, we note an oscillation whose amplitude is small and will be probably washed out by any average processes.

We consider now, the case of time resolved experiment and we look only for the time delay dependence at a fixed experimental time. In fact, there is also the possibility of measuring the time dependence of the correlation for a fixed time delay of the detection set-up. We will discuss such possibility in a following paper. In figure IV, we show the time delay dependence of the correlation of the scattered intensity. In all the cases we observe a rise time which corresponds to the lifetime of the excited state level of the target whereas the decay is essentially governed by the coherence time of the single photon wave packet. We have superimposed the curves corresponding to different spectral linewidth. We note, like in stationary experiments, a strong dependence associated to the overlapping of both target and field resonances. The picture IV.c exhibits an inversion of the relative importance of the correlation associated to its analytical dependence with the off resonance parameter. On the contrary, pictures V show the influence of the off-resonance parameter for various spectral linewidth. As expected, the influence of $(\Delta_k-\Delta_p+E_g)$ decreases with the increase of Γ_k. At last, figures VI and VII exhibit the dependence of the correlation with the energy gap of the ground levels. The most peculiar feature is the very strong influence of the spectral linewidth on the coherent summation of the various scattered photons. Also, we note that this phenomenon is much more efficient for time resolved experiments than for stationary ones.

VII. Conclusion

We have dealt with the description of correlation measurements on a scattered beam. Also we have analyzed the relevance for such measurements of the various physical parameters. In all the cases, we observe a stronger dependence for the correlation measurements than for the intensity ones. Concerning the stationary experiments, it appears that we can obtain by stationary excitation conditions the same informations resulting from pulsed intensity measurements. This can be of interest for practical experimental situations. Also these measurements must be more sensitive to the coherence properties of the light. Moreover, we have seen that in both, stationary and pulsed experiments, we observe a coherent contribution associated to the final ground levels. This observation can be relevant in order to follow rapid relaxation dynamics of target in these levels.

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FIGURE CAPTIONS

Figure I : Plot of the correlation function of the scattered intensity as a function of the time delay $z_1-T_1-z_2+T_2$. We consider a stationary experiment performed with a single resonance target model. The case of various spectral linewidth Γ_k have been superimposed.

$\uparrow$ $(\Gamma_k = 0{,}5\,\Gamma_p)$; $\leftarrow$ $(\Gamma_k = 0{,}1\,\Gamma_p)$; $\downarrow$ $(\Gamma_k = 0{,}01\,\Gamma_p)$
for three different off resonance parameter $\Delta_k-\Delta_p+E_g$

Figure II : Plot of the scattered intensity correlation analogous to the one described in figure I. We compare here the influence of the off resonance parameter

$\uparrow$ $(\Delta_k-\Delta_p+E_g=0)$; $\leftarrow$ $(\Delta_k-\Delta_p+E_g=0{,}2\,\Gamma_p)$; $\downarrow$ $(\Delta_k-\Delta_p+E_g=\Gamma_p)$
for various spectral linewidths Γ_k. The oscillation corresponds to the structure in the fundamental configuration with $\Delta E_{g_i} = 10\,\Gamma_p$

Figure III : We represent the same correlation function described in figure I, but here the curves are superimposed for various energy gap $\Delta E_g = E_{g'}-E_g$ between the levels pertaining to the fundamental configuration

$\uparrow$ $(\Delta E_g = 0)$; $\leftarrow$ $(\Delta E_g = 10\,\Gamma_p)$; $\downarrow$ $(\Delta E_g = 50\,\Gamma_p)$
for various off resonance parameters.

Figure IV : Plot of the scattered intensity correlation as a function of the time delay $z_1-T_1-z_2+T_2$ in the case of pulsed experiments performed with a single resonance target model. The cases of different spectral linewidths have been considered

$\uparrow$ $(\Gamma_k = 10\,\Gamma_p)$; $\leftarrow$ $(\Gamma_k = 30\,\Gamma_p)$; $\downarrow$ $(\Gamma_k = 100\,\Gamma_p)$
for various off resonance parameters.

Figure V : Plot of the scattered intensity correlation analogous to the one described in figure IV. We compare here the influence of the off resonance parameter

$\uparrow$ $(\Delta_k - \Delta_p + E_g = 0)$; $\leftarrow$ $(\Delta_k - \Delta_p + E_g = 3\Gamma_p)$; $\rightarrow$ $(\Delta_k - \Delta_p + E_g = 5\Gamma_p)$;
$\downarrow$ $(\Delta_k - \Delta_p + E_g = 10\,\Gamma_p)$

for various spectral linewidths.

