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NONCLASSICAL EFFECTS IN COLLECTIVE DOUBLE OPTICAL RESONANCE

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I. INTRODUCTION

In many nonlinear systems involving the interaction between light and medium, some nonclassical effects are observed. The reviews of such nonclassical effects in optics are given in papers by Loudon $^{/1/}$. Paul $^{/2/}$, and Walls $^{/3'}$. The most well-known example of these effects is the photon antibunching which was observed in the experimental works by Kimble et al $^{4,5/}$.

Another example of nonclassical effects is the violation of the Cauchy -Schwarz (C-S) inequality which was observed in the work by Clauser $^{6/}$. The violation of the C-S inequality was also predicted in the two-photon laser $^{7/}$ and parametric amplifier $\frac{78}{8}$.

In recent years a large number of theoretical and experimental works is concentrated on the problem of squeezed states of light /9-18, 31-32/ which were observed in the experimental works by Slusher et al. $^{/20/}$. Shelby et al. $\frac{21}{2}$, and Kimble et al. $\frac{22}{2}$.

In this paper we present the violation of the C-S inequality and the squeezing in a fluorescence from a system of three-level atoms (Fig. 1) interaction with two driving monochromatic resonant fields and with an emitted field in the context of double optical resonance $\frac{23\cdot25}{}$.

II. MASTER EQUATION

The N three-level atoms are assumed to be concentrated in a region small compared to the wavelength of all the relevant radiation modes. In treating the external field as C-numbers, the master equation for the atomic system alone ρ with the Markovian and rotating wave approximation is $\frac{27}{}$.



Fig. 1. Three-level system of atoms interacting with the two monochromatic applied fields and with emitted field.

$$\frac{\partial \rho}{\partial t} = -iG[(\cos \alpha J_{12} + \sin \alpha J_{23} + H.C.), \rho] - - \gamma_{21} (J_{21} J_{12} \rho - 2J_{12} \rho J_{21} + \rho J_{21} J_{12}) - - \gamma_{32} (J_{32} J_{23} \rho - 2J_{23} \rho J_{32} + \rho J_{32} J_{23}) = L\rho,$$
(1)

where $2\gamma_{21}$ and $2\gamma_{32}$ are radiative spontaneous transition probabilities per unit time for a single atom to change from the level $|2\rangle$ to $|1\rangle$ and from $|3\rangle$ to $|2\rangle$, respectively; $G = (G_1^2 + G_2^2)^{\frac{1}{2}}$ and $tga = G_2/G_1$, where G_1 and G_2 are the Rabi frequencies for the atomic transitions from level $|2\rangle$ to $|1\rangle$ and from $|3\rangle$ to $|2\rangle$, respectively; and

$$J_{ij} = \sum_{k=1}^{N} |i\rangle_{k} \leq j | \qquad (i, j = 1, 2, 3)$$

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

$$[\mathbf{J}_{ij} \ , \ \mathbf{J}_{i'j'}] = \mathbf{J}_{ij'} \ \delta_{i'j} \ - \mathbf{J}_{i'j} \ \delta_{ij'} \ .$$

As in refs. $^{19,25,28/}$, we introduce the Schwinger representation for angular momentum

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

where operators C, obey the boson commutation relation

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

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

$$\mathbf{G} \gg \mathbf{N} \boldsymbol{\gamma}_{21} , \mathbf{N} \boldsymbol{\gamma}_{32} . \tag{2}$$

After performing the canonical transformation

$$C_{3} = -\frac{1}{\sqrt{2}} \sin a Q_{1} + \cos a Q_{2} + \frac{1}{\sqrt{2}} \sin a Q_{3} ,$$

$$C_{2} = \frac{1}{\sqrt{2}} Q_{1} + \frac{1}{\sqrt{2}} Q_{3} ,$$
(3)

$$C_{1} = -\frac{1}{\sqrt{2}}\cos a\hat{\omega}_{1} - \sin a\hat{\omega}_{2} + \frac{1}{\sqrt{2}}\cos a\hat{\omega}_{3} , \qquad (3)$$

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 nG(n = 1,2,3,4). For the case when relation (2) is fulfilled, we make the secular approximation, i.e., retain only a slowly varying part $^{(25,30)}$. Correction of the results obtained in this fashion will be of an order of $(\gamma_{21}N/G)^2$ or $(\gamma_{32}N/G)^2$.

