INTENSITY CORRELATIONS IN RESONANCE FLUORESCENCE OF TWO ATOMS, COHERENTLY DRIVEN BY A STRONG RESONANT LASER FIELD

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We consider normalized second-order correlations functions for systems of two radiatively interacting atoms. In the strong-field limit we obtain analytical solutions for the intensity correlations, separately for the case of the point model (S^2-invariant system) and for the case when the atoms are separated by a distance r_{12} (S^2-breaking system). In both cases the intensity correlations contain additional components beside those of the usual one atom case. These components modify drastically the light statistics; moreover, light scattered by an S^2-invariant system has a statistics different from that of light scattered by an S^2-breaking system. In the latter case the statistics depends on the interatomic separation r_{12} as well as on the direction of observation, see Figs 2,3. The feasibility of observing photon antibunching in these systems is also discussed.

Introduction

A most interesting problem, recently considered by many authors[1-6], is the existence of additional sidebands in many-atom resonance fluorescence in addition to the usual three peaks of the isolated atom[7-10]. Agarwal et al[4] and Mavroyannis[5] have recently obtained analytical formulae for the spectrum of two-atom resonance fluorescence in the case of strong cooperativity (S^2-invariant system) and have shown that the spectrum contains additional sidebands in 2Ω with an amplitude proportional to $1/\Omega^2$. As pointed out in our recent paper[1], the spectrum of an S^2-breaking system of two atoms also contains additional sidebands in 2Ω with amplitude dependent on $(a/\Omega)^2$ where a is a parameter dependent on interatomic separation, and vanishes when the atoms are far apart. As stated by Agarwal et al[4], the experimental detection of these lines is hardly feasible.

Agarwal et al[11] have also considered intensity correlations in an S^2-invariant system, and have shown that in the case of a high Rabi frequency Ω and high cooperation number S (i.e. for a great number of atoms) it involves additional sidebands in 2Ω with amplitude of zeroth order in the parameter $1/\Omega$. This has been recently confirmed by Mavroyannis[12] and Kilin[6,23]. However, from their results, we cannot discuss the dependence of the intensity correlations on interatomic separation or the angular distribution. Steudel and Richter[13] and Steudel[14] have considered this problem for the case of incoherently pumped two- and three - atomic systems respectively.

In this paper, we consider the intensity correlations in resonance fluorescence of two atoms, equally driven by a strong resonant laser field, for different interatomic distances, as well as in strong cooperativity. To this aim we have adopted the model described in our recent work[1] and obtained a closed system of 15 equations of motion for the atomic correlation functions which we solved by the Laplace transform method. As shown in[1,23] the steady - state solutions for the case of strong cooperativity $a=1$ differ from those for the case $a\neq 1$. This is connected with the S^2-conservation breaking in the case $a\neq 1$. The differences between the solutions for these two cases, for spontaneous emission, have been discussed by Agarwal[15].

In the strong field limit, we obtain separately approximate analytical formulae for the intensity correlations function in the case of strong cooperativity $a=1$ and in the case $a\neq 1$.

Time-evolution of atomic variables

In our model, we consider two like two-level atoms, distant by r_{12}, in the field of an intense resonant beam of laser light. We assume dipolar transitions as permitted between the two levels of either atom. We moreover assume the intense pumping laser beam to be in the coherent state $|\alpha\rangle$. On these assumptions, the Lehmberg[16] master equation approach leads to the following equations of motion describing the time evolution of the pseudo-spin operators of an individual atom

$$\begin{aligned}
\dot{S}_1^+ &= -\frac{\gamma}{2} S_1^+ - i\Omega S_1^z + \gamma_{12} S_2^+ S_1^z \,, \\
\dot{S}_1^- &= -\frac{\gamma}{2} S_1^- + i\Omega S_1^z + \gamma_{12} S_1^z S_2^- \,, \\
\dot{S}_1^z &= -\gamma\left(S_1^z + \tfrac{1}{2}\right) + \tfrac{1}{2} i\Omega\left(S_1^+ - S_1^-\right) - \tfrac{1}{2}\gamma_{12}\left(S_1^+ S_2^- + S_2^+ S_1^-\right) \,,
\end{aligned} \tag{1}$$

where S_1^+ and $S_1^- = (S_1^+)^\dagger$ are operators raising and lowering the energy of atom 1, and S_1^z describes its energy. The equations for the operators of the other atom are of the same form as (1), albeit with the interchange $1 \leftrightarrow 2$ of the indices.
In (1), γ is the Einstein coefficient A, whereas γ_{12}, which is dependent on r_{12} is introduced by Lehmberg[16] factor describes the radiative interaction between the atoms.

