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# VIOLATION OF THE CAUCHY - SCHWARZ INEQUALITY IN COLLECTIVE RAMAN SCATTERING

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In recent years, a large number of works has been concentrated on the nonclassical properties of the light. The most interesting nonclassical effects such as photon antibunching[1-2], violation of the Cauchy - Schwarz (C-S) inequality[3] and squeezing[4-5] have been observed experimentally. The applications of the quantum light in the precision measurements, information transfer and detection of a gravitational wave are widely discussed[6-9].

The violation of the Cauchy - Schwarz inequality has been observed by Clausser[3] in the radiation emitted in an atomic two-photon cascade. This effect has also been predicted in the two-photon laser[10], parametric amplifier[11], collective resonance fluorescence[12] collective double resonance[13-14], and harmonic generation[15].

In this paper we discuss the violation of the Cauchy - Schwarz inequality in the correlations between the spectrum components of the Rayleigh and Stoke lines. It has been shown that the violation of the C-S inequality is present only in the Rayleigh line, moreover for the correlations between the sidebands of the Rayleigh line the violation of the C-S inequality is present for a large number of atoms. It means that this quantum effect has the macroscopic nature. Analogous conclusions have been made in recent works[12-14].

We consider a small system (the Dicke model) of N three level atoms interacting with one mode of a monochromatic driving field of frequency $\omega_L$ and with the vacuum of other modes. A schematic diagram of atomic energy levels is shown in fig.1. The ground state $|1\rangle$ is coupled to the excited state $|2\rangle$ by the strong driving field. The state $|3\rangle$ may be a low-lying vibrational or rotational excitation accessible from the ground state. To keep the discussion general, we will not specify those states only 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. The transition $|3\rangle \to |1\rangle$ is caused by an atomic reservoir and assumed to be nonradiative[16].

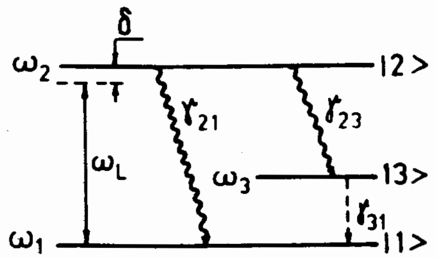


*Fig.1. Level scheme of the atomic system.*



On treating the exciting laser field classically and making standard (Born and Markov) approximations to describe the system reservoir couplings, one obtains a master equation for the reduced density operator of the atomic system alone in the following form[17] ($\hbar \equiv 1$ units are used)

$$\frac{\partial\rho}{\partial t} = -i\left[\frac{\delta}{2}(J_{22}-J_{11}) + G(J_{12}+J_{21}) - \Omega_3 J_{33}, \rho\right] - \gamma_{21}(J_{21}J_{12}\rho - J_{12}\rho J_{21} + H.C.) - \gamma_{23}(J_{23}J_{32}\rho - J_{32}\rho J_{23} + H.C.) - \gamma_{31}(J_{31}J_{13}\rho - J_{13}\rho J_{31} + H.C.), \qquad (1)$$

where $2\gamma_{ij}$ $(i,j = 1,2,3)$ are the transition rate from level $|i\rangle \to |j\rangle$ due to the atomic interaction with the reservoir; $\Omega_3 = \omega_{23} - \frac{\omega_{21}}{2}$ (where $\omega_{ij} = \omega_i - \omega_j$), $\delta = \omega_{21} - \omega_L$ is the detuning of the laser frequency from the atomic resonance frequency $\omega_{21}$ ; $G = -d_{21}E_0$ is the resonance Rabi frequency describing the interaction of the driving field with the atomic system, $J_{ij}$ $(i,j = 1,2,3)$ are the collective angular momenta of the atomic system having in the Schwinger representation the following form

$$J_{ij} = C_i^+ C_j \, , \qquad (2)$$

where the operators $C_i$ and $C_i^+$ obey the boson communication relation

$$[C_i, C_j^+] = \delta_{ij} \, , \qquad (3)$$

and can be treated as the annihilation and creation operators for the atoms being populated in the level $|i\rangle$.

