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STATISTICAL PROPERTIES OF PHOTONS IN COLLECTIVE RESONANT RAMAN SCATTERING

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Statistical properties of the photons in collective resonant Raman scattering are investigated. The anticorrelation between Stokes and Rayleigh lines is observed.

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

Статистические свойства фотонов в коллективном резонансном рассеянии Рамана

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Исследованы статистические свойства фотонов в коллективном резонансном рассеянии Рамана. Наблюдена антикорреляция между стоксовой и рэлеевской линиями.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

The photon statistics of various nonlinear optical processes has been a subject of increasing interest in recent years [2-17]. In particular, the photon statistics for stimulated Raman scattering has been analysed in works [8,13].

In the present paper we consider the photon statistics for collective resonant Raman scattering (Fig.1). It will be shown that under a suitable condition the anticorrelation bitween Stokes and Rayleigh lines is observed.

We consider a small system (the Dicke model, 1954) of N three-level atoms interacting with a resonant driving field of the frequency ω and with a field of radiation (Fig.1). Let us label the ground state by $|1\rangle$; the real excited state by $|3\rangle$

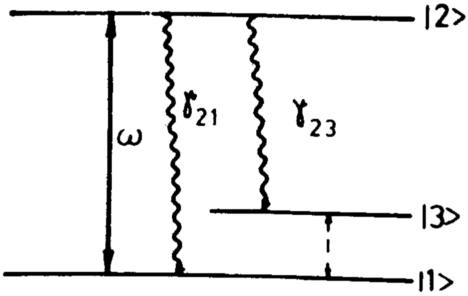

Fig.1. Three-level system of atoms interacting with the monochromatic applied field.

and the resonant intermediate state by $|2\rangle$ with energies ω_1, ω_3 and ω_2, respectively (the system of $h = 1$). 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 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. The transition $|3\rangle \to |1\rangle$ is caused by an atomic reservoir and assumed to be nonradiative [18]. For simplicity the external driving field is assumed to be in resonance with the level separation $\omega_2 - \omega_1 = \omega_{21} = \omega$.

In treating the external field classically and using the Born and Markov approximation, one can obtain a master equation for the reduced density matrix ρ for the system alone in the form [1,8].

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

where $2\gamma_{21}$ and $2\gamma_{23}$ are 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 nonradiative rate for transition $|3\rangle$ to $|1\rangle$. The coherence part of Hamiltonian H_{coh} in the interaction picture has the form

$$H_{coh} = \Omega_3 J_{33} + \Omega (J_{21} + J_{12}),$$

where $\Omega_3 = \frac{\omega_{21}}{2} - \omega_{23}$; ω is the Radi frequency for the atomic transition from the level $|2\rangle$ to $|1\rangle$. And $J_{ij} = \sum_{k=1}^{N} |i\rangle_k \langle j|_k$ $(i, j = 1, 2, 3)$ 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.[6,19], we shall use the Schwinger representation for the angular momentum

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

where C_j obey the boson commutation relation

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

Further, we investigate only the case of an intense external field so that

$$\Omega \gg N\gamma_{21}, N\gamma_{23} \quad \text{and} \quad N\nu_{31} . \qquad (2)$$

After performing the canonical transformation

$$C_1 = \frac{1}{\sqrt{2}} Q_1 + \frac{1}{\sqrt{2}} Q_2$$

$$C_2 = -\frac{1}{\sqrt{2}} Q_1 + \frac{1}{\sqrt{2}} Q_2 \qquad (3)$$

$$C_3 = Q_3$$

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, one can make the secular approximation, i.e., retain only a slowly varying part[11,6]. Correction to 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

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

where U is a unitary operator representing the canonical transformation (3); $X = \nu_{31}/\gamma_{23}$

$$Z = \frac{(N+1)X^{N+2} - (N+2)X^{N+1} + 1}{(X-1)^2} ,$$

$|R, N_1\rangle$ is an eigenstate of the operator $R = R_{22} + R_{11}$, R_{11} and $\hat{N} = R_{11} + R_{22} + R_{33}$. Here $R_{ij} = Q_i^+ Q_j$ $(i, j = 1, 2, 3)$. The operators Q_i satisfy the boson commutation relation

$$[Q_i , Q_j^+] = \delta_{ij}$$

so that

$$[R_{ij} , R_{i'j'}] = R_{ij'} \delta_{i'j} - R_{i'j} \delta_{ij'} . \qquad (5)$$

From solution (4) it is easy to see that the stationary characteristics of the system depend only on the number of atoms N and the relation of spontaneous transition probability γ_{23} and nonradiative rate ν_{31}.

By using solution (4) one can define the characteristic function [20]

$$\chi_R(\xi) = \langle e^{i\xi R} \rangle_s = Z^{-1} \frac{(N+1)Y^{N+2} - (N+2)Y^{N+1} + 1}{(Y-1)^2},$$

where $Y = xe^{i\xi}$. Here $\langle A \rangle_s$ denotes the expectation value of an operator A in the steady state (4).