In deriving equations (1), for simplicity, we assumed the laser beam frequency as strictly equal to that of the atomic transition ω_0 i.e. zero detuning, and moreover neglected shifts in the levels and dipole-dipole interactions. Moreover, the validity of equations hinges on the assumption that the system is a Markovian one, i.e. that

$$(|\vec{r}_{12}|)_{max} \ll c \cdot \Delta t \quad , \tag{2}$$

where Δt is a time required for appreciable changes in the atomic levels.
Despite these restrictions, equations (1) have been applied successfully to the description of extended systems[18].
The atomic operators of eq. (1) are slowly-varying parts of the full operators:

$$\tilde{S}_1^+(t) = S_1^+(t)\, e^{i\omega_0 t}, \quad \tilde{S}_1^-(t) = S_1^-(t)\, e^{-i\omega_0 t}, \quad \tilde{S}_1^z(t) = S_1^z(t) \quad . \tag{3}$$

The set of equations (1) leads to a finite hierarchy of equations for the correlation functions of the system which can be solved by the Laplace transform method. In the case of two atoms we have a set of 15 equations. The set under consideration can be transformed into the following independent subsets: three of dimension 3 and one of dimension 6.

$$\frac{dU}{d\tau} = AU \quad , \quad \frac{dV}{d\tau} = BV \quad , \quad \frac{dW}{d\tau} = CW \quad , \quad \frac{dX}{d\tau} = DX + m \quad , \tag{4}$$

with

$$A = \begin{bmatrix} -\frac{1}{2}(1-\underline{a}) & 4\beta & \underline{a} \\ -\beta & -1 & 0 \\ 0 & -2\beta & -\frac{1}{2}(3-\underline{a}) \end{bmatrix}, \quad B = \begin{bmatrix} -\frac{1}{2}(1-\underline{a}) & \underline{a} & 0 \\ 0 & -\frac{1}{2}(3-\underline{a}) & 2\beta \\ -\beta & -2\beta & -1 \end{bmatrix}$$

$$C = \begin{bmatrix} -\frac{1}{2}(1+\underline{a}) & \underline{a} & 0 \\ 0 & -\frac{1}{2}(3+\underline{a}) & -2\beta \\ -\beta & 2\beta & -1 \end{bmatrix} \qquad D = \begin{bmatrix} -\frac{1}{2}(1+\underline{a}) & 4\beta & 0 & 0 & \underline{a} & 0 \\ -\beta & -1 & -\underline{a} & 0 & 0 & 0 \\ -\beta & -\underline{a} & -1 & 0 & 2\beta & 4\underline{a} \\ -\beta & 0 & 0 & -1 & 2\beta & 0 \\ 0 & -2\beta & -2\beta & -2\beta & -\frac{1}{2}(3+\underline{a}) & 8\beta \\ 0 & 0 & 0 & 0 & -\beta & -2 \end{bmatrix} \tag{5}$$

and

$$m_i = -4\beta\delta_{1i} \quad ,$$

$$U_1 = S_1^- + S_1^+ - S_2^- - S_2^+ \quad , \qquad U_2 = S_1^+S_1^- - S_2^+S_2^- \quad ,$$

$$U_3 = S_1^+S_1^-S_2^- + S_1^+S_2^+S_1^- - S_2^+S_2^-S_1^- - S_1^+S_2^+S_2^- \quad ,$$

$$V_1 = S_1^- - S_1^+ - S_2^- + S_2^+ \quad , \qquad V_2 = S_1^+S_1^-S_2^- - S_1^+S_2^+S_1^- - S_2^+S_2^-S_1^- + S_2^+S_1^+S_2^- \quad ,$$

$$V_3 = S_2^+S_1^- - S_1^+S_2^- \quad ,$$

$$W_1 = S_1^- - S_1^+ + S_2^- - S_2^+ \quad , \qquad W_2 = S_1^+S_1^-S_2^- - S_1^+S_2^+S_1^- + S_2^+S_2^-S_1^- - S_1^+S_2^+S_2^- \quad ,$$