Futher, we investigate the case of an intense external field or of large detuning $\delta$ so that the following relation is fulfilled:

$$\Omega = \left(\frac{1}{4}\delta^2 + G^2\right)^{1/2} \gg N\gamma_{ij} \quad (i,j = 1-3). \qquad (4)$$

After the cannonical transformation

$$C_1 = Q_1 \cos\phi + Q_2 \sin\phi ,$$

$$C_2 = -Q_1 \sin\phi + Q_2 \cos\phi , \qquad (5)$$

$$C_3 = Q_3 ,$$

where

$$\mathrm{tg}\, 2\phi = 2G/\delta ,$$

and after performing the secular approximation[18], i.e. ignoring the part of the Liouville operator L containing rapidly oscillating terms with frequencies $n\Omega (n = 2.4)$, one can find a stationary solution of the master equation in the form

$$\tilde{\rho} = U\rho U^+ = A^{-1} \sum_{P=0}^{N} X^P \sum_{M=0}^{P} Z^M |F, M\rangle\langle M, P| , \qquad (6)$$

where U is the unitary operator representing the cannonical transformation (5)

$$X = \gamma_{31}/(\gamma_{23}\, \mathrm{ctg}^2\phi ), \qquad (7)$$

$$Z = \mathrm{ctg}^4\phi , \qquad (8)$$

$$A = \frac{Z}{Z-1} \cdot \frac{(XZ)^{N+1} - 1}{XZ - 1} - \frac{1}{Z-1} \cdot \frac{X^{N+1} - 1}{X - 1} , \qquad (9)$$

$|F, M\rangle$ is an eigenstate of the operators $R = R_{11} + R_{22}$, $R_{11}$ and the operator of number of atoms $\hat{N} = R_{11} + R_{22} + R_{33}$ here $R_{k\ell} = Q_k^+ Q_\ell \quad (k, \ell = 1,2,3)$ are the collective operators of "dressed" atoms. The operators $Q_k, Q_k^+$ satisfy the boson commutation relation

$$[Q_k, Q_\ell^+] = \delta_{k\ell} , \qquad (10)$$

and consequently,

$$[R_{k\ell}, R_{k'\ell'}] = R_{k\ell'}\delta_{k'\ell} - R_{k'\ell}\delta_{k\ell'} . \qquad (11)$$

By using eq.(6) the characteristic function can be defined

$$\chi_{R_{11},R}(\eta, \xi) = \langle \ell^{i\eta R_{11} + i\xi R} \rangle =$$

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

where $Y_1 = X\ell^{i\xi}$, $Y_2 = Z\ell^{i\eta}$ and $\langle B\rangle$ denotes the expectation value of the operator B over the steady state described by the density matrix (6). Once the characteristic function is known, it is easy to calculate the statistical moments

$$\langle R^n R_{11}^m \rangle = \frac{\partial^n}{\partial (i\xi)^n} \cdot \frac{\partial^m}{\partial (i\eta)^m} \chi_{R_{11},R}(\eta, \xi)\Big|_{\substack{i\eta = 0 \\ i\xi = 0}} . \qquad (12)$$

Further we discuss the violation of the C-S inequality in the correlations between spectrum components of the Rayleigh line due to atomic transition $|2\rangle \to |1\rangle$.

With the use of the dressing transformation (5) the atomic collective operator $J_{21}$ can be written in the form

$$J_{21} = \sin\phi \cdot \cos\phi\, (R - 2R_{11}) + \cos^2\phi R_{21} - \sin^2\phi R_{12} . \qquad (13)$$

Following the work[18-19] we can consider the operators $\cos^2\phi R_{21}$, $\sin\phi\cos\phi(R - 2R_{11})$ and $-\sin^2\phi R_{12}$ – as the amplitude operators for the sources of Mollow's triplet of the Rayleigh line contered at the frequencies $\omega_L + 2\Omega$, $\omega_L$ and $\omega_L - 2\Omega$ and for simplicity we denote these operators by $S_1^+$, $S_0$ and $S_{-1}^+$, respectively.

Following London[20], we define a nondelayed second-order correlation function for the spectrum components $S_i$ and $S_j$ in the form

$$G_{i,j}^{(2)} = \frac{\langle S_i^+ S_j^+ S_j S_i \rangle}{\langle S_i^+ S_i \rangle \cdot \langle S_j^+ S_j \rangle} \qquad (i, j = 0, \pm 1) . \qquad (14)$$

Since the operator $S_i$ does not commute with the operator $S_j$, in the general case we have

$$G_{i,j}^{(2)} \neq G_{j,i}^{(2)} \qquad (i \neq j) .$$

The quantities $G_{i,i}^{(2)}$ $(i = 0, \pm 1)$ describe the photon statistics of the spectrum components $S_i$ and the quantities $G_{i,j}^{(2)}$ $(i \neq j)$ describe the cross-correlation between spectrum components $S_i$ and $S_j$.

