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COLLECTIVE EFFECTS
IN RESONANT RAMANN SCATTERING
OF INTENSE OPTICAL WAVES

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1. Introduction

Many theoretical and experimental studies[2-15] of the coopera-
tive effects in the interaction of atoms and molecules with a laser
field and the vacuum of radiation have been carried out since the
early work on superradiance by Dicke[1].

In this work the theory of collective resonant Raman scattering
in the presence of intense incident and scattered light waves (fig.1)
has been developed by using the quantum-mechanical master equation
approach and secular approximation[3]. The analytic expressions for
the steady state collective spectrum of scattered light are given.
The intensity correlation for the spectrum components of scattered
light is investigated too. In the cooperative limit $N \to \infty$ the
intensities and normalized intensity correlation functions for each
component of the spectra have a discontinuous behaviour reminiscent
of a typical nonequilibrium first-order phase transition[21]

II. Master Equation

The N three-level molecules are assumed to be concentrated in
a region small compared to the wavelength of all the relevant radi-
ation modes.

Let us label the ground state $|1\rangle$, the real excited state
$|3\rangle$ and the resonant intermediate state $|2\rangle$ with energies $\hbar \omega_1$,
$\hbar \omega_3$ and $\hbar \omega_2$, respectively (fig.1). The real excited state
$|3\rangle$ may be a low-lying vibrational or rotational excitation from the
ground state. In order to keep the discussion general, we will not
specify these states beyond saying that the intermediate state $|2\rangle$
can be connected via the electromagnetic interaction Hamiltonian to
both the state $|1\rangle$ and $|3\rangle$ (in the dipole approximation) but the
states $|3\rangle$ and $|1\rangle$ are not connected by the dipole Hamiltonian beca-
use of parity consideration. The transitions $|3\rangle \to |1\rangle$ and $|1\rangle \to |3\rangle$
are caused by an atomic reservoir and assumed to be nonradiative[16].

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In treating the external fields 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[2]

$$\frac{\partial \rho}{\partial t} = -i\left[H_{coh}, \rho\right] - \gamma_{21}\left(J_{21}J_{12}\rho - 2J_{12}\rho J_{21} + \rho J_{21}J_{12}\right)$$
$$- \gamma_{23}\left(J_{23}J_{32}\rho - 2J_{32}\rho J_{23} + \rho J_{23}J_{32}\right)$$
$$- \nu_{31}\left(J_{31}J_{13}\rho - 2J_{13}\rho J_{31} + \rho J_{31}J_{13}\right) \qquad (1)$$
$$- \nu_{13}\left(J_{13}J_{31}\rho - 2J_{31}\rho J_{13} + \rho J_{13}J_{31}\right),$$

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 $|2\rangle$ to $|3\rangle$, respectively. $2\nu_{31}$ and $2\nu_{13}$ are nonradiative rates for transitions $|3\rangle$ to $|1\rangle$ and $|1\rangle$ to $|3\rangle$, respectively.

$$H_{coh} = \Omega\left(\cos\alpha\, J_{12} + \sin\alpha\, J_{32} + H.C.\right),$$

where $\Omega = (\Omega_1^2 + \Omega_2^2)^{1/2}$ and $tg\alpha = \Omega_2/\Omega_1$. Here Ω_1 and Ω_2 are the Rabi frequencies for the atomic transitions from the level $|2\rangle$ to $|1\rangle$ and $|2\rangle$ to $|3\rangle$, respectively. And $J_{ij} = \sum_{k=1}^{N} |i\rangle_k \langle j|$ ($i,j = 1,2,3$) are the collective angular momenta of the atoms.

$$[J_{ij}, J_{i'j'}] = J_{ij'}\delta_{ji'} - J_{i'j}\delta_{ij'} \qquad (i,j = 1,2,3).$$

The atomic coherence phenomena can parhaps be illustrated with greater licidity by introducing the Schwinger representation for angular momentum[4,14,20].

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

where c_i obey boson commutation relation.

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

In the case of sufficiently intense Ω, so that

$$\Omega \gg N\gamma_{21}, N\gamma_{23}, N\nu_{31} \qquad \text{and} \qquad N\nu_{13} \qquad (2)$$

it is possible to develope an approximation scheme that enables us to obtain analytic results.