$$W_3 = S_1^-S_2^- - S_1^+S_2^+ \quad , \tag{6}$$

$$X_1 = S_1^+ + S_2^+ + S_1^- + S_2^- \quad , \qquad X_2 = S_1^+S_1^- + S_2^+S_2^- \quad ,$$

$$X_3 = S_1^+S_2^- + S_2^+S_1^- \quad , \qquad X_4 = S_1^+S_2^+ + S_1^-S_2^- \quad ,$$

$$X_5 = S_1^+S_1^-S_2^- + S_1^+S_2^+S_1^- + S_2^+S_2^-S_1^- + S_1^+S_2^+S_2^- \quad , \qquad X_6 = S_1^+S_2^+S_1^-S_2^- \quad .$$

We have introduced the notation:

$$\tau = \gamma t \quad , \quad \beta = \frac{\Omega}{2\gamma} \quad , \quad \alpha = \frac{\gamma_{12}}{\gamma} \quad . \tag{7}$$

From (4) it is evident that U and V represent antisymmetric combinations of atomic operators while W and X are symmetric combinations. Non-zero steady-state solutions arise from the X quantities only. It is worth noting that the steady-state solutions as calculated from (1) lead to expectation values of the atomic operators other than in the case $a=1$, considered by Agarwal et al[4]. This fact is connected with the S^2 conservation breaking in our case. The determinant of the D matrix becomes zero as $a=1$. This involves a reduction of the dimension of the set from 6 to 5 because of the linear dependence of the variables due to S^2-conservation. The reduced set is identical with that considered by Agarwal et al[4]. Also the so-called scaling factor discussed previously[24] appears in the system with S^2 conserved and does not appear in the steady-state solutions of (1), which have the form:

$$\langle X_1\rangle_s = -\frac{1}{\beta}\left(1 + \frac{a-1}{8\beta^2}\right) \quad , \qquad \langle X_2\rangle_s = \left(1 - \frac{1}{8\beta^2}\right) \quad ,$$

$$\langle X_3\rangle_s = \frac{1}{8\beta^2} \quad , \qquad \langle X_4\rangle_s = \frac{a+1}{8\beta^2} \quad , \tag{8}$$

$$\langle X_5\rangle_s = -\frac{1}{2\beta}\left(1 - \frac{1}{4\beta^2}\right) \quad , \qquad \langle X_6\rangle_s = \frac{1}{4}\left(1 - \frac{1}{4\beta^2}\right) \quad .$$

For $\underline{a}=1$, the solutions of (1) have the following form:

$$\langle X_1\rangle_s = -\frac{4}{3\beta}\left(1-\frac{1}{12\beta^2}\right) , \quad \langle X_2\rangle_s = \left(1-\frac{1}{6\beta^2}\right) ,$$

$$\langle X_3\rangle_s = \frac{1}{3}\left(1+\frac{1}{6\beta^2}\right) , \quad \langle X_4\rangle_s = \frac{1}{3\beta^2} , \tag{9}$$

$$\langle X_5\rangle_s = -\frac{4}{3\beta}\left(1-\frac{1}{3\beta^2}\right) , \quad \langle X_6\rangle_s = \frac{1}{3}\left(1-\frac{1}{3\beta^2}\right) ,$$

Because the square of the total spin of the system can be expressed by the X operators as follows:

$$S^2 = 2 - X_2 + X_3 + 2X_6 , \tag{10}$$

its steady-state expectation value, according to (8) , is

$$\langle S^2\rangle_s = 2 - \frac{1}{2}\left(1-\frac{1}{4\beta^2}\right) . \tag{11}$$

The system reaches this steady-state value even for very small deviations of the parameter $\underline{a}$ from unity i.e. in the majority of real situations.