The validity of the Cauchy - Schwarz (C-S) inequality is presented by the classical theory for the nondelayed second-order correlation functions[20]

$$(G^{(2)}_{i,j})^2 < G^{(2)}_{i,i} \cdot G^{(2)}_{j,j} \quad (i \neq j). \tag{15}$$

Further we discuss the conditions when the C-S inequality (15) for the correlation between spectrum components $S_i$ and $S_j$ $(i \neq j,\ i,j = 0,\pm 1)$ is violated.

We introduce the factor (if $G^{(2)}_{i,j} \neq 0$)

$$K_{i,j} = (G^{(2)}_{i,i} \cdot G^{(2)}_{j,j})/(G^{(2)}_{i,j})^2. \tag{16}$$

The Cauchy - Schwarz inequality is violated for the correlation between spectrum components $S_i$ and $S_j$ if the factor $K_{ij}$ is less than unity, thus the factor $K_{ij}$ describes the degree of the violation of the C-S inequality.

For the single-atom case, using the well-known operator relation

$$R_{ij}\, R_{i'j'} = R_{ij'}\, \delta_{i'j}, \tag{17}$$

one shows that

$$G^{(2)}_{1,1} = G^{(2)}_{-1,-1} = 0, \tag{18}$$

$$G^{(2)}_{0,0} = G^{(2)}_{0,\pm 1} = G^{(2)}_{+1,0} = \frac{XZ + X + 1}{XZ + X} > 1, \tag{19}$$

$$G^{(2)}_{-1,1} = \frac{XZ + X + 1}{X} > 1, \tag{20}$$

$$G^{(2)}_{1,-1} = \frac{XZ + X + 1}{XZ} > 1. \tag{21}$$

Equations (18)-(21) yield $K_{i,j} = 0$ for $i \neq j$, $i,j = 0,\pm 1$ and it means that in the single-atom case the C-S inequality is violated for any two spectrum components of the triplet of the Rayleigh line.

Further, we discuss the violation of the C-S inequality in the collective case. The following calculations show that in the collective case the C-S inequality is violated only for the correlations between the two sidebands $S_{\pm 1}$. By using the solution (6) and commutation relations (10)-(11) one can find

$$K_{1,-1} = \frac{\langle R_{21}R_{21}R_{12}R_{12}\rangle \cdot \langle R_{12}R_{12}R_{21}R_{21}\rangle}{(\langle R_{21}R_{12}R_{21}R_{12}\rangle)^2}, \tag{22}$$



$$K_{-1,1} = \frac{\langle R_{21}R_{21}R_{12}R_{12}\rangle \cdot \langle R_{12}R_{12}R_{21}R_{21}\rangle}{(\langle R_{12}R_{21}R_{12}R_{21}\rangle)^2}, \tag{23}$$

where

$$\begin{aligned}\langle R_{21}R_{12}R_{21}R_{12}\rangle = {} & \langle R^4_{11}\rangle - 2\langle RR^3_{11}\rangle + 2\langle R^3_{11}\rangle + \\ & + \langle R^2R^2_{11}\rangle - 4\langle RR^2_{11}\rangle + \langle R^2_{11}\rangle + \\ & + 2\langle R^2R_{11}\rangle - 2\langle RR_{11}\rangle + \langle R^2\rangle,\end{aligned} \tag{24}$$

$$\begin{aligned}\langle R_{12}R_{21}R_{12}R_{21}\rangle = {} & \langle R^4_{11}\rangle - 2\langle RR^3_{11}\rangle - 2\langle R^3_{11}\rangle + \\ & + \langle R^2R^2_{11}\rangle + 2\langle RR^2_{11}\rangle + \langle R^2_{11}\rangle,\end{aligned} \tag{25}$$

$$\begin{aligned}\langle R_{12}R_{12}R_{21}R_{21}\rangle = {} & \langle R^4_{11}\rangle - 2\langle RR^3_{11}\rangle - 4\langle R^3_{11}\rangle + \\ & + \langle R^2R^2_{11}\rangle + 5\langle RR^2_{11}\rangle + 5\langle R^2_{11}\rangle - \\ & - \langle R^2R_{11}\rangle - 3\langle RR_{11}\rangle - 2\langle R_{11}\rangle,\end{aligned} \tag{26}$$

$$\begin{aligned}\langle R_{21}R_{21}R_{12}R_{12}\rangle = {} & \langle R^4_{11}\rangle - 2\langle RR^3_{11}\rangle + 4\langle R^3_{11}\rangle + \\ & + \langle R^2R^2_{11}\rangle - 7\langle RR^2_{11}\rangle + 5\langle R^2_{11}\rangle + 3\langle R^2R_{11}\rangle - \\ & - 7\langle RR_{11}\rangle + 2\langle R_{11}\rangle + 2\langle R^2\rangle - 2\langle R\ \rangle.\end{aligned} \tag{27}$$

In eqs.(24)-(27) the expectation values $\langle R^n R^m_{11}\rangle$ can be found according to the relation (12).