After performing the canonical transformation

$$C_3 = -\frac{\sin\alpha}{\sqrt{2}}Q_1 + \cos\alpha\, Q_2 + \frac{\sin\alpha}{\sqrt{2}}Q_3$$
$$C_2 = \frac{1}{\sqrt{2}}Q_1 + \frac{1}{\sqrt{2}}Q_3 \qquad (3)$$
$$C_1 = -\frac{\cos\alpha}{\sqrt{2}}Q_1 - \sin\alpha\, Q_2 + \frac{\cos\alpha}{\sqrt{2}}Q_3$$

one can find that the Liouville operator L appearing in equation (1) splits into two components L_o and L_1. The component L_o is slowly varying in time whereas L_1 contains rapidly oscillating terms at frequencies 2Ω and 4Ω. For intense fields, it is reasonable to make the secular approximation, i.e., to retain only the slowly varying part[3]. Corrections to the results obtained in this fashion will be of an order of $(\gamma_{21}N/\Omega)^2$,

$$(\gamma_{23}N/\Omega)^2, (\nu_{13}N/\Omega)^2 \qquad \text{or} \qquad (\nu_{31}N/\Omega)^2.$$

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

$$\tilde{\rho} = U\rho U^+ = A\sum_{S=0}^{N} Z^S \sum_{M=-S,-S+2,\cdots}^{S} |S,M\rangle\langle M,S|, \qquad (4)$$

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

$$Z = \frac{\nu_{31}\cos^4\alpha + \nu_{13}\sin^4\alpha}{\gamma_{21}\sin^2\alpha + \gamma_{23}\cos^2\alpha + \nu_{31}\sin^4\alpha + \nu_{13}\cos^4\alpha}$$

$$A = (Z-1)^2 / \left[(N+1)Z^{N+2} - (N+2)Z^{N+1} + 1\right].$$

$|S,M\rangle$ is an eigenstate of the operators $R = R_{33} + R_{11}$, $\Delta_3 = R_{33} - R_{11}$ and the operator of total number of atoms

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

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

The operators Q_i satisfy the boson commutation relation

$$[a_i, a_j^+] = \delta_{ij}$$

so
$$[R_{ij}, R_{i'j'}] = R_{ij'}\delta_{i'j} - R_{i'j}\delta_{ij'}. \tag{5}$$

In the case of $N = 1$, the solution (4) reduces to the single atom solution of Agarwal and Jha[16].

By using eq.(4), the characteristic function can be defined similar to Louisell

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

where $\quad Y = Z \cdot e^{i\xi}.$

Here $\langle B\rangle_s$ indicates 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

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

In particular, we find

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

$$\langle R^2\rangle_s = \Big[N^2(N+1)Z^{N+4} - N(3N^2+6N-1)Z^{N+3} +$$
$$+ (N+2)(3N^2+3N-2)Z^{N+2} - (N+1)^2(N+2)Z^{N+1} + 4Z^2 + 2Z\Big]\Big/ \tag{8}$$
$$\Big/\left[(Z-1)^2\left((N+1)Z^{N+2} - (N+2)Z^{N+1} + 1\right)\right].$$

In the case of $Z = 1$ the relations (7-8) reduce to

$$\langle R\rangle_s = \frac{2}{3}N, \tag{9}$$

$$\langle R^2\rangle_s = \frac{1}{2}N(N+\frac{1}{5}). \tag{10}$$

III. Spectrum of Scattered Light

Due to Agarwal and Jha[16] the steady state spectrum of the spontaneous emission corresponding to transition $|2\rangle \to |3\rangle$ (Stokes lines) is proportional to the Fourier transform of the atomic correlation function

$$\langle J_{23}(\tau)J_{32}\rangle_s = \lim_{t\to\infty}\langle J_{23}(t+\tau)J_{32}(t)\rangle.$$

