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Correlation of photons in collective Raman scattering

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Résumé. — On détermine la statistique de photons des composantes du spectre de la raie Stokes dans la diffusion Raman collective. On calcule aussi le groupement et le dégroupement entre les composantes du spectre de la raie Stokes, et entre la raie Stokes et la raie Rayleigh.

Abstract. — The photon statistics of spectrum components of the Stokes line in collective Raman scattering are investigated. The cross bunching and cross-antibunching between the spectrum components of the Stokes line and between the Stokes and Rayleigh lines are investigated too.

1. Introduction.

In the last few years the interest in the statistical properties of photons in various nonlinear optical processes has increased. The antibunching, anticorrelation, and squeezing have been a subject of various theoretical and experimental studies [2-20]. In the theoretical work by Cohen-Tannoudji and Reynaud [3], Apanasevich and Kilin [4], and the experimental work by Aspect *et al.* [2] the cross-correlation between the spectrum components of resonance fluorescence for the one-atom case has been investigated. The anticorrelation between the Stokes and Rayleigh lines in the resonance Raman scattering of the one-atom case have been predicted by Agarwal and Jha [23]. The recent publications deal with collective effects in photon statistics in resonance fluorescence [8-10], double resonance [17] and Raman scattering [6, 13].

In the present paper the correlations of photons in the collective Raman scattering in an intense driving field are studied (Fig. 1). The photon statistics of spectrum components, correlation and anticorrelation between spectrum components of the Stokes line and between the Stokes and Rayleigh lines are investigated.

2. Master equation.

We consider a small system (the Dicke model, 1954) of N three-level atoms interacting with a mono-

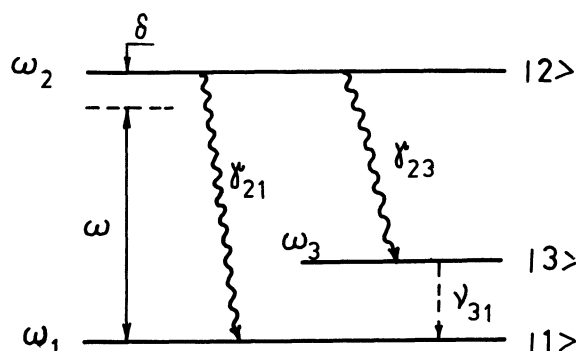


Fig. 1. — Three-level system of atoms interacting with the monochromatic applied field.

chromatic driving field of a frequency ω and the radiation vacuum (Fig. 1). Let us label the ground state by $|1\rangle$, the real excited state by $|3\rangle$ and the resonant intermediate state by $|2\rangle$ with energies ω_1 , ω_3 and ω_2 , respectively (the system of $\hbar \equiv 1$). The real excited state $|3\rangle$ may be a low-lying vibrational or rotational excitation from the ground state. To keep the discussion general, we will not specify these states besides saying that the intermediate state $|2\rangle$ can be connected *via* the electromagnetic interaction Hamiltonian with both the states $|1\rangle$ and $|3\rangle$ (in the dipole approximation), but the states $|3\rangle$ and $|1\rangle$ are not connected by the dipole Hamiltonian because of parity consideration.

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The transition $|3\rangle - |1\rangle$ is caused by an atomic reservoir and assumed to be nonradiative [22].

In treating the external field classically and using the Born and Markov approximation with respect to the coupling of the system with the vacuum field and atomic reservoir, one can obtain a master equation for the reduced density matrix ρ for the system alone in the form [1, 6]

$$\begin{aligned} \frac{\partial \rho}{\partial t} = & -i[H_{\text{coh}}, \rho] - \\ & -\gamma_{21}(J_{21}J_{12}\rho - J_{12}\rho J_{21} + H.C.) \\ & -\gamma_{23}(J_{23}J_{32}\rho - J_{32}\rho J_{23} + H.C.) \\ & -\nu_{31}(J_{31}J_{13}\rho - J_{13}\rho J_{31} + H.C.) \equiv L\rho, \quad (1) \end{aligned}$$

where $2\gamma_{21}$ and $2\gamma_{23}$ are the radiative spontaneous transition probabilities per unit time for a single atom to change from the level $|2\rangle$ to $|1\rangle$ and from $|2\rangle$ to $|3\rangle$, respectively; $2\nu_{31}$ is the nonradiative rate for atomic transition from $|3\rangle$ to $|1\rangle$.