The behaviour of the factors $K_{1,-1}$ and $K_{-1,1}$ as functions of the parameter $ctg^2\phi$ is plotted in figs.2 and 3 for several values of N and $\gamma_{23}/\gamma_{31}$. It is easy to see from figures 2-3 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 a macroscopic quantum effect.

Finally we discuss the violation of the C-S inequality in the stoke line. One can find from the cannonical transformation (5) that

$$J_{23} = -\sin\phi\, R_{13} + \cos\phi\, R_{23}. \tag{28}$$

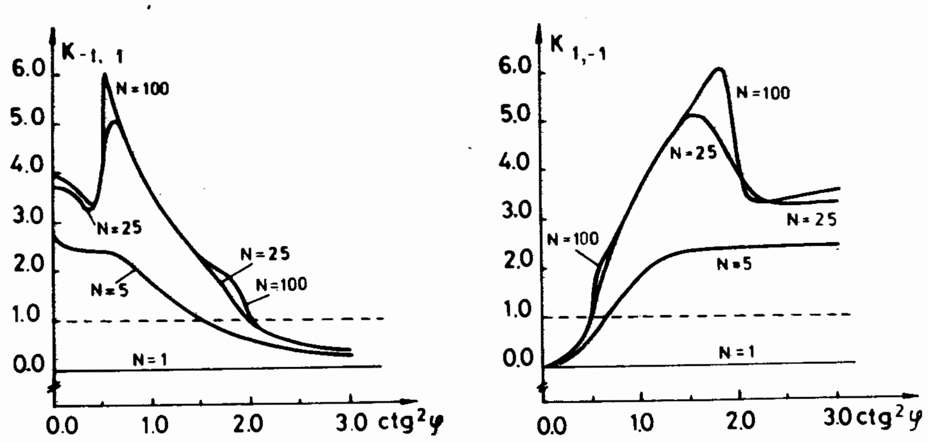


*Fig. 2. The factors* $K_{-1,1}$ *and* $K_{1,-1}$ *as functions of* $ctg^2\phi$ *for* $\gamma_{31}/\gamma_{23}$ *= 0.5.*

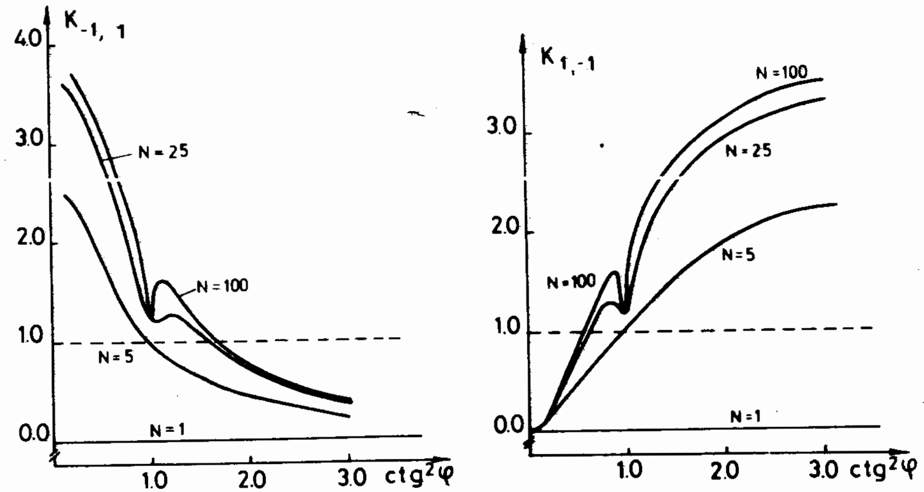


*Fig. 3. The factor* $K_{-1,1}$ *and* $K_{1,-1}$ *as a function of* $ctg^2\phi$ *for* $\gamma_{31}/\gamma_{23}$ *= 2.*