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

$$\widetilde{\rho} = Z \frac{1}{R} \sum_{R=0}^{N} X \frac{R}{M} \sum_{R=0}^{R} |\mathbf{M}, \mathbf{R}\rangle \langle \mathbf{R}, \mathbf{M}| , \qquad (4)$$

where $\tilde{\rho} = U\rho U^{\dagger}$, here U is the unitary operator representing the canonical transformation (3)

X =
$$\frac{\gamma_{32}\cos^2 a}{\gamma_{21}\sin^2 a}$$
; Z = $\frac{(N+1)X^{N+2} - (N+2)X^{N+1} + 1}{(X-1)^2}$.

The state $|M, R\rangle$ is an eigenstate of the operator R_{11} , $R = R_{11} + R_{33}$ 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

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

so

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

By using solution (4) the characteristic function can be defined similarly to Louisell $^{/29/}$

$$\chi_{R}(\xi) = \langle \mathbf{e}^{i\xi R} \rangle_{\mathbf{s}} = \mathbf{Z}^{-1} \frac{(N+1)\mathbf{Y}^{N+2} - (N+2)\mathbf{Y}^{N+1} + 1}{(\mathbf{Y}-1)^{2}}, \quad (7)$$

2

3

$$\frac{\partial \rho}{\partial t} = -iG[(\cos \alpha J_{12} + \sin \alpha J_{23} + H.C.), \rho] - - \gamma_{21} (J_{21} J_{12} \rho - 2J_{12} \rho J_{21} + \rho J_{21} J_{12}) - - \gamma_{32} (J_{32} J_{23} \rho - 2J_{23} \rho J_{32} + \rho J_{32} J_{23}) = L\rho,$$
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where $2\gamma_{21}$ and $2\gamma_{32}$ are radiative spontaneous transition probabilities per unit time for a single atom to change from the level $|2\rangle$ to $|1\rangle$ and from $|3\rangle$ to $|2\rangle$, respectively; $G = (G_1^2 + G_2^2)^{\frac{1}{2}}$ and $tga = G_2/G_1$, where G_1 and G_2 are the Rabi frequencies for the atomic transitions from level $|2\rangle$ to $|1\rangle$ and from $|3\rangle$ to $|2\rangle$, respectively; and

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$$\widetilde{\rho} = Z \frac{-1}{R} \sum_{R=0}^{N} X \frac{R}{M} \sum_{R=0}^{R} |M, R\rangle \langle R, M| , \qquad (4)$$

where $\tilde{\rho} = U\rho U^{\dagger}$, here U is the unitary operator representing the canonical transformation (3)

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2

3

where $Y = Xe^{i\xi}$ and $\langle B \rangle_s$ indicates the expectation value of an operator B in the steady state (3).

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

$$\langle \mathbf{R}^{\mathbf{n}} \rangle_{\mathbf{s}} = \frac{\partial^{\mathbf{n}}}{\partial (\mathbf{i}\xi)^{\mathbf{n}}} \chi_{\mathbf{R}}(\xi) |_{\mathbf{i}\xi = 0}$$
 (8)

III. SQUEEZING IN THE FLUORESCENT LIGHT

In this section we discuss squeezing in the fluorescent light in the collective resonant process.