Figure VI : We represent the same function described in figure IV, but here the curves are superimposed for different spectral linewidths

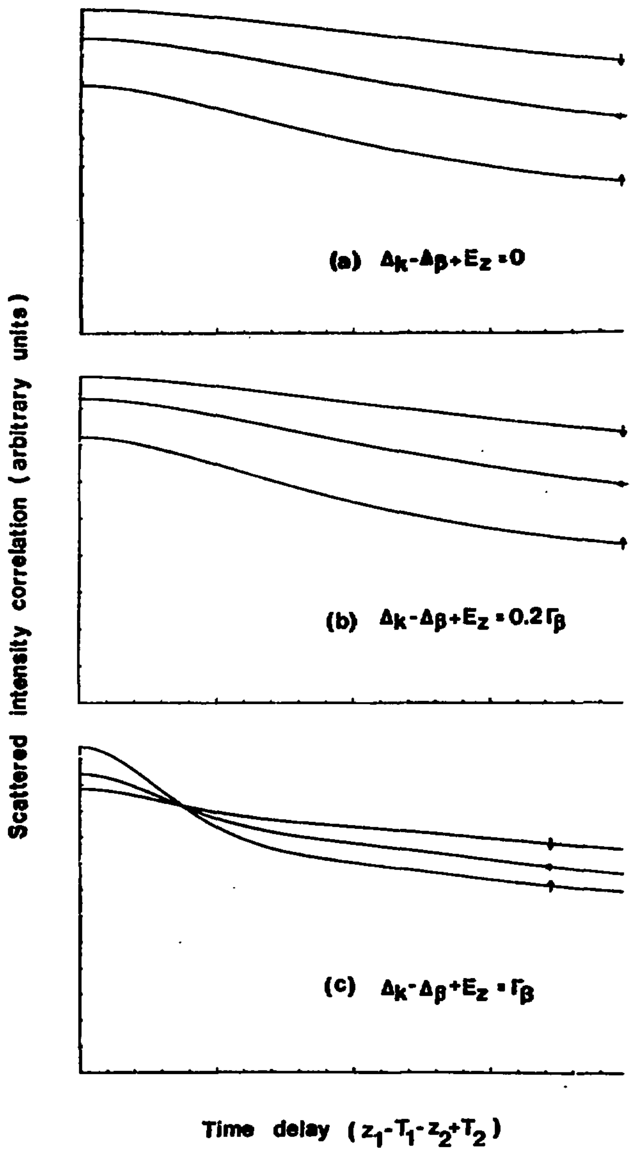
$\uparrow$ $(\Gamma_k = 10\,\Gamma_p)$; $\leftarrow$ $(\Gamma_k = 30\,\Gamma_p)$; $\downarrow$ $(\Gamma_k = 100\,\Gamma_p)$

The cases of various energy gap $\Delta E_g = E_{g'} - E_g$ between the levels pertaining to the fundamental configuration are considered.

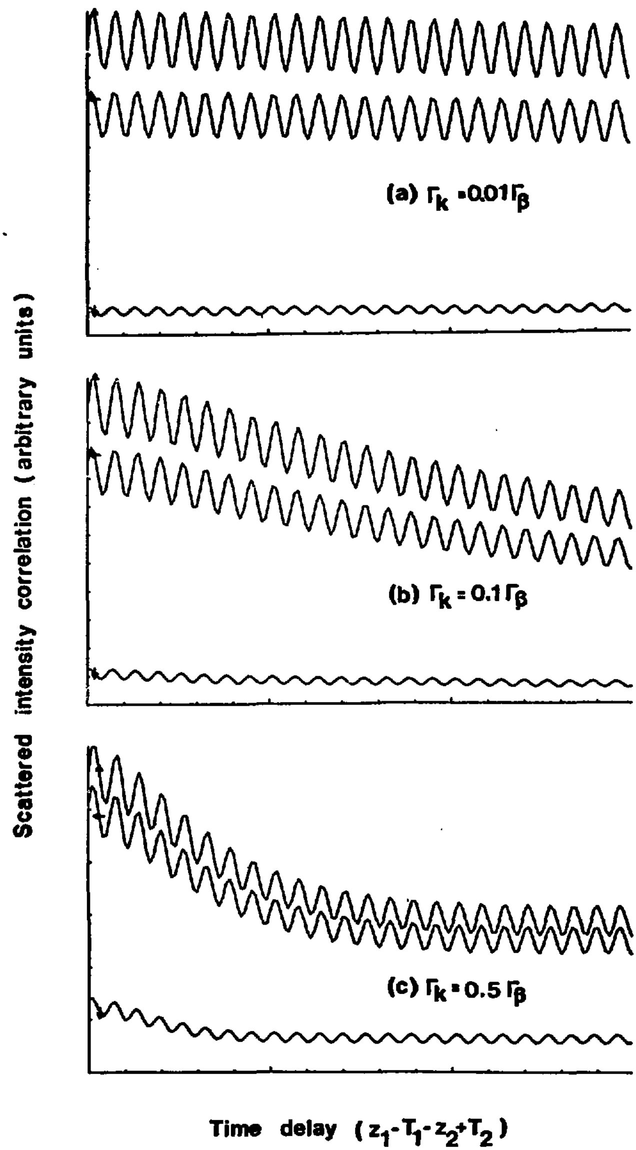
Figure VII : Plot of the scattered intensity correlation analogous to the one described in figure IV. The curves for various energy gap levels in the fundamental configuration are superimposed