It is easy to see that the operators $-\sin\phi R_{13}$ and $\cos\phi R_{23}$ can be considered as the sources of two spectrum components (Autler-Townes doublet) of the stoke line of frequencies $\omega_{23} - \frac{\delta}{2} - \Omega$ and $\omega_{23} - \frac{\delta}{2} + \Omega$ and for simplicity we call them $A_{-\Omega}$ and $A_{+\Omega}$, respectively. For the single-atom case, by using the operator relation (12) one can find the nondelayed second-order correlation function (14) for the Authler - Townes doublet $A_{\pm\Omega}$ in the form

$$G^{(2)}_{-\Omega,-\Omega} = G^{(2)}_{+\Omega,+\Omega} = 0, \qquad G^{(2)}_{-\Omega,+\Omega} = G^{(2)}_{+\Omega,-\Omega} = 0, \tag{29}$$

thus the validity of the C-S inequality (15) is predicted.

For the collective case, as a result of the anticorrelation between the spectrum components of the stoke line /19/ the C-S inequality for the correlation between spectrum components of the Authler - Townes doublet is valid. In other words, in the stoke line the violation of the C-S inequality is absent either in the single atom case or in the collective case.

In conclusion note that the violation of the C-S inequality for correlations between the Rayleigh and Stoke lines can be investigated by the above described procedure.

## REFERENCES


1. Kimble H.J., Dagenais M., Mandel L. - Phys.Rev.Lett., 1977, 39, p.691.
2. Kimble H.J., Dagenais M., Mandel L. - Phys.Rev., 1978, 118, p.201.
3. Clauser F. - Phys.Rev., 1974, D9, p.853.
4. Slusher R.E., Hollberg L.W., Yurke B., Mertz J.C., Vallet J.F. - Phys.Rev.Lett., 1985, 55, p.2409.
5. Ling-An Wu, Kimble H.J., Hall J.L., Huija Wu. - Phys.Rev. Lett., 1986, 57, p.2520.
6. Cave C.M. - Phys.Rev., 1981, 23D, p.1693.
7. Bondurant R.S., Shapiro J.H. - Phys.Rev., 1984, 30D, p.2548.
8. Yamanoto Y., Haus H.A. - Rev. of Mod. Phys., 1986, 58, p.1001.
9. Golubev Yu.M., Socolov Y-V. - Sov.J.Exper.Theor.Phys., 1984, 87, p.408.
10. Zubairy M.S. - Phys.Lett., 1982, 187, p.162.
11. McNail K.J., Gardiner C.W. - Phys.Rev., 1983, A28, p.1560.
12. Bogolubov N.N.(Jr.), Shumovsky A.S., Tran Quang. - Phys. Lett., 1987, A123, p.71.
13. Bogolubov N.N.(Jr.), Shumovsky A.S., Tran Quang. - J.de Phys., 1987, 48, p.1925.
14. Lawande S.V., Jagatap B.N. - Phys.Lett., 1988, A126, p.329.
15. Kozierowski M. - Phys.Rev., 1987, 36A, p.2973.
16. Agarwal G.S., Jha S. - J.Phys., 1979, B12, p.2655.

17. Agarwal G.S. Springer Trast in Modern Phys. Springer Berlin, 1974.
18. Bogolubov N.N.(Jr.), Shumovsky A.S., Tran Quang. - Physica, 1987, 144A, p.503.
19. Bogolubov N.N.(Jr.), Shumovsky A.S., Tran Quang. - J. de Physique, 1987, 48, p.1671.
20. London R. - Rep.Prog.Phys., 1980, 43, p.38.

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Шумовский А.С., Чан Куанг E17-88-475
Нарушение неравенства Коши - Шварца
в коллективном рассеянии Рамана

Обсуждено нарушение неравенства Коши - Шварца для корреляций между спектральными компонентами рэлеевской линии и между компонентами стоксовой линии коллективного рассеяния Рамана. Показано, что нарушение неравенства Коши - Шварца существует только в рэлеевской линии, кроме этого, для крайних компонент рэлеевской линии нарушение неравенства Коши - Шварца существует для случая большого числа атомов, что означает макроскопичность этого квантового эффекта.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

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Shumovsky A.S., Tran Quang E17-88-475
Violation of the Cauchy - Schwarz Inequality
in Collective Raman Scattering

The violation of Cauchy - Scharz (C-S) inequality for correlations between spectrum components of the Rayleigh line and between spectrum components of the Stoke line in the collective Raman scattering is discussed. It is shown that the violation of the C-S inequality occurs only in the Rayleigh line, moreover, for the sidebands of the Rayleigh line the violation of the C-S inequality takes place for a large number of atoms, which means that this quantum effect has the macroscopic nature.

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

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