The coherence part of Hamiltonian H_{coh} in the interaction picture has the form

$$H_{\text{coh}} = \frac{\delta}{2}(J_{22} - J_{11}) + G(J_{21} + J_{12}) - \Omega_3 J_{33}.$$

Here $\Omega_3 = \omega_{23} - \frac{\omega_{21}}{2}$ (where $\omega_{ij} = \omega_i - \omega_j$; $i, j = 1, 2, 3$); $\delta = \omega_{21} - \omega$ is the frequency detuning of resonance; $G = -\mathbf{d}_{2i} \mathbf{E}_0$ is the matrix element of the driving field and atomic interaction, and

$$J_{ij} = \sum_{K=1}^N |i\rangle_K \langle j| \quad (i, j = 1, 2, 3)$$

are the collective angular momenta of the atoms. They satisfy the commutation relation:

$$[J_{ij}, J_{i'j'}] = J_{ij'} \delta_{ji'} - J_{i'j} \delta_{ij'}.$$

As in references [24, 5] we introduce the Schwinger representation for the angular momentum

$$J_{ij} = C_i^+ C_j \quad (i, j = 1, 2, 3),$$

where C_i obey the boson commutation relation:

$$[C_i, C_j^+] = \delta_{ij}.$$

Further we only investigate the case of an intense external field or large detuning δ , so that:

$$\Omega = \left(\frac{1}{4} \delta^2 + G^2 \right)^{1/2} \gg N\gamma_{21}, N\gamma_{23}, N\nu_{31}. \quad (2)$$

We make the canonical transformation which permits us to move from a system of atoms to a system of dressed atoms

$$\begin{aligned} C_1 &= Q_1 \cos \varphi + Q_2 \sin \varphi, \\ C_2 &= -Q_1 \sin \varphi + Q_2 \cos \varphi, \\ C_3 &= Q_3, \end{aligned} \quad (3)$$

where

$$\tan 2\varphi = 2G/\delta.$$

After performing the canonical transformation (3) one can find that the Liouville operator L appearing in equation (1) splits into two components L_0 and L_1 . The component L_0 is slowly varying in time whereas L_1 contains rapidly oscillating terms at frequencies 2Ω and 4Ω . For the case when relation (2) is fulfilled we make the secular approximation, i.e., retain only a slowly varying part [5, 8]. Correction of the results obtained in this fashion will be of an order $(\gamma_{21}N/\Omega)^2$, $(\gamma_{23}N/\Omega)^2$ or $(\nu_{31}N/\Omega)^2$.

Making the secular approximation, one can find the stationary solution of the master equation:

$$\begin{aligned} \tilde{\rho} = U\rho U^+ = & A^{-1} \sum_{P=0}^N X^P \times \\ & \times \sum_{N_1=0}^P Z^{N_1} |P, N_1\rangle \langle N_1, P|, \quad (4) \end{aligned}$$

where U is the unitary operator representing the canonical transformation (3)

$$X = \frac{\nu_{31}}{\gamma_{23}} \tan^2 \varphi$$

$$Z = \cot^4 \varphi$$

$$A = \frac{Z}{Z-1} \cdot \frac{(XZ)^{N+1} - 1}{XZ - 1} - \frac{1}{Z-1} \cdot \frac{X^N - 1}{X - 1}.$$

$|P, N_1\rangle$ is an eigenstate of the operators $R = R_{11} + R_{22}$, R_{11} and

$$\hat{N} = R_{11} + R_{22} + R_{33} = J_{11} + J_{22} + J_{33},$$

i.e.

$$R|P, N_1\rangle = P|P, N_1\rangle,$$

$$R_{11}|P, N_1\rangle = N_1|P, N_1\rangle,$$

$$\hat{N}|P, N_1\rangle = N|P, N_1\rangle.$$

Here $R_{ij} = Q_i^+ Q_j$ ($i, j = 1, 2, 3$) are the collective angular momenta of the dressed atoms.