Using the secular approximation and the quantum regression theorem[18], one can find the equations of motion for the correlation functions $\langle R_{ij}(\tau)J_{32}\rangle_s$ in the form

$$\frac{d}{d\tau}\langle \Delta_3(\tau)J_{32}\rangle_s = \langle\Gamma_1(\tau)\Delta_3(\tau)J_{32}\rangle_s$$

$$\frac{d}{d\tau}\langle R_{12}(\tau)J_{32}\rangle_s = -i\Omega\langle R_{12}(\tau)J_{32}\rangle_s - \langle\Gamma_2(\tau)R_{12}(\tau)J_{32}\rangle_s$$

$$\frac{d}{d\tau}\langle R_{32}(\tau)J_{32}\rangle_s = i\Omega\langle R_{32}(\tau)J_{32}\rangle_s - \langle\Gamma_2(\tau)R_{32}(\tau)J_{32}\rangle_s \tag{11}$$

$$\frac{d}{d\tau}\langle R_{13}(\tau)J_{32}\rangle_s = -2i\Omega\langle R_{13}(\tau)J_{32}\rangle_s - \langle\Gamma_3(\tau)R_{13}(\tau)J_{32}\rangle_s,$$

where

$$\Gamma_1(\tau) = \gamma_{21} + \gamma_{23}\sin^2\alpha + \nu_{31}\sin^2\alpha + \nu_{13}\cos^2\alpha +$$
$$+ \frac{Z(1-Z)R_{22}(\tau)}{\nu_{31}\cos^4\alpha + \nu_{13}\sin^4\alpha}. \tag{12}$$

$$\Gamma_2(\tau) = \frac{1}{2}\gamma_{21}(1+\sin^2\alpha) + \frac{1}{2}\gamma_{23}(1+\cos^2\alpha) +$$
$$+ \nu_{13}\cos^2\alpha(1+\frac{3}{2}\sin^2\alpha) + \nu_{31}\sin^2\alpha(1+\frac{3}{2}\cos^2\alpha) +$$
$$+ \frac{Z(1-Z)(R_{22}(\tau)-\frac{N}{2})}{\nu_{31}\cos^4\alpha + \nu_{13}\sin^4\alpha}. \tag{13}$$

$$\Gamma_3(\tau) = \gamma_1(1+\frac{\cos^2\alpha}{2}) + \gamma_{23}(1+\frac{\sin^2\alpha}{2}) + \nu_{31}\sin^2\alpha(1-\frac{1}{2}\cos^2\alpha)$$
$$+ \nu_{13}\cos^2\alpha(1-\frac{1}{2}\sin^2\alpha) + \frac{Z(1-Z)R_{22}(\tau)}{\nu_{31}\cos^4\alpha + \nu_{13}\sin^4\alpha}. \tag{14}$$

By analogy with Compagno and Persico[6] we factorize

$$\langle \Gamma_\mu(\tau) R_{ij}(\tau) J_{32} \rangle_s = \langle \Gamma_\mu \rangle_s \langle R_{ij}(\tau) J_{32} \rangle_s . \qquad (15)$$

Using the factorization (15) and solution (4), one can find the solutions of equation (11) and write the atomic correlation function in the form

$$\langle J_{23}(\tau) J_{32} \rangle_s = \frac{1}{4} \sin^2 \alpha \langle \Delta_3^2 \rangle_s \, e^{-\langle \Gamma_1 \rangle_s \tau} +$$
$$+ \cos^2 \alpha \langle R_{12} R_{21} \rangle_s \, e^{-\langle \Gamma_2 \rangle_s \tau} \cos(\Omega \tau) \qquad (16)$$
$$+ \frac{1}{2} \sin^2 \alpha \langle R_{31} R_{13} \rangle_s \, e^{-\langle \Gamma_3 \rangle_s \tau} \cos(2\Omega \tau),$$

where

$$\langle \Delta_3^2 \rangle_s = 2 \langle R_{31} R_{13} \rangle_s = \frac{1}{3} \left(2 \langle R \rangle_s + \langle R^2 \rangle_s \right) \qquad (17)$$

$$\langle R_{12} R_{21} \rangle_s = \frac{1}{2}(N+1) \langle R \rangle_s - \frac{1}{2} \langle R^2 \rangle_s . \qquad (18)$$

Expression (16) yields the give-peaked structure of spectrum of Stokes line.

