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N.N.Bogolubov,Jr., A.S.Shumovsky, Tran Quang

CORRELATION OF PHOTONS IN COLLECTIVE RAMAN SCATTERING

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I. Introduction

In the last few years the interest in the statistical properties of a photon in various nonlinear optical processes has increased. The antibunching, anticorrelation, superbunching and squeezing have been a subject of various theoretical and experimental works /2-20/. In the theoretical works by Cohen - Tannoudji and Reynaud /3/, Apanasevich and Kilin /4/, and the experimental work by Aspect et al. /2/ the cross-correlation of spectrum components of resonance fluorescence for the one-atom case has been investigated. The recent publications deal with collective effects in photon statistics in resonance fluorescence /8-10/ and double resonance /17/.

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

II. Master equation

We consider a small system (the Dicke model, 1954) of N three-level atoms interacting with a monochromatic driving field of a frequency ω and with a field of radiation (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 $|3\rangle$ and $|1\rangle$ (in the dipole approximation), but the states $|3\rangle$ and $|1\rangle$ are not connected by the dipole Hamiltonian because of parity consideration. The transition $|3\rangle \rightarrow |1\rangle$ 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/

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

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

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

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; $G = -\vec{d}_{21}\cdot\vec{E}_0$ is the matrix element of the driving field and atomic interaction, and

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

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 refs. /23,5/ we introduce the Schwinger representation for angular momentum

$$
J_{ij} = c_i^{+} c_j \qquad (i,j = 1,2,3),
$$

where c_i obey the boson commutation relation

$$
[c_i, c_j^{+}] = \delta_{ij}.
$$

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

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

After performing the canonical transformation

$$
\begin{aligned}
c_1 &= Q_1 \cos\varphi + Q_2 \sin\varphi \\
c_2 &= -Q_1 \sin\varphi + Q_2 \cos\varphi \\
c_3 &= Q_3 ,
\end{aligned}
\tag{3}
$$

where $\operatorname{tg} 2\varphi = \frac{2G}{\delta}$, one can find that the Liouville operator L appearing in eq. (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 of $(\gamma_{21}N/\Omega)^2$, $(\gamma_{23}N/\Omega)^2$ or $(\nu_{31}N/\Omega)^2$.

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

$$
\tilde{\rho} = U\rho U^{+} = A^{-1}\sum_{R=0}^{N} X^{R} \sum_{N_1=0}^{R} Z^{N_1}\, |R, N_1\rangle\langle N_1, R| ,
\tag{4}
$$

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

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

$$Z = ctg^4 \varphi$$

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

$|R, N_1\rangle$ is an eigenstate of the operator $R = R_{11} + R_{22}$, R_{11} and the operator of a number of atoms

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

Here $\qquad R_{ij} = Q_i^+ Q_j \qquad (i, j = 1, 2, 3)$

The operators Q_i satisfy the boson commutation relation

$$[Q_i, Q_j] = \delta_{ij} \tag{5}$$

so that

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

As in ref./24/, for simplicity we introduce the characteristic function

$$\chi_{R_{11}, R}(\xi, \zeta) = \langle e^{i\xi R_{11} + i\zeta R} \rangle_s =$$

$$= A^{-1} \left[\frac{Y_2}{Y_2 - 1} \cdot \frac{(Y_1 Y_2)^{N+1} - 1}{Y_1 Y_2 - 1} - \frac{1}{Y_2 - 1} \cdot \frac{Y_1^{N+1} - 1}{} \right] , \tag{7}$$

where $\qquad Y_1 = X e^{i\zeta}$

$$Y_2 = Z \cdot e^{i\xi} . \tag{8}$$

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

$$\langle R_{11}^m R^n \rangle = \frac{\partial^m}{\partial (i\xi)^m} \cdot \frac{\partial^n}{\partial (i\zeta)^n} \chi_{R_{11}, R}(\xi, \zeta) \Big|_{\substack{i\xi = 0 \\ i\zeta = 0}} \tag{9}$$

III. Intensity correlation of spectrum components of scattered light

In this section we study the influence of collective effects on the intensity correlation of the components of the steady-state spectrum of the Stoke line. One can find from canonical transformation (3) that

$$J_{23} = -\sin\varphi \, R_{13} + \cos\varphi \, R_{23}$$

$$J_{32} = -\sin\varphi \, R_{31} + \cos\varphi \, R_{32} . \tag{10}$$

It is easy to see that the operators $R_{13}(t)$ and $R_{23}(t)$ can be considered as the sources of spectrum components of the Stoke 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 $g^{(2)}_{-\Omega}$ and $g^{(2)}_{+\Omega}$. By using the stationary solution (4) and commutation relations (5-6), one can find correlation functions $g^{(2)}_{\pm\Omega}$ in the form

$$g^{(2)}_{-\Omega} = \langle R_{13} R_{13} R_{31} R_{31} \rangle_s / \langle R_{13} R_{31} \rangle_s^2 \tag{11}$$

$$g^{(2)}_{+\Omega} = \langle R_{23} R_{23} R_{32} R_{32} \rangle_s / \langle R_{23} R_{32} \rangle_s^2 , \tag{12}$$

where $\langle R_{13} R_{13} R_{31} R_{31} \rangle_s = (N^2 + 3N + 2)(\langle R_{11}^2 \rangle_s - \langle R_{11} \rangle_s) -$

$$- (2N+3)(\langle R_{11}^2 R \rangle_s - \langle R_{11} R \rangle_s) + \langle R_{11}^2 R^2 \rangle_s - \langle R_{11} R^2 \rangle_s \tag{13}$$

$$\langle R_{23}R_{23}R_{32}R_{32}\rangle_s = (N^2+3N+2)(\langle R^2\rangle_s - \langle R\rangle_s - 2\langle R_{11}R\rangle_s + \langle R_{11}\rangle_s + \langle R_{11}^2\rangle_s) - (2N+3)(\langle R^3\rangle_s - \langle R^2\rangle_s - 2\langle R_{11}^2 R\rangle_s + \langle R_{11}R\rangle_s + \langle R_{11}^2 R\rangle_s) + \langle R^4\rangle_s - \langle R^3\rangle_s - 2\langle R_{11}R^3\rangle_s + \langle R_{11}R^2\rangle_s + \langle R_{11}^2 R^2\rangle_s , \tag{14}$$

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

$$\langle R_{23}R_{32}\rangle_s = (N+1)(\langle R\rangle_s - \langle R_{11}\rangle_s) - \langle R^2\rangle_s + \langle R_{11}R\rangle_s . \tag{16}$$

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

The behaviour of the function $g^{(2)}_{\pm\Omega}$ against the parameter $ctg^2\varphi$ when $\nu_{31}/\gamma_{23}=1$ and against the parameter ν_{31}/γ_{23} when $ctg^2\varphi = 1$ are plotted in fig.2. and fig. 3, respectively. For the one atom case all two spectrum components of the Stoke line have subpoissonian statistics $(g^{(2)}_{\pm\Omega}=0)$ for all values of parameters $ctg^2\varphi$ and ν_{31}/γ_{23}. The collective effects reduce the antibunching of spectrum components. For the atom case the spectrum components have subpoissonian statistics only for a suitable region of parameters $ctg^2\varphi$ or ν_{31}/γ_{23}. It is interesting to note that two spectrum components of Stokes line have the same photon statistic only in the case of resonance $ctg^2\varphi = 1$. For some region of the parameter $ctg^2\varphi$ the one-spectrum component has the superpoissonian statistics, while the other, has subpoissonian statistics.

IV. Cross-correlation between spectrum components of the Stoke line

More interesting is the questin 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. By using solution (4) and commutation relation (5-6), one can find

$$C^{(2)}_{+\Omega,-\Omega} = \langle R_{23}R_{13}R_{31}R_{32}\rangle_s / \langle R_{23}R_{32}\rangle_s \langle R_{13}R_{31}\rangle_s = C^{(2)}_{-\Omega,+\Omega} \equiv C^{(2)}_s , \tag{17}$$

where

$$\langle R_{23}R_{13}R_{31}R_{32}\rangle_s = \langle R_{13}R_{23}R_{32}R_{31}\rangle_s = (N^2+3N+2)(\langle R_{11}R\rangle_s - \langle R_{11}^2\rangle_s) - (2N+3)(\langle R_{11}R^2\rangle_s - \langle R_{11}^2 R\rangle_s) + \langle R_{11}R^3\rangle_s - \langle R_{11}^2 R^2\rangle_s . \tag{18}$$

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 eqs. (15-16); and the statistical moments $\langle R_{11}^m R^n\rangle_s$ in eq. (9).