The operators Q_i satisfy the boson commutation relation

$$[Q_i, Q_j^+] = \delta_{ij}, \quad (5)$$

so that

$$[R_{ij}, R_{i'j'}] = R_{ij'} \delta_{i'j} - R_{i'j} \delta_{ij'}. \quad (6)$$

As in reference [25], for simplicity we introduce the characteristic function:

$$\begin{aligned} \chi_{R_{11}, R}(\eta, \xi) &= \langle e^{i\eta R_{11} + i\xi R} \rangle_S = \\ &= A^{-1} \left[\frac{Y_2}{Y_2 - 1} \cdot \frac{(Y_1 Y_2)^{N+1} - 1}{Y_1 Y_2 - 1} \right. \\ &\quad \left. - \frac{1}{Y_2 - 1} \cdot \frac{Y_1^{N+1} - 1}{Y_1 - 1} \right], \quad (7) \end{aligned}$$

where

$$\begin{aligned} Y_1 &= X e^{i\xi}, \\ Y_2 &= Z e^{i\eta}. \end{aligned} \quad (8)$$

Here $\langle B \rangle_s$ denotes the expectation value of an operator B in the steady-state (4).

Once the characteristic function is known, it is easy to calculate the statistical moments:

$$\langle R_{11}^m R^n \rangle_s = \frac{\partial^m}{\partial (i\eta)^m} \cdot \frac{\partial^n}{\partial (i\xi)^n} \chi_{R_{11}, R}(\eta, \xi) \Big|_{i\eta=0, i\xi=0}. \quad (9)$$

3. Photon statistics of spectrum components of scattered light.

In this section we study the influence of collective effects on the photon statistics of the components of the Stokes line. One can find from the canonical transformation (3) that:

$$\begin{aligned} J_{23} &= -\sin \varphi R_{13} + \cos \varphi R_{23}, \\ J_{32} &= -\sin \varphi R_{31} + \cos \varphi R_{32}. \end{aligned} \quad (10)$$

It is easy to see that the operators $R_{13}(t)$ and $R_{23}(t)$ can be considered as the sources of the spectrum components of the Stokes line at frequencies $\omega_{23} - \frac{\delta}{2} - \Omega$ and $\omega_{23} - \frac{\delta}{2} + \Omega$, respectively. For simplicity, we call them $S_{-\Omega}$ and $S_{+\Omega}$ and the steady-state normalized intensity correlation functions of spectrum components $g_{-\Omega}^{(2)}$ and $g_{+\Omega}^{(2)}$. By using the stationary solution (4) and commutation relations (5-6), one can find the correlation functions $g_{\pm\Omega}^{(2)}$ in the form:

$$g_{-\Omega}^{(2)} = \langle R_{13} R_{13} R_{31} R_{31} \rangle_s / \langle R_{13} R_{31} \rangle_s^2, \quad (11)$$

$$g_{+\Omega}^{(2)} = \langle R_{23} R_{23} R_{32} R_{32} \rangle_s / \langle R_{23} R_{32} \rangle_s^2, \quad (12)$$

where

$$\begin{aligned} \langle R_{13} R_{13} R_{31} R_{31} \rangle_s &= \\ &= (N^2 + 3N + 2)(\langle R_{11}^2 \rangle_s - \langle R_{11} \rangle_s^2) - \\ &- (2N + 3)(\langle R_{11}^2 R \rangle_s - \langle R_{11} R \rangle_s^2) \\ &+ \langle R_{11}^2 R^2 \rangle_s - \langle R_{11} R^2 \rangle_s^2, \end{aligned} \quad (13)$$

$$\begin{aligned} \langle R_{23} R_{23} R_{32} R_{32} \rangle_s &= (N^2 + 3N + 2) \times \\ &\times (\langle R^2 \rangle_s - \langle R \rangle_s^2 - 2\langle R_{11} R \rangle_s + \langle R_{11} \rangle_s^2) \\ &+ \langle R_{11}^2 \rangle_s - (2N + 3). \end{aligned} \quad (14)$$

$$\begin{aligned} &(\langle R^3 \rangle_s - \langle R^2 \rangle_s^2 - 2\langle R_{11}^2 R \rangle_s + \langle R_{11} R \rangle_s^2 + \\ &+ \langle R_{11}^2 R \rangle_s) + \langle R^4 \rangle_s - \langle R^3 \rangle_s^2 - 2\langle R_{11} R^3 \rangle_s \\ &+ \langle R_{11} R^2 \rangle_s + \langle R_{11}^2 R \rangle_s, \end{aligned}$$

$$\langle R_{13} R_{31} \rangle_s = (N + 1) \langle R_{11} \rangle_s - \langle R_{11} R \rangle_s, \quad (15)$$

$$\begin{aligned} \langle R_{23} R_{32} \rangle_s &= (N + 1)(\langle R \rangle_s - \langle R_{11} \rangle_s) - \\ &- \langle R^2 \rangle_s + \langle R_{11} R \rangle_s. \end{aligned} \quad (16)$$

Here the value $\langle R_{11}^m R^n \rangle_s$ can be found in equation (9).