It is easy to show, using the relations (9-10), (11-14) and (16-18), that in the case of $Z = 1$ the peak intensity of each spectrum component of Stoke line in (16) varies as N^2 while the width of each component is the same as in the single-molecule spectrum. An analogous behaviour for the triplet of resonance fluorescence in the two-level system is discussed by Agarwal et al.[3], Compagno and Persico[6]. The picture changes for the case of $Z \neq 1$ and the number of atoms N is large enough. For the case of $Z < 1$ the widths of all spectrum components of Stoke line vary as N. The peak intensities of the three lines located at the frequencies ω_{23}, $\omega_{23} \pm 2\Omega$ are independent of N while the peak intensities of the two lines located at the frequencies $\omega_{23} \pm \Omega$ are proportional to N. For the case of $Z > 1$ the widths of the three lines located at the frequencies ω_{23}, $\omega_{23} \pm 2\Omega$ are independent of N and their peak intensities very as N^2 while the widths and peak intensities of the two lines located at the frequencies $\omega_{23} \pm \Omega$ are proportional to N.

Fig. 1. Schematic representation of three-level system interacting with resonant incident and scattered coherent waves.
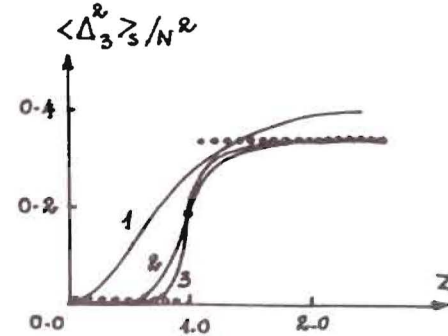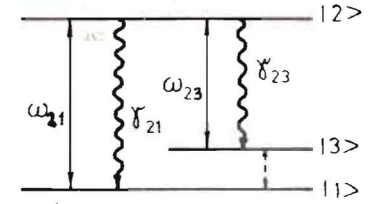

Fig. 2. Relative intensity ($\langle \Delta_3^2 \rangle_s / N^2$) of spectrum components located at frequencies ω_{23}, $\omega_{23} \pm 2\Omega$ as function of Z. Curves marked 1 to 3 correspond to $N = 5, 25, 50$, respectively. The dotted curve indicates the behaviour as $N \to \infty$.
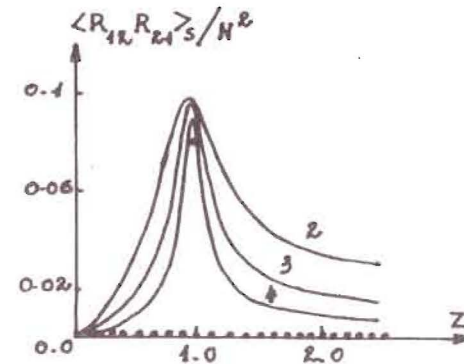

Fig. 3. Relative intensity ($\langle R_{12} R_{21} \rangle_s / N^2$) of spectrum components located at frequencies $\omega_{23} \pm \Omega$ as function of Z. Curves marked 2 to 4 correspond to $N = 25, 50, 100$, (respectively). The dotted curve indicates the behaviour as $N \to \infty$.

The detailed behaviour of the functions $\langle \Delta_3^2 \rangle_s / N^2$ and $\langle R_{12} R_{21} \rangle_s / N^2$ which are proportional to intensities per atom of the three lines located at the frequencies ω_{23}, $\omega_{23} \pm 2\Omega$ and of the two lines located at the frequencies $\omega_{23} \pm \Omega$, respectively, are shown in figs.2 and 3.