$\uparrow$ $(\Delta E_g = 0)$; $\leftarrow$ $(\Delta E_g = 50\,\Gamma_p)$; $\downarrow$ $(\Delta E_g = 100\,\Gamma_p)$

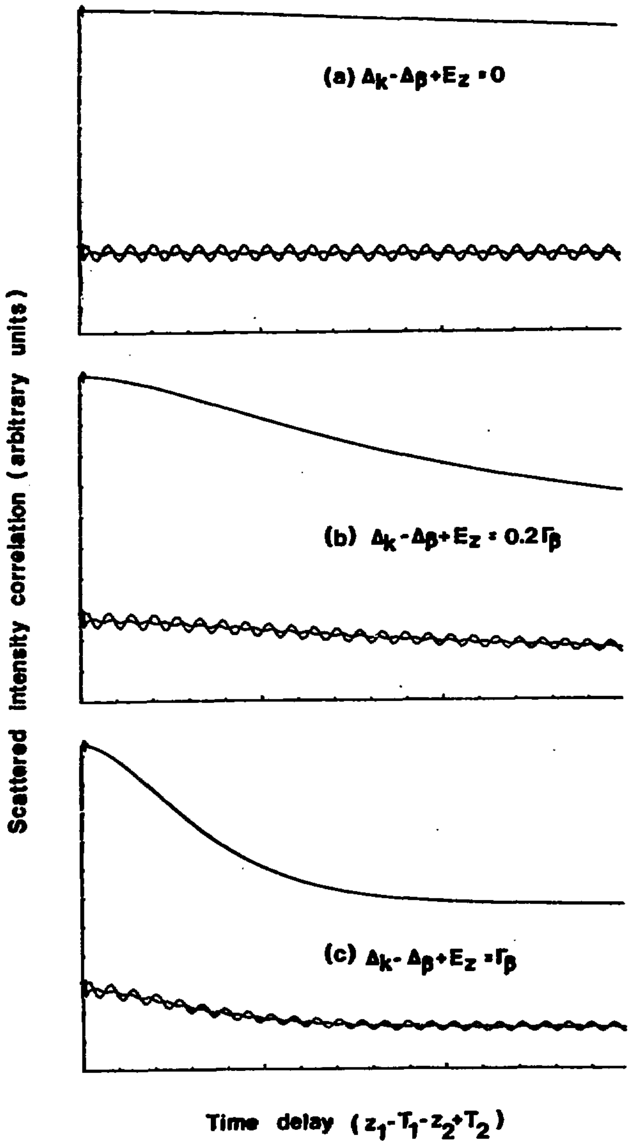
The cases of various spectral linewidths are considered.