The behaviour of the function $g_{\pm\Omega}^{(2)}$ against the parameter $\text{ctg}^2 \varphi$ when $\nu_{31}/\gamma_{23} = 1$ and against the parameter ν_{31}/γ_{23} when $\text{ctg}^2 \varphi = 1$ are plotted in figure 2 and figure 3, respectively. For the one atom case both spectrum components of the Stokes line have subPoissonian statistics ($g_{\pm\Omega}^{(2)} = 0$) for all values of the parameters $\text{ctg}^2 \varphi$ and ν_{31}/γ_{23} . The collective effects reduce the antibunching of spectrum components. For the two-atom case the spectrum compo-

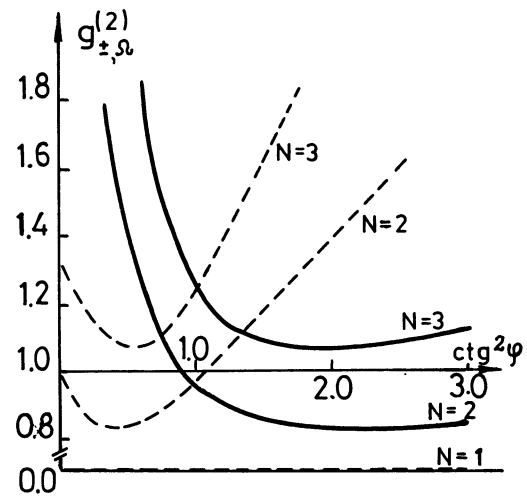


Fig. 2. — Normalized intensity correlation function $g_{-\Omega}^{(2)}$ (solid curves) and $g_{+\Omega}^{(2)}$ (dashed curves) graphed against the parameter $\text{ctg}^2 \varphi$ when $\nu_{31}/\gamma_{23} = 1$.

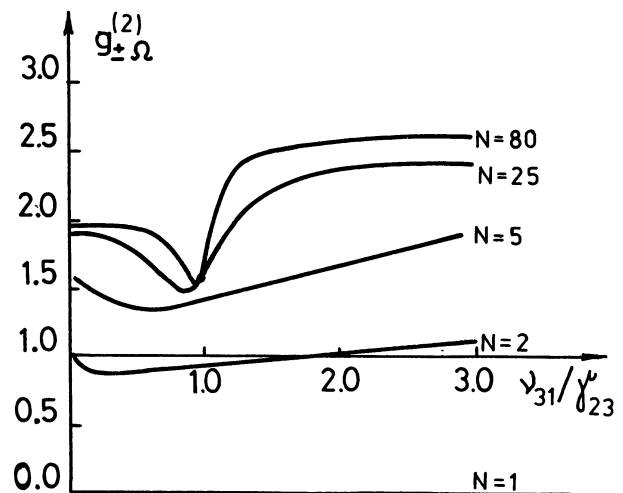


Fig. 3. — Normalized intensity correlation function $g_{\pm\Omega}^{(2)}$ graphed against the parameter ν_{31}/γ_{23} when $\text{ctg}^2 \varphi = 1$.

nents have subPoissonian statistics only for a suitable region of parameters $\text{ctg}^2 \varphi$ or ν_{31}/γ_{23} . It is interesting to note that for the collective case the two spectrum components of Stokes line have the same photon statistics only in the case of resonance $\text{ctg}^2 \varphi = 1$. For some region of the parameter $\text{ctg}^2 \varphi$ the one-spectrum component has the super-Poissonian statistics, while the other has subPoissonian statistics.