The variance of the fluctuations in the fluorescent field may be derived by using the following relations between the radiation field and the atomic operator in the far-field limit $^{/16/}$

$$E_{1}^{(+)}(\vec{x}, t') = E_{1, \text{ free}}^{(+)}(\vec{x}, t') + \psi_{12}(\vec{x}) J_{12}(t) e^{-i\Omega_{1}t} ,$$

$$E_{1}^{(-)}(\vec{x}, t') = (E_{1}^{(+)}(\vec{x}, t'))^{+} ,$$
(9)

$$E_{2}^{(+)}(\vec{x}, t') = E_{2, \text{free}}^{(+)}(\vec{x}, t') + \psi_{23}(\vec{x}) J_{23}(t) e^{-i\Omega_{2}t} ,$$

$$E_{2}^{(-)}(\vec{x}, t') = (E_{2}^{(+)}(\vec{x}, t'))^{+} ,$$
(10)

where t = t' - t/C,

$$\psi_{12}(\vec{x}) = \frac{\omega_{21}}{2\pi\epsilon_0 C^2} \cdot \frac{\vec{x} \times (\vec{d}_{12} \times \vec{x})}{r^3},$$

$$\psi_{23}(\vec{x}) = \frac{\omega_{32}}{2\pi\epsilon_0 C^2} \cdot \frac{\vec{x} \times (\vec{d}_{23} \times \vec{x})}{r^3},$$

the values of ψ_{12} and ψ_{23} are assumed to be real, \vec{d} and \vec{x} are the dipole vector of the atoms and the observation point vector, respectively; $\mathbf{r} = |\vec{x}|$; $\mathbf{E}_{1}^{(+)}$ and $\mathbf{E}_{2}^{(+)}$ are the positive-frequency parts of the fluorescent fields corresponding to the lower and upper atomic transitions $|2\rangle$ to $|1\rangle$ and $|3\rangle$ to $|2\rangle$, respectively.

From the canonical transformation (3), one can write the collective angular moments J_{12} and J_{23} in the form

$$J_{12} = \frac{1}{2} \cos \alpha D_3 + \frac{1}{2} \cos \alpha (R_{31} - R_{13}) - \frac{1}{\sqrt{2}} \sin \alpha (R_{23} + R_{21}), \qquad (11)$$

$$J_{23} = \frac{1}{2} \sin a D_3 + \frac{1}{2} \sin a (R_{31} - R_{13}) + \frac{1}{\sqrt{2}} \cos a (R_{12} + R_{32}), \quad (12)$$

where $D_3 = R_{33} - R_{11}$.

Following the works 29,16 we can consider the operators $-\frac{1}{2}\cos \alpha R_{13}(t)$, $-\frac{1}{\sqrt{2}}\sin \alpha R_{23}(t)$, $\frac{1}{2}\cos \alpha \cdot D_3(t)$, $-\frac{1}{\sqrt{2}}\sin \alpha R_{21}(t)$ and $\frac{1}{2}\cos \alpha R_{31}(t)$ as the amplitude-operators for the source of the spectrum components at the frequencies $\Omega_1^{-2}G$, $\Omega_1^{-}G$, Ω_1 , $\Omega_1^{+}G$ and $\Omega_1^{+2}G$: and for simplicity we denote these operators by S_{-2}^{-} , S_{-1}^{-} , S_0^{-} , S_1^{-} and S_2^{-} , respectively.

Analogously, the operators $-\frac{1}{2}\sin\alpha R_{13}(t)$, $\frac{1}{\sqrt{2}}\cos\alpha R_{12}(t)$,

 $\frac{1}{2}\sin a D_3(t)$, $\frac{1}{\sqrt{2}}\cos a R_3(t)$ and $\frac{1}{2}\sin a R_{31}(t)$ can be considered as the amplitude operators for the sources of the spectrum components at the frequencies Ω_2 -2G, Ω_2 -G, Ω_2 , Ω_2 +G and Ω_2 +2G and for simplicity we denote these operators by T_{-2} , T_{-1} , T_0 , T_1 and T_2 , respectively.