A. VILLAEYS fig I



A. VILLAEYS Fig II



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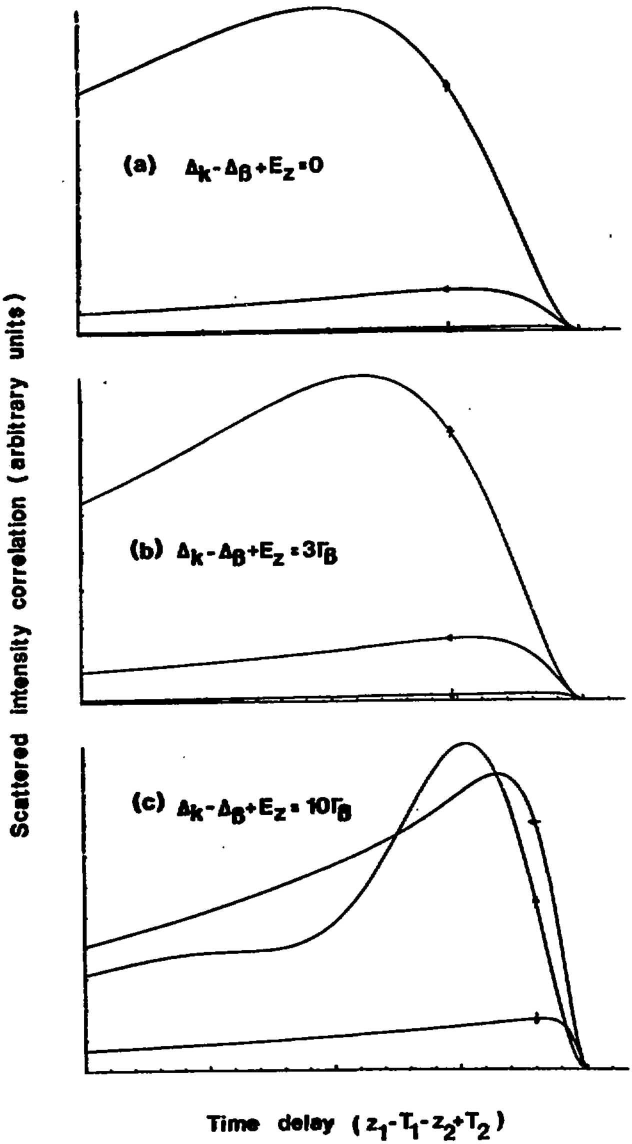
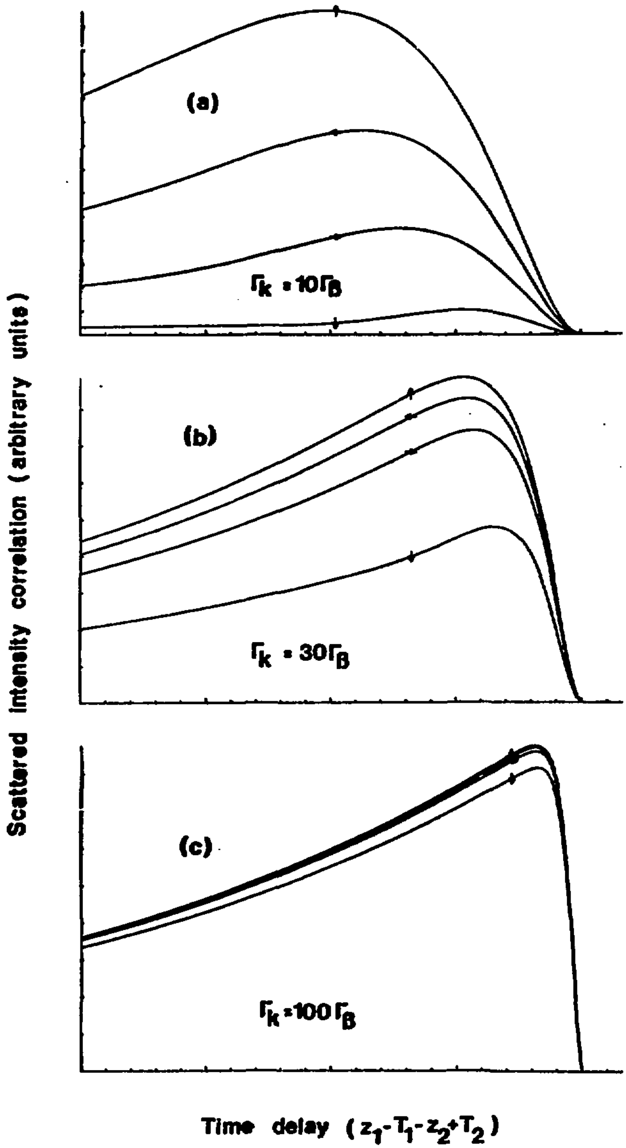