Intensity correlations

We consider the second-order correlation function

$$G^{(2)}(\vec{R}_1,t\,;\,\vec{R}_2,t+\tau) = \langle \hat{\vec{E}}^{(-)}(\vec{R}_1,t)\hat{\vec{E}}^{(-)}(\vec{R}_2,t+\tau)\hat{\vec{E}}^{(+)}(\vec{R}_2,t+\tau)\hat{\vec{E}}^{(+)}(\vec{R}_1,t)\rangle , \tag{12}$$

for two points $\vec{R}_1$, $\vec{R}_2$ in the far field, i.e. $|\vec{R}_1| = |\vec{R}_2| = R \gg \frac{\omega_0}{c}$, $|\vec{r}_{12}|$ and the normalized second-order correlation function(intensity correlation) defined as:

$$g^{(2)}(\vec{R}_1,t\,;\,\vec{R}_2,t+\tau) = \frac{G^{(2)}(\vec{R}_1,t\,;\,\vec{R}_2,t+\tau)}{G^{(1)}(\vec{R}_1,t)\,G^{(1)}(\vec{R}_2,t+\tau)} \tag{13}$$

For two atoms we have[13] :

$$G^{(2)}(\vec{R}_1,t\,,\vec{R}_2,t+\tau) = \left(\frac{4\pi\hbar k_0\gamma}{R^2}\right)^2 |u(\vec{R}_1)|^2\,|u(\vec{R}_2)|^2$$

$$\sum_{i,j,k,l=1}^{2} \langle S_i^+(t)\,S_j^+(t+\tau)\,S_k^-(t+\tau)\,S_l^-(t)\rangle \exp\left[ik_0\left(\vec{r}_{il}\cdot\vec{R}_1^0 + \vec{r}_{ik}\cdot\vec{R}_2^0\right)\right] , \tag{14}$$

$$G^{(1)}(\vec{R},t) = \left(\frac{4\pi\hbar k_0\gamma}{R^2}\right)|u(\vec{R})|^2 \sum_{i,j=1}^{2} \langle S_i^+(t)\,S_j^-(t)\rangle \exp\left(ik_0\,\vec{r}_{ij}\cdot\vec{R}^0\right) , \tag{15}$$

with: $|u(\vec{R})|^2 = \frac{3}{4\pi}(1-\cos\theta)$, with θ the angle between the observation direction unit vector $\vec{R}^0$ and the atomic transition dipole moment $\vec{\mu}$; $k_0 = \frac{\omega_0}{c} = \frac{2\pi}{\lambda}$, and $\vec{r}_{12}$ the vector connecting both atoms.

From eq. (1) we obtain that, in the steady state, the Laplace transform of (14) has the form:

$$G^{(z)}(z) = N_0 \left\{ \frac{N(z)}{M(z)} - \frac{\left[\beta(2z+3-a)\langle X_3\rangle_s - (2z+1-a)(2z+3-a)\langle X_6\rangle_s\right]\sin\alpha_1 \sin\alpha_2}{(z+1)(2z+1-a)(2z+3-a) + 8\beta^2(2z+1)} \right\} \quad (16)$$

with:

$$N(z) = -\tfrac{1}{4}\beta\Big\{(z+1)(z+2)(z+1-a)(2z+3+a) + 16\beta^2\left[2z^2+(5-2a)z+(1-a)(3+a)\right] +$$

$$+\left[(z+1)(z+2)(z+1-a)(2z+3+a) + 8\beta^2 z(z+1+a)\right]\cos\alpha_2\Big\}\Big[(1+\cos\alpha_1)\langle X_5\rangle_s - \frac{4\beta}{z}\left(\langle X_2\rangle_s + \right.$$

$$\left. + \langle X_3\rangle_s \cos\alpha_1\right)\Big] + \Big\{\tfrac{1}{4}(z+1)^2(z+2)(2z+1+a)(2z+3+a) + 4\beta^2(z+1)\left[4z^2+(8-a)z+3-a\right] -$$

$$-a\left[\tfrac{1}{4}(z+1)(z+2)(2z+1+a)(2z+3+a) + 2\beta^2\left(5z^2+2(4+a)z+2(a+1)\right)\right]\cos\alpha_1\Big\}\langle X_6\rangle_s -$$

$$-\cos\alpha_2\Big\{\tfrac{1}{4}a(z+1)(z+2)(2z+1+a)(2z+3+a) + 4\beta^2(z+1)\left[2z^2+(6-a)z+(4+a-a^2)\right] +$$

$$+2a\beta^2\left[5z^2+2(4+a)z+2(1+a)\right] + 32\beta^4(z+1+a)\Big\}\langle X_6\rangle_s +$$

$$+\cos\alpha_1\cos\alpha_2\Big\{\tfrac{1}{4}(z+1)^2(z+2)(2z+1+a)(2z+3+a) - 4a\beta^2(z+1)(z+2) +$$