The following calculations show that the squeezing is absent for all separate spectrum components S_1 and T_1 $(i = 0, \pm 1, \pm 2)$. The squeezing is also absent for the whole fluorescent fields corresponding to the lower and upper atomic transitions. One can find that the squeezing exists only in the mixtures of the spectrum components S_1 and T_{-1} or S_{-1} and T_1 ; moreover, the degrees of squeezing in the mixture of S_1 and T_{-1} and in the mixture of S_{-1} and T_1 are equal. Further, we discuss only the squeezing in the mixture of the two spectrum components S_{-1} and T_{+1} . After substituting the operator J_{12} in the relation (9) by the operator S_{-1} and the operator J_{23} in relation (10) by the operator T_{+1} and using the steadystate solution (4) one finds the normally ordered variance of fluctuations for the hermitian amplitude operators of the mixture of two spectrum components S_{-1} and T_1 in the form

$$<: (\Delta a_{1,2})^{2} :> = \frac{1}{4} \{ \psi_{12}^{2} \sin^{2} \alpha < R_{32} R_{23} >_{s} + \psi_{23}^{2} \cos^{2} \alpha < R_{23} R_{32} >_{s} \mp (13)$$

$$\mp \psi_{12} \psi_{23} \cos \alpha \cdot \sin \alpha (< R_{32} R_{23} >_{s} + < R_{23} R_{32} >_{s}),$$

where $a = E_1^{(+)} + E_2^{(+)}$, $a^+ = E_1^{(-)} + E_2^{(-)}$, $a_1 = \frac{1}{2}(a^++a)$, $a_2 = \frac{-i}{2}(a-a^+)$. The statistical moments $\langle R_{32}R_{23} \rangle_s$ and $\langle R_{23}R_{32} \rangle_s$ can be written in the form

$$<\mathbf{R}_{23} \mathbf{R}_{32}>_{s} = \frac{1}{2}(N-2) < \mathbf{R}>_{s} - \frac{<\mathbf{R}^{2}>_{s}}{2} + N,$$
 (14)

$$<\mathbf{R}_{32} | \mathbf{R}_{23} >_{\mathbf{s}} = \frac{1}{2} (\mathbf{N} + 1) < \mathbf{R} >_{\mathbf{s}} - \frac{1}{2} < \mathbf{R}^2 >_{\mathbf{s}},$$
 (15)

here $\langle R \rangle_s$ and $\langle R^2 \rangle_s$ can be found in relation (8). We speak of squeezing in the mixture of two spectrum components S_{-1} and T_{+1} if the normally ordered variance of the operators a_1 or a_2 is less than zero /16-18/

$$<: (\Delta a_i)^2 :> < 0 \qquad i = 1 \text{ or } 2.$$
 (16)

It is easy to see that in the case of x = 1, we have $\langle R_{23} R_{32} \rangle_s = \langle R_{32} R_{23} \rangle_s$ and the relation (13) reduces to

$$<: (\Delta a_{1,2})^2 :> = \frac{1}{4} < R_{23} R_{32} >_{s} \times (\psi_{12} \sin a - \mp \psi_{23} \cos a)^2 \ge 0$$
 (17)

thus, the squeezing is adsent for this case. It is easy to see that the squeezing is also absent for the case of $\operatorname{ctg} a \to 0$ or $\operatorname{ctg} a \to \infty$.

The detailed behaviour of the $\langle:(\Delta a_1)^2:\rangle$ (in the relative unity $\frac{1}{4}\psi_{12}^2$) as a function of the relation of the pumping field intensities $\operatorname{ctg} 2_a$ in the case of $\gamma_{32} = \gamma_{21}$ and $\psi_{12} = \psi_{23} > 0$ is plotted in fig. 2 for various numbers of atoms. As is shown in fig. 2, the substantial squeezing occurs for the mixture of two spectrum components S_{-1} and T_1 (or S_1 and T_{-1}). The squeezing is absent for all separate spectrum components or for the whole field of fluorescence. As in the work $^{/17/}$, we can define the factor of squeezing for the atomic operators

$$A_{1} = \frac{1}{2}(A + A^{+}), \quad A_{2} = -\frac{i}{2}(A - A^{+}),$$

where

$$A = \psi_{12} S_{-1} + \psi_{23} T_{1},$$

in the form

$$F_{1,2} = \frac{\langle (\Delta A_{1,2})^2 : \rangle_s}{\frac{1}{2} |\langle [A_1, A_2] \rangle_s|}.$$
 (18)