A. VILLAEYS fig IV



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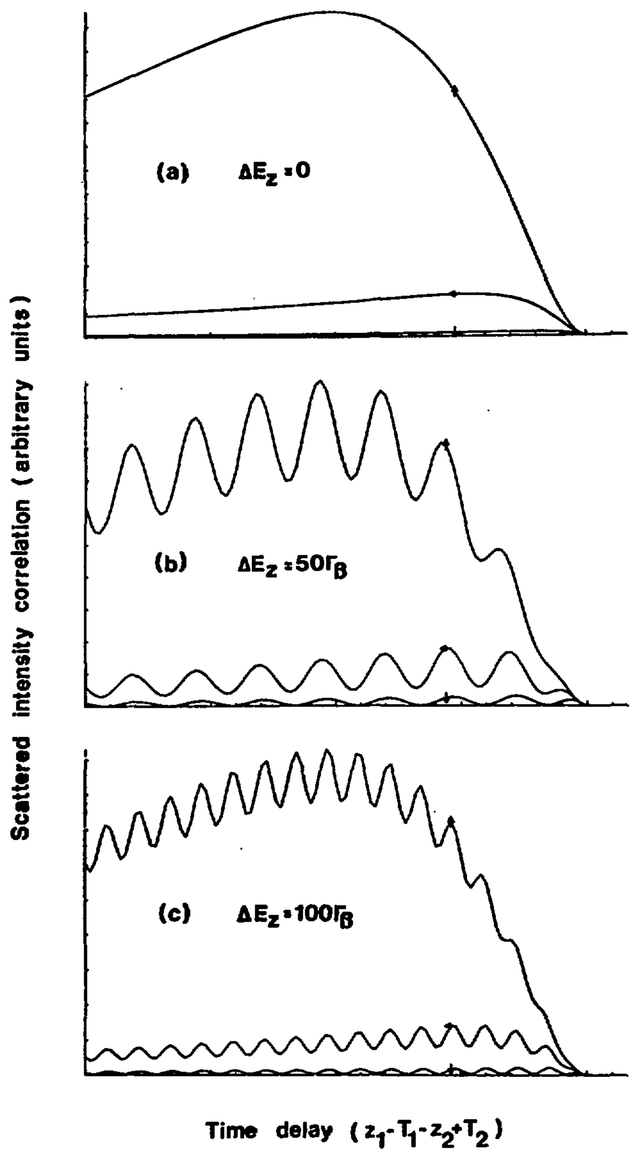
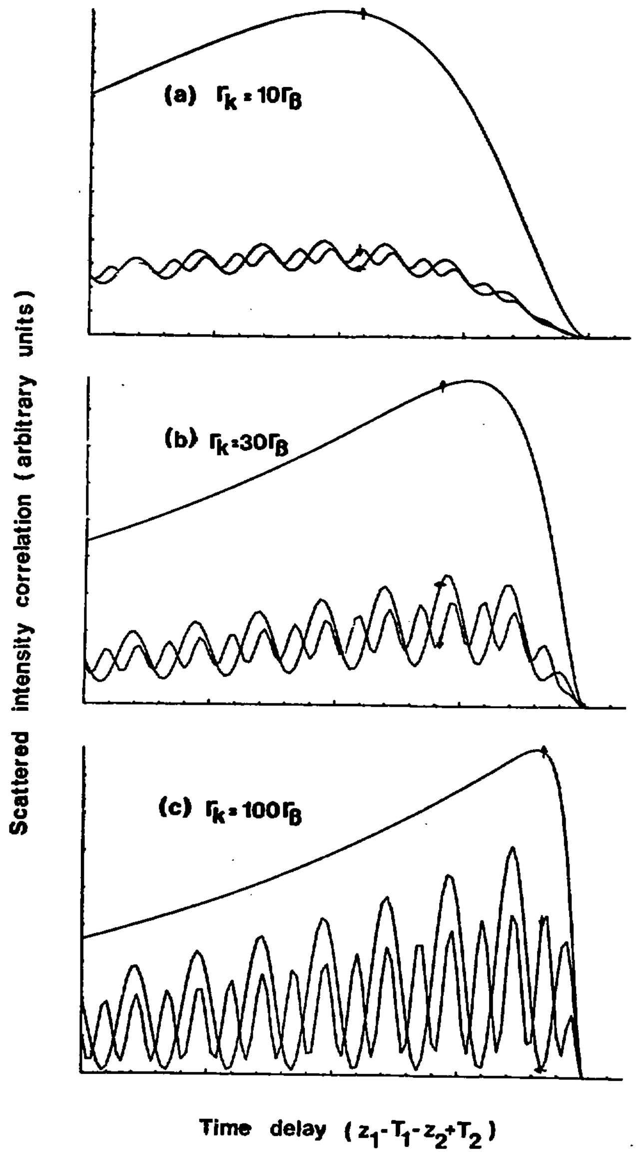


A. VILLAEYS fig VI



A. VILLAEYS fig VII