For all finite values of N, one observes a smooth variation of functions $\langle \Delta_3^2 \rangle_s / N^2$ and $\langle R_{12} R_{21} \rangle_s / N^2$ with Z. For the cooperative limit $N \to \infty$, one can find from relations (7-10) and (17,18) that

$$\langle \Delta_3^2 \rangle_s / N^2 = \begin{cases} 1/3 & \text{if} \quad Z > 1 \\ 1/6 & \quad Z = 1 \\ 0 & \quad Z < 1 \end{cases} \qquad (19)$$

and

$$\langle R_{12} R_{21} \rangle_s / N^2 = \begin{cases} 0 & \text{if} \quad Z > 1 \\ 1/12 & \quad Z = 1 \\ 0 & \quad Z < 1 \end{cases} \qquad (20)$$

Thus (in cooperative limit $N \to \infty$) the functions $\langle \Delta_3^2 \rangle_s / N^2$ and $\langle R_{12} R_{21} \rangle_s / N^2$ have a discontinuous (see Figs. 2 and 3) behaviour analogous to a typical nonequilibrium first-order phase transition[21] at the critical point $Z = 1$. This result is different from the critical behaviour of resonance fluorescence where the system exhibits a sharp transition reminiscent of a typical second-order phase transition[11,12,21].

One can show that the characteristics for spectra of Rayleight line can be obtained using an analogous approach.

IV. Intensity Correlation of Spectrum Components of Scattered Light

Due to Apanasevich and Kilin[17], the normalized intensity correlation functions of the spectrum components of Stokes line are defined as

$$g_{0,0}^{(2)} = \langle \Delta_3^4 \rangle_s / (\langle \Delta_3^2 \rangle_s)^2$$

$$g_{1,1}^{(2)} = \langle R_{12} R_{12} R_{21} R_{21} \rangle_s / (\langle R_{12} R_{21} \rangle_s)^2$$

$$g_{2,2}^{(2)} = \langle R_{31} R_{31} R_{13} R_{13} \rangle_s / (\langle R_{31} R_{13} \rangle_s)^2 .$$

where $g_{0,0}^{(2)}$, $g_{1,1}^{(2)}$ and $g_{2,2}^{(2)}$ are normalized intensity correlation functions for the spectrum line located at frequencies ω_{23}, $\omega_{23} \pm \Omega$ and $\omega_{23} \pm 2\Omega$, respectively.

Using eqs.(4,5), one can find

$$g_{0,0}^{(2)} = \frac{3}{5} \frac{3\langle R^4 \rangle_s + 12 \langle R^3 \rangle_s + 8\langle R^2 \rangle_s - 8\langle R \rangle_s}{(\langle R^2 \rangle_s + 2\langle R \rangle_s)^2} \qquad (21)$$

$$g_{1,1}^{(2)} = \frac{4}{3} \frac{\langle R^4 \rangle_s - 2(N+2)\langle R^3 \rangle_s + (N^2+5N+5)\langle R^2 \rangle_s - (N+1)(N+2)\langle R \rangle_s}{(\langle R \rangle_s (N+1) - \langle R^2 \rangle_s)^2} \qquad (22)$$

$$g_{2,2}^{(2)} = \frac{6}{5} \frac{\langle R^4 \rangle_s + 4\langle R^3 \rangle_s + \langle R^2 \rangle_s - 6\langle R \rangle_s}{(\langle R^2 \rangle_s + 2\langle R \rangle_s)^2} . \qquad (23)$$

Here the values $\langle R^n \rangle_s$ can be found in (6). The detailed behaviour for normalized intensity correlation functions $g_{0,0}^{(2)}$, $g_{1,1}^{(2)}$ and $g_{2,2}^{(2)}$ are shown in figs.4,5 and 6, respectively, where they are plotted as functions of the parameter Z. For the cooperative limit $N \to \infty$ the relations (21-23) reduce to

$$g_{0,0}^{(2)} = \begin{cases} 6 & \text{if} \quad Z \to 1- \\ 12/5 & \quad Z = 1 \\ 9/5 & \quad Z > 1 \end{cases}$$

$$g_{1,1}^{(2)} = \begin{cases} 2 & \text{if} \quad Z < 1 \\ 8/5 & \quad Z = 1 \\ 8/3 & \quad Z > 1 \end{cases}$$

$$g_{2,2}^{(2)} = \begin{cases} 4 & \text{if} \quad Z < 1 \\ 8/5 & \quad Z = 1 \\ 6/5 & \quad Z > 1 \end{cases} .$$