4. Cross-correlation between spectrum components of the Stokes line.

More interesting is the question of cross-correlation between the spectrum components. The magnitude of the cross correlation between the spectrum components $S_{+\Omega}$ and $S_{-\Omega}$ can be characterized by the steady-state cross-correlation function $C_{+\Omega, -\Omega}^{(2)}$. By using solution (4) and commutation relations (5-6) one can find :

$$C_{+\Omega, -\Omega}^{(2)} = \langle R_{23} R_{13} R_{31} R_{32} \rangle_S / \langle R_{23} R_{32} \rangle_S \times \langle R_{13} \cdot R_{31} \rangle_S = C_{-\Omega, +\Omega}^{(2)} \equiv C_S^{(2)}, \quad (17)$$

where

$$\begin{aligned} \langle R_{23} R_{13} R_{31} R_{32} \rangle_S &= \langle R_{13} R_{23} R_{32} R_{31} \rangle_S = \\ &= (N^2 + 3N + 2)(\langle R_{11} R \rangle_S - \langle R_{11}^2 \rangle_S) \\ &- (2N + 3)(\langle R_{11} R^2 \rangle_S - \langle R_{11}^2 R \rangle_S) \\ &+ \langle R_{11} R^3 \rangle_S - \langle R_{11}^2 R^2 \rangle_S. \end{aligned} \quad (18)$$

Here the values $\langle R_{23} R_{32} \rangle_S$ and $\langle R_{13} R_{31} \rangle_S$ in equation (17) can be found in equations (15-16) and the statistical moments $\langle R_{11}^m R^n \rangle_S$ in equation (9).

We speak about anticorrelation (cross-antibunching [15]) or correlation (cross-bunching) between spectrum components $S_{\pm\Omega}$ when the cross-correlation function $C_S^{(2)}$ is less or more than unity. The dependence of the function $C_S^{(2)}$ on the parameter $\text{ctg}^2 \varphi$ is shown in figure 4 for the case of $\nu_{31}/\gamma_{23} = 1$ (dashed curves) and $\nu_{31}/\gamma_{23} = 0.4$ (solid curves).

For the one-atom case there is a cross-antibunching between spectrum components ($C_S^{(2)} = 0$) for all values of the parameter $\text{ctg}^2 \varphi$. For the collective case the cross-antibunching between spectrum com-

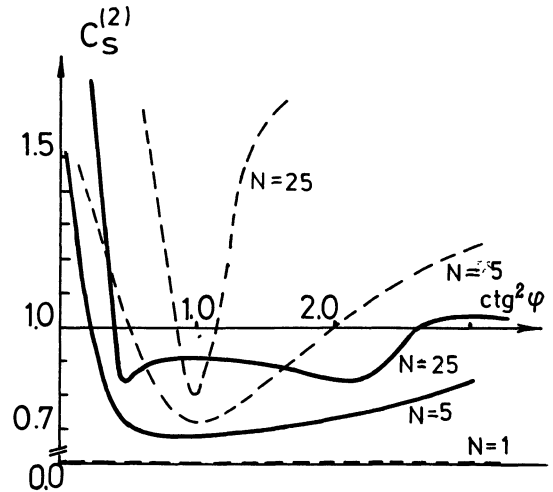


Fig. 4. — Cross-correlation function $C_S^{(2)}$ graphed against the parameter $\text{ctg}^2 \varphi$ for the case of $\nu_{31}/\gamma_{23} = 1$ (dashed curves) and $\nu_{31}/\gamma_{23} = 0.4$ (solid curves).

ponents of the Stokes line is reduced and exists only for a suitable region of the parameter $\text{ctg}^2 \varphi$ (see Fig. 4). Thus, in some region of the parameters $\text{ctg}^2 \varphi$ and ν_{31}/γ_{23} the atoms have a tendency to emit simultaneously only the photon of one spectrum component and in the other region of the parameters $\text{ctg}^2 \varphi$ and ν_{31}/γ_{23} the spectrum components $S_{\pm\Omega}$ have a tendency to be emitted in pairs. As is shown in figure 4, the cross-antibunching is presented for a large number of atoms and in this sense the cross-antibunching is a macroscopic effect.