$$+4\beta^2(z+1)\left[4z^2+(9+2a)z+5+3a\right] + 48\beta^4\left(z+\tfrac{4}{3}\right)\Big\}\langle X_6\rangle_s \quad , \quad (17)$$

$$M(z) = 32\beta^4\left[2z^2+(5-2a)z+(1-a)(3+a)\right] +$$

$$+2\beta^2(z+1-a)\left[2a^2(2z+1+a) + 2(z+1+a)(2z+1+a)(2z+3-a) - a(z+2)(5z+2a+4) + \right. \quad (18)$$

$$\left. +(z+1)(z+2)(2z+3+a)\right] + (z+1)(z+2)(z+1-a)(z+1+a)\left[z^2+(2+a)z+\tfrac{1}{4}(1+a)(3+a)\right] ,$$

$$N_0 = 2\left(\frac{4\pi\hbar K_0 \gamma}{R^2}\right)^2 |u(\vec{R}_1)|^2 |u(\vec{R}_2)|^2 \quad , \quad \alpha_i = K_0 \vec{r}_{12}\cdot\vec{R}_i^0 \quad . \quad (19)$$

In the strong field limit $\beta \gg 1$, the approximate roots of the denominator of (18) can be found and an analytical formula describing the intensity correlations can be obtained.

We shall determine these functions separately for strict cooperativity $a=1$ and $a \neq 1$. On applying the inverse Laplace transform to (16), making use of (15), (8) and (9) we find that, in the steady state, the intensity correlations in two-atom resonance fluorescence have the form:

strong cooperativity : $a=1$:

$$g^{(2)}(\vec{R},\tau) = 1 - \frac{3}{8} e^{-\frac{3}{4}\tau} \cos(2\beta\tau) + \frac{1}{32} e^{-\frac{3}{2}\tau} + \frac{3}{32} e^{-\frac{5}{2}\tau} \cos(4\beta\tau) \quad , \tag{20}$$

and for $a \neq 1$:

$$g^{(2)}(\vec{R}_1,\vec{R}_2,\tau) = 1 - \frac{1}{2} e^{-\frac{3}{4}\tau} \cos(2\beta\tau) + \frac{1}{2} e^{-\frac{1}{2}(\frac{5}{2}-a)\tau} \cos(2\beta\tau) \sin\alpha_1 \sin\alpha_2 +$$

$$+ \frac{1}{8} e^{-\frac{1}{2}(3+2a)\tau} \cos(4\beta\tau) \cos\alpha_1 \cos\alpha_2 + \frac{1}{8}\left[3\cosh\left(\frac{1}{4}u\tau\right) + \frac{1+6a}{u} \sinh\left(\frac{1}{4}u\tau\right)\right] e^{-\frac{1}{4}(5-2a)\tau} \cos\alpha_1 \cos\alpha_2 \, , \tag{21}$$

with $u = (12a^2 - 4a + 1)^{\frac{1}{2}}$.

Summary and conclusions

Eqs (20) and (21) are analytical formulae, describing the time evolution of the intensity correlations function for light scattered by the system of two atoms. A plot of the intensity correlations, which follows from eq. (20) for $\beta = 3$, is shown in Fig.1. It is in agreement with the numerical result of Agarwal et al[19] . We note that for $\tau = 0$ $g^{(2)} = 0.75$ and the function grows for small τ . The last result confirms those of Carmichael[20] and Drummond[21] . The situation is the opposite if the atoms are at a distance $r_{12} \neq 0$. Figs 2 and 3 give the shape of (21) for $\beta = 3$, $\alpha_1 = \alpha_2$, different distances between the atoms, and various emission directions with respect to the line connecting both atoms. We find that in this case, for $\tau = 0$, $g^{(2)}(0) = 1$, the function decreases for small τ . If $\alpha_1 \neq \alpha_2$ and $\tau = 0$,

$$g^{(2)}(\vec{R}_1,\vec{R}_2,0) = \frac{1}{2}\left\{1 + \cos K_0 \vec{r}_{12}(\vec{R}_1^0 - \vec{R}_2^0)\right\} \quad , \tag{22}$$

hence, if

$$K_0 \vec{r}_{12}(\vec{R}_1^0 - \vec{R}_2^0) = 2\pi n \quad , \qquad n = 0,1,2,\ldots \quad , \tag{23}$$

we have $g^{(2)}(0) = 1$, whereas for

$$K_0 \vec{r}_{12}(\vec{R}_1^0 - \vec{R}_2^0) = \pi(2n+1), \quad n = 0,1,2,\ldots \quad , \tag{24}$$

we have $g^{(2)}(0) = 0$.