The behaviour of the factor of squeezing F_1 as a function of $\operatorname{ctg}^2 a$ in the case of $\gamma_{32} = \gamma_{21}$, $\psi_{12} = \psi_{23} > 0$ is plotted in fig. 3. As is shown in fig. 3 the factors of squeezing for atomic operators A_1 , A_2 are independent of the number of atoms and can tend to the value $F_1 = -0.5$ (i.e., the 50% of squ-



Fig. 2 (a-b) The normally ordered variance $\langle : (\Delta a_1)^2 : \rangle$ as the function of $\operatorname{ctg}^2 a$ for the case of $\gamma_{32} = \gamma_{21}$ and $\psi_{12} = \psi_{23} > 0$.



eezing) in a region of the point $\operatorname{ctg}^2 a = 1$. One can show that $[A_1, A_2] = 0$ when $\operatorname{ctg}^2 a = 1$. Thus in this case, however $F_1 = -0.05$, the conception of squeezing for commutation operators A_1 and A_2 loses sense.

We note that as a result of the influence of the free parts of the fluorescent field $E_{1, \text{ free}}^{(\pm)}$ and $E_{2, \text{ free}}^{(\pm)}$ (see relation 9-10) the factors of squeezing F_1 , F_2 for the atomic operators A_1 , A_2 are not coincident with the factors of squeezing for the field operators a_1 and a_2 .

IV. VIOLATION OF THE CAUCHY - SCHWARZ (C-S) INEQUALITY

As Zubairy 77 , we define a degree of second-order coherence between the spectrum components S_i and S_i in the form

$$G_{i,j}^{(2)} = \frac{\langle S_{i}^{+} S_{j}^{+} S_{j} S_{i} \rangle_{s}}{\langle S_{i}^{+} S_{i} \rangle_{s} \cdot \langle S_{j}^{+} S_{j} \rangle_{s}} \qquad (i,j = 0, \pm 1, \pm 2).$$
(19)

The photon antibunching is exhibited for the spectrum component S_i (i = 0, ± 1, ± 2) which satisfies the inequality

$$G_{i,i}^{(2)}$$
 < 1,

i.e., the degree of second-order coherence is less than unity. Such properties occurring for the four sidebands S_i (i = ±1,±2) in the case of one or several atoms are investigated in the work $^{/19/}$.

Further, we shall discuss another nonclassical effect — the violation of the C-S inequality in the stationary fluorescent field.

We speak about the violation of the C-S inequality for the correlation between two spectrum components S_i and S_j $(i, j = 0, \pm 1, \pm 2)$ if the following condition is satisfied $^{/33/}$:

$$K_{i,j} = \frac{G_{i,i}^{(2)} \cdot G_{j,j}^{(2)}}{(G_{i,j}^{(2)})^2} < 1.$$
(20)

The factor $K_{i,j}$ descrides the degree of violation of the C-S inequality for the correlation between two spectrum components S_i and S_j .

By using commutations (5-6) and stationary solution (4), one can find:

$$K_{2,-2} = K_{-2,2} = \frac{(\langle R_{31}R_{31}R_{13}R_{13}R_{13}\rangle_{s})^{2}}{(\langle R_{31}R_{13}R_{13}R_{31}R_{13}\rangle_{s})^{2}}, \qquad (21)$$

$$K_{0,2} = K_{2,0} := K_{0,-2} = K_{-2,0} = \frac{\langle R_{31}R_{31}R_{13}R_{13} \rangle_{s} \cdot \langle D_{3}^{*} \rangle_{s}}{(\langle D_{3}R_{31}R_{13}R_{13}R_{13}D_{s} \rangle_{s})^{2}},$$
(22)

$$K_{0,1} = K_{0,-1} = \frac{\langle R_{32} R_{32} R_{23} R_{23} R_{23} \rangle_{s} \cdot \langle D_{3}^{4} \rangle_{s}}{(\langle D_{3} R_{32} R_{23} D_{3} \rangle_{s})^{2}}, \qquad (23)$$