Thus, (in the cooperative limit $N \to \infty$) the normalized intensity correlation functions for spectrum components of scattered light have a discontinuous transition (see figs. 4-6), reminiscent of a typical nonequilibrium first-order phase transition, at the critical point $Z = 1$. Because of the quantum fluctuation, the normalized correlation functions $g_{0,0}^{(2)}$, $g_{1,1}^{(2)}$ and $g_{2,2}^{(2)}$ differ from unit
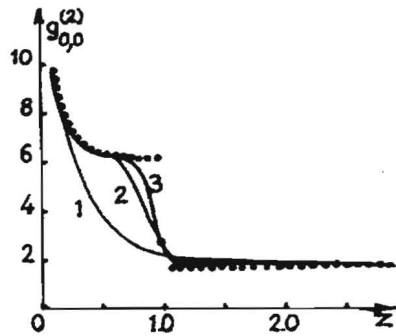
Fig. 4. Normalized intensity correlation function $g_{0,0}^{(2)}$ graphed against the parameter Z. Curves marked 1 to 3 correspond to $N = 5,25,50$ (respectively). The dotted curve indicates the behaviour as $N \to \infty$.

Fig. 5. Normalized intensity correlation function $g_{1,1}^{(2)}$ graphed against the parameter Z. Curves marked 1 to 4, correspond to $N = 5, 25, 50, 100$ (respectively). The dotted curve indicates the behaviour as $N \to \infty$.

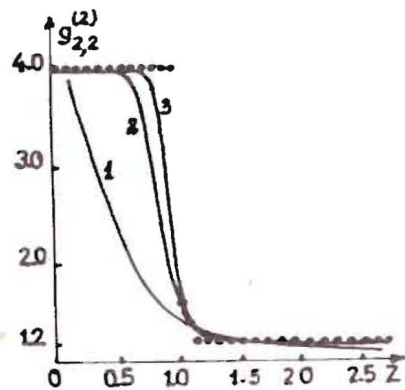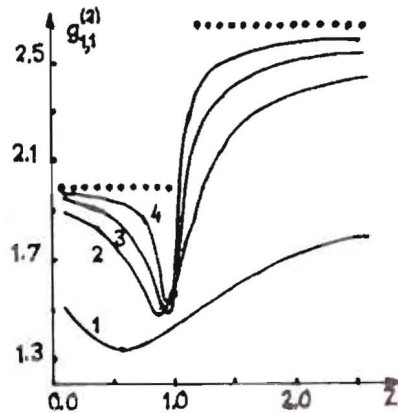Fig.6. Normalized intensity correlation function $g_{2,2}^{(2)}$ graphed against the parameter Z. Curves marked 1 to 3 correspond to $N = 5, 25, 50$, respectively. The dotted curve indicates the behaviour as $N \to \infty$.

not only in the critical region $Z \sim 1$ but for all the values of parameter Z including the case of cooperative limit $N \to \infty$. Consequently, the system cannot be considered classically for the case of large number of atoms.

In conclusion we have shown that in the difference with resonance fluorescence in two-level atoms, the critical point $Z = 1$ in the three-level case is invariant and does not depend on N. However, in the limit $N \to \infty$ it is also necessary to increase Ω so that the condition (2) is satisfied.

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Коллективные эффекты резонансного рассеяния Рамана
в сильных оптических волнах

Исследованы коллективные эффекты резонансного рассеяния
Рамана в присутствии сильных внешнего и рассеянного полей.
Получены аналитические формулы для стационарных спектров сток-
совой линии и нормированных интенсивных корреляционных функ-
ций спектральных компонент.

В коллективном пределе $N \to \infty$ относительные интенсивности
и нормированные интенсивные корреляционные функции имеют пре-
рывное поведение, аналогичное неравновесным фазовым переходам
первого рода.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

Bogolubov N.N.,Jr.,Shumovsky A.S., Tran Quang   E17-85-679
Collective Effects in Resonant Ramann Scattering
of Intense Optical Waves

The collective effects in resonant Ramann scattering
in the presence of intense incident and scattered light
waves are investigated. The analytic expressions for the
steady-state spectrum and normalized intensity correlation
functions for components of the spectra are given. In the
cooperative limit $N \to \infty$ the relative intensities and nor-
malized intensity correlation functions for each component
of the spectra have a discontinuous behaviour reminiscent
of a typical nonequilibrium first order phase transition.

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