It is noteworthy that no photon anticorrelation effect ($g^{(2)}(0) < 1$) takes place if the interatomic distance r_{12} and observation direction fulfil the relation (23). Moreover, if $\alpha_1 = \alpha_2$ i.e. if the photons are recorded in one direction, $g^{(2)}(0) = 1$ always. This confirms Kimble and Mandel's[17] suggestion that anticorrelation of photons, emitted by a system of many atoms, cannot be observed using a single photodetector.

Photon anticorrelation ($g^{(2)}(0) < 1$) is accessible to observation with a setup involving two photodetectors. Then, $\alpha_1 \neq \alpha_2$ provided that the condition (24) is fulfilled.

Eq. (22) leads to yet another conclusion, namely, that $g^{(2)}(0)$ is independent of the radiative interaction between the atoms (it is determined by the parameter a). This is so because the two atoms are pumped by the same laser beam, so that a well defined phase relationship exists provided that the distance between the two atoms is constant between vibrations of the dipole of either atoms. It is independent of a , since the process does not result from the emission of a photon by one of the atoms and its absorption by the other, but is due solely to coherent pumping, which imposes a fixed difference of phase.

When deriving Eqs (20) and (21) we omitted dipole-dipole interaction between the atoms; however, as shown by Freedhoff[22] and Kilin[23] , such interaction does not affect the shape of $g^{(2)}(\tau)$ significantly.

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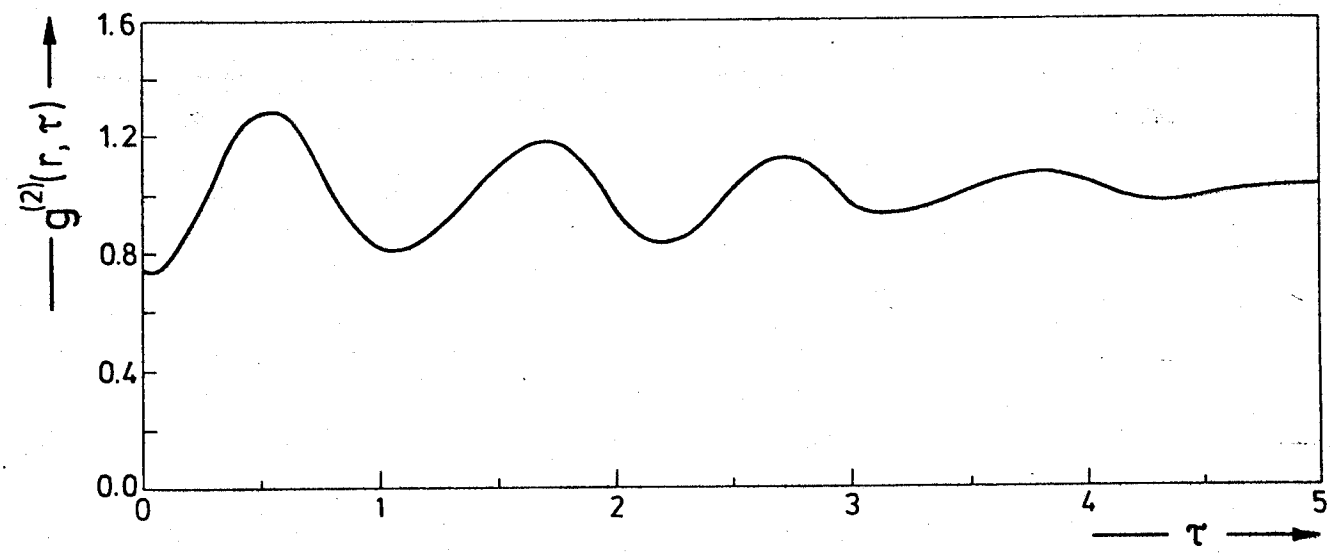

Fig.1. The normalized second-order correlation function $g^{(2)}(\tau)$ for S^2-invariant system plotted versus the dimensionless time τ with $\beta = 3$.