$$K_{2,-1} = K_{-2,1} = \frac{\langle R_{31}R_{31}R_{13}R_{13}R_{13}\rangle_{s} \cdot \langle R_{12}R_{12}R_{21}R_{21}R_{21}\rangle_{s}}{(\langle R_{31}R_{12}R_{21}R_{13}\rangle_{s})^{2}}, \quad (24)$$

$$K_{-1,2} = K_{1,-2} = \frac{\langle R_{31}R_{31}R_{13}R_{13}\rangle_{s} \cdot \langle R_{12}R_{12}R_{21}R_{21}\rangle_{s}}{(\langle R_{12}R_{31}R_{13}R_{21}\rangle_{s})^{2}}, \quad (25)$$

where

$$\langle D_3^4 \rangle_s = \frac{1}{15} (3 \langle R^4 \rangle_s + 12 \langle R^3 \rangle_s + 8 \langle R^2 \rangle_s - 8 \langle R \rangle_s),$$
 (26)

$$< R_{12} R_{12} R_{21} R_{21} >_{s} = \frac{1}{3} (< R^{4} >_{s} - 2(N+2) < R^{3} >_{s} + (N^{2} + 5N + 5) < R^{2} >_{s} - (N+1)(N+2) < R >_{s}),$$

$$(27)$$

$$< R_{31}R_{31}R_{13}R_{13} = \frac{1}{30}(< R^4 > + 4 < R^3 > + < R^2 > - 6 < R >),$$
 (28)

$$< R_{31} R_{13} R_{31} R_{13} >_{s} = \frac{1}{12} (-< R^{4} >_{s} + N < R^{3} >_{s} + (N + 3) < R^{2} >_{s} - 2(N + 1) < R >_{s}),$$

$$(29)$$

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$$< D_{3}R_{31}R_{13}D_{3}>_{s} = \frac{1}{30}(< R^{4}>_{s} + 4< R^{3}>_{s} + 6< R^{2}>_{s} + 4< R>_{s}),$$
 (30)

$$\langle D_3 R_{32} R_{23} D_3 \rangle_s = \frac{1}{6} (-\langle R^4 \rangle_s + (N-1) \langle R^3 \rangle_s + 2(N+1) \langle R^2 \rangle_s),$$
 (31)

$$_{s} = \frac{1}{12}(-_{s} + (N+2)_{s} + (32)$$

$$+ (3N + 1) < R^2 > + 2(N + 1) < R >$$

$$_{s} = \frac{1}{12}(-_{s} + N_{s} + (N+3)_{s}).$$
 (33)

The following calculations show that another factors are more than unity for any number of atoms

 $K_{1,-1} = K_{-1,1} > 1,$ $K_{2,1} = K_{1,2} = K_{-1,-2} = K_{-2,-1} > 1,$ $K_{1,0} = K_{-1,0} > 1.$

It means that the classical C-S inequality occurs in these cases.

The behaviour of the functions $K_{2,-2}$, $K_{0,2}$, $K_{0,1}$, $K_{2,-1}$ and $K_{-1,2}$ against the parameter x is shown in figs. 4 (a-e) for various numbers of atoms. It is easy to see from these figures that the strong violation of the C-S inequality exists for a large number of atoms. It means that in contrast with the effect of photon antibunching, the violation of the C-S inequality is the macroscopic quantum effect.

To conclude, we note that the investigation of the violation of the C-S inequality for the correlations between spectrum components corresponding to the upper atomic transition $|3\rangle \rightarrow |2\rangle$ and between spectra of the upper and lower atomic transitions can be carried out by using an analogous approach.

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Неклассические эффекты

в коллективном двойном оптическом резонансе

Обсуждены сжатие и нарушение неравенства Коши — Шварца в поле флуоресценции коллективного двойного оптического резонансного процесса.

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The squeezing and violation of the Cauchy - Schwarz inequality in the fluorescent field of collective double resonant process are discussed.

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

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