5. Cross-correlation between the Stokes and Rayleigh lines.

The cross-correlation between the Stokes and Rayleigh line of the Raman scattering for one atom excited by two resonant fields has been considered in reference [23]. In this section we discuss the cross-correlation between the Stokes and Rayleigh lines of the collective Raman scattering. For simplicity we only consider the case of resonance $\text{ctg}^2 \varphi = 1$. By using the canonical transformation (3), stationary solution (4) and commutation relations (5-6), one can find the steady-state cross-correlation function between the Stokes and Rayleigh lines $C_{S,R}^{(2)}$ in the form

$$\begin{aligned} C_{S,R}^{(2)} &= \langle J_{23} J_{21} J_{12} J_{32} \rangle_S / \langle J_{23} J_{32} \rangle_S \cdot \langle J_{21} J_{12} \rangle_S = C_{R,S}^{(2)} = \\ &= \frac{-\langle R^4 \rangle_S + N \langle R^3 \rangle_S + (N+3) \langle R^2 \rangle_S - 2(N+1) \langle R \rangle_S}{(\langle R^2 \rangle_S + 2 \langle R \rangle_S) \cdot ((N+1) \langle R \rangle_S - \langle R^2 \rangle_S)}. \end{aligned} \quad (19)$$

The statistical moments $\langle R^m \rangle_S$ in relation (19) can be found in equation (9).

The dependence of the cross-correlation function $C_{S,R}^{(2)}$ on the parameter ν_{31}/γ_{23} is plotted in figure 5.

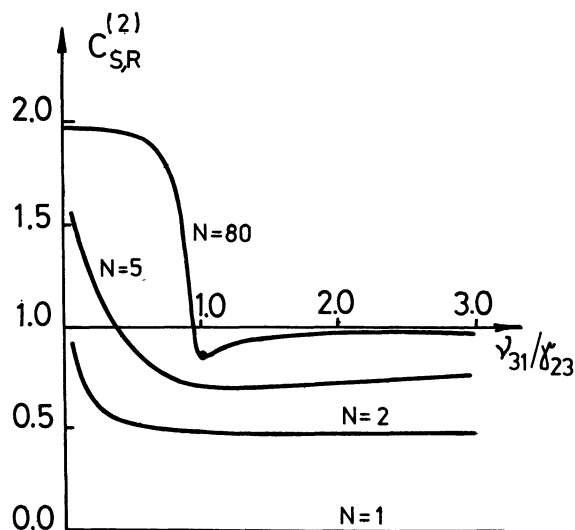


Fig. 5. — Cross-correlation function $C_{S,R}^{(2)}$ graphed against the parameter ν_{31}/γ_{23} for the case of $\text{ctg}^2 \varphi = 1$.

For the one atom case $C_{S,R}^{(2)} = 0$; thus, there is a cross-antibunching between the Stokes and Rayleigh lines for all values of the parameter ν_{31}/γ_{23} ; this conclusion is in agreement with the work by Agarwal and Jha [23]. For the collective case, one can see from figure 5 that the cross-bunching between the Stokes and Rayleigh lines appears in some region of the parameter ν_{31}/γ_{23} and cross-antibunching in the other region.

We mention that the characteristics for the three spectrum components of the Rayleigh line can be obtained using an analogous approach as in sec-

tions 3 and 4. It is interesting to note that the two sidebands located at frequencies $\omega \pm 2\Omega$ of the Rayleigh line have a tendency to be emitted in pairs (cross-bunching) for all numbers of atoms N and all values of the parameters $\text{ctg}^2 \varphi$ and ν_{31}/γ_{23} while the anticorrelation between the central spectrum component located at frequency ω and sidebands exists only for the collective case $N \geq 2$ and in a suitable region of the parameters $\text{ctg}^2 \varphi$ and ν_{31}/γ_{23} ; thus, the cross-correlation functions between the central spectrum component and sidebands of the Rayleigh line provide a new tool for the study of cooperative effects.

6. Conclusion.

In this paper we have presented analytical results for the steady-state correlation of photons in the collective Raman scattering in an intense driving field and investigated the influence of the frequency detuning of resonance relation and other parameters of the system on the statistical properties of the scattering field. The results may be checked by an experiment of the type described by Aspect *et al.* for the resonance fluorescence [2]. In the presence of another field applied to the transition $|2\rangle \rightarrow |3\rangle$, the spectrum picture is complicated and the radiation field has richer statistical properties. The photon statistics of the spectrum components of the collective Raman scattering in two intense exact resonant fields has been discussed in paper [6].

The two time intensity-intensity correlation and the dynamics of the collective Raman scattering are the object of a future investigation.

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