We speak about anticorrelation (cross-antibunching /15/) or correlation (cross-bunching) between spectrum components $S_{\pm\Omega}$ when the cross-correlation function $C^{(2)}_s$ is less or more than unity. The dependence of the function $C^{(2)}_s$ on the parameter $ctg^2\varphi$ is shown in fig.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 the cross-antibunching between spectrum components comes into existence ($C^{(2)}_s = 0$) for all values of the parameter $ctg^2\varphi$. For the collective case the cross-antibunching between spectrum components of the Stoke line reduce and come into existence only for a suitable region of the parameter $ctg^2\varphi$ (see fig.4). Thus, in some region of the paramters $ctg^2\varphi$ and ν_{31}/γ_{23} the atoms have a terdency to emit simultaneously only the photon of one spectrum component and in the other region of the parameters $ctg^2\varphi$ and ν_{31}/γ_{23} the spectrum components $S_{\pm\Omega}$ have a tendency to be emitted in pairs. However, it is interesting to note that the cross-antibunching, contrast with the antibunching effect, is the macroscopic effect. The

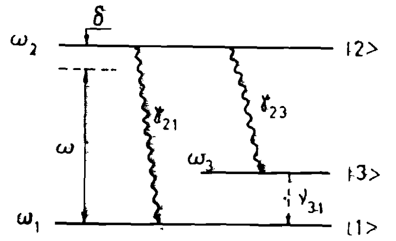

Fig. 1. Three-level system of atom interacting with the monochromatic applied field.

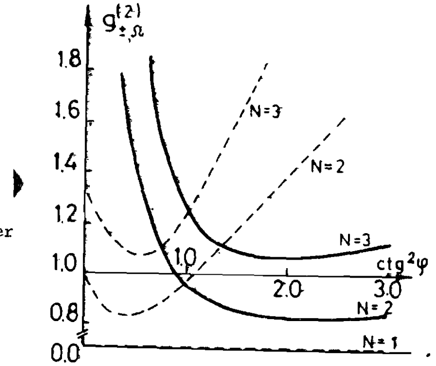

Fig. 2. Normalized intensity correlation function $g^{(2)}_{-\Omega}$ (solid curves) and $g^{(2)}_{+\Omega}$ (dashed curves) graphed againt tne parameter $ctg^2\varphi$ when $\nu_{31}/\gamma_{23} = 1$.

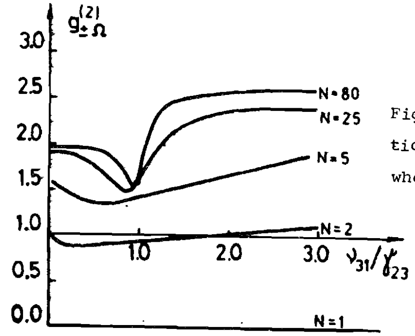

Fig. 3. Normalized intensity correlation function $g^{(2)}_{\pm\Omega}$ graphed against the parameter ν_{31}/γ_{23} when $ctg^2\varphi = 1$.

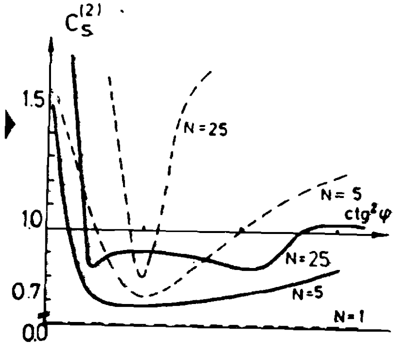

Fig. 4. Cross-correlation function $C^{(2)}_S$ graphed against the parameter $ctg^2\varphi$ for the case of $\nu_{31}/\gamma_{23} = 1$ (dashed curves) and $\nu_{31}/\gamma_{23} = 0.4$ (solid curves).

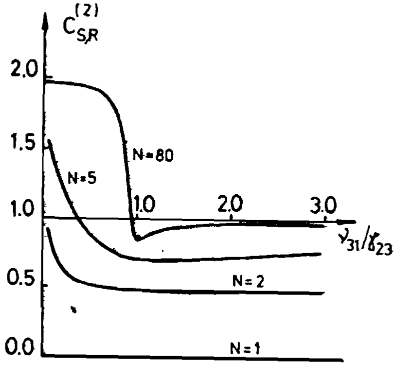

Fig. 5. Cross-correlation function $C^{(2)}_{S,R}$ graphed against the parameter ν_{31}/γ_{23} for the case of $ctg^2\varphi = 1$.

cross-antibunching between spectrum components of Stoke line, as is shown in fig.4, is presented for a large number of atoms. By using eqs. (17-18), one can show that in the cooperative limit $N \to \infty$ the cross-antibunching between spectra $S_{\pm\Omega}$ comes into existence ($C^{(2)}_S$ =0.8) only for the case of $ctg^2\varphi = 1$ and $\nu_{31}/\gamma_{23} = 1$.

V. <u>Cross-correlation between the Stoke and Rayleight lines</u>

The cross-correlations between the emission of photons from an excited three-level atom with one transition more strongly driven than the other studies have recently been studied in the paper /15/. In this section we discuss cross-correlation between the Stoke and Rayleight lines of the collective Raman scattering.

For simplicity we consider only the case of resonance $ctg^2\varphi = 1$. 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 Stoke and Rayleight line $C^{(2)}_{S,R}$ in the form

$$C^{(2)}_{S,R} = \langle J_{23} J_{21} J_{12} J_{32} \rangle_S / \langle J_{23} J_{32} \rangle_S \cdot \langle J_{21} J_{12} \rangle_S = C^{(2)}_{R,S}$$

$$= \frac{-\langle R^4\rangle_S + N\langle R^3\rangle_S + (N+3)\langle R^2\rangle_S - 2(N+1)\langle R\rangle_S}{(\langle R^2\rangle_S + 2\langle R\rangle_S)\,((N+1)\langle R\rangle_S - \langle R^2\rangle_S)} . \qquad (19)$$

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

The dependence of the cross-correlation function $C^{(2)}_{S,R}$ on the parameter ν_{31}/γ_{23} is plotted in fig.5. For the one atom case $C^{(2)}_{S,R} = 0$; thus, the cross-antibunching between the Stoke and Rayleight lines comes into existence for all values of the parameter ν_{31}/γ_{23} . For the collective case, one can see from fig.5 that the cross-bunching between the Stoke and Rayleight lines appear in some region of the parameter ν_{31}/γ_{23} and cross-antibunching between them takes the place in the other region of the parameter.

In conclusion, we have to show that the characteristics for the three spectrum components of the Rayleight line can be obtained using an analogous approach as in sections III and IV. It is interesting to note that the two sidebands located at frequencies $\omega \pm 2\Omega$ of the Rayleight line have a tendency to be emitted in pairs (cross-bunching) for all numbers of atoms N and parameters $c\,tg^2\varphi$ and ν_{31}/γ_{23} while the anticorrelation between the central spectrum component located at frequency ω and sidebands comes into existence for the collective case $N \geq 2$ in a suitable region of the parameters $c\,tg^2\varphi$ and ν_{31}/γ_{23} ; thus, the cross-antibunching between the central spectrum component and sidebands of the Rayleight line can be considered as characteristics of collective effects.

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Боголюбов Н.Н. (мл.), Шумовский А.С., Чан Куанг E4-86-703

Корреляция фотонов в коллективном рассеянии Рамана

Исследована статистика фотонов спектральных компонентов линии в коллективном рассеянии Рамана. Обсуждены кросс-группировка и кросс-антигруппировка между спектральными компонентами стоксовой линии между стоксовой и рэлеевской линиями.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

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Correlation of Photons in Collective Raman Scattering

The photon statistics of spectrum components of the Stoke line in collective Raman scattering is investigated. The cross-bunching and cross-antibunching between spectrun components of the Stoke line and between the Stoke and Rayleyght lines are investigated too.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

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