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

$$\langle R^n \rangle_s = \frac{\partial^n}{\partial(i\xi)^n} \chi_R(\xi) \Big|_{i\xi=0}. \tag{6}$$

Now, we discuss the influence of collective effects and relation $X = \frac{\nu_{31}}{\gamma_{23}}$ on the photon statistics of the Stokes line and the cross-correlation between Stokes and Rayleigh lines.

By using the canonical transformation (3), stationary solution (4) and commutation relation (5) one can find the steady-state normalized intensity correlation function of Stokes line $g^{(2)}_{s,s}$ and the cross-correlation function between Stokes and Rayleigh lines $C^{(2)}_{S,R}$ in the form

$$g^{(2)}_{s,s} = \langle J_{23} J_{23} J_{32} J_{32} \rangle_s / \langle J_{23} J_{32} \rangle_s^2$$

$$= \frac{4}{3} \frac{\langle R^4 \rangle_s - 2(N+2)\langle R^3 \rangle_s + (N^2+5N+5)\langle R^2 \rangle_s - (N+1)(N+2)\langle R \rangle_s}{(\langle R^2 \rangle_s - (N+1)\langle R \rangle_s)^2} \tag{7}$$

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

$$= \frac{-\langle R^4 \rangle_s + N\langle R^3 \rangle_s + (N+3)\langle R^2 \rangle_s - 2(N+1)\langle R \rangle_s}{\langle R^2 + 2R \rangle_s \cdot \langle (N+1)R - R^2 \rangle_s} \tag{8}$$

The statistical moments $\langle R^n \rangle_s$ in relations (7,8) can be found in equation (6).

The dependence of the normalized intensity correlation function $g^{(2)}_{s,s}$ and cross-correlation function $C^{(2)}_{S,R}$ on the

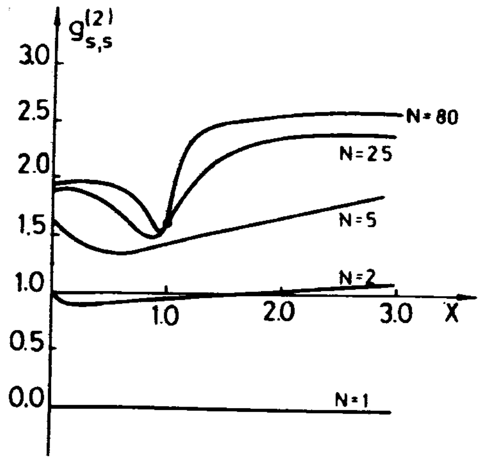

Fig. 2. Normalized intensity correlation function $g^{(2)}_{s,s}$ graphed against the parameter X.

parameter X is plotted in Figs. 2 and 3, respectively. From these Figures one can see that:

i) For the one-atom case $g^{(2)}_{s,s} = 0$ and $C^{(2)}_{S,R} = 0$, thus the Stokes line has subpoissonian statistics and the anticorrelation between Stokes and Rayleigh lines comes into existence for all values of the parameters X.

ii) The collective effects reduce the antibunching of Stokes line. For the case of several atoms, the Stokes line has subpoissonian statistics ($g^{(2)}_{s,s} < 1$) only for a suitable region of the parameter X and for the case of $N \geq 5$ Stokes line has superpoissonian statistics ($g^{(2)}_{s,s} > 1$) for all values of the parameter X.

iii) For a suitable region of the parameter X.(Fig.3) the anticorrelation between Stokes and Rayleigh lines ($C^{(2)}_{S,R} < 1$) comes into existence for the various number of atoms N. For the collective limit $N \to \infty$ the anticorrelation between Stokes and Rayleigh lines is presented only for the case of $X = 1 (C^{(2)}_{S,R} = 0.8)$.

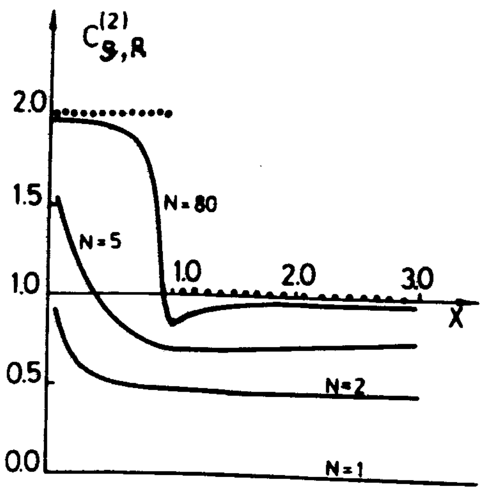

Fig. 3. Cross-correlation function $C^{(2)}_{S,R}$ graphed against the parameter X. *The dotted curve indicates the case of* $N \to \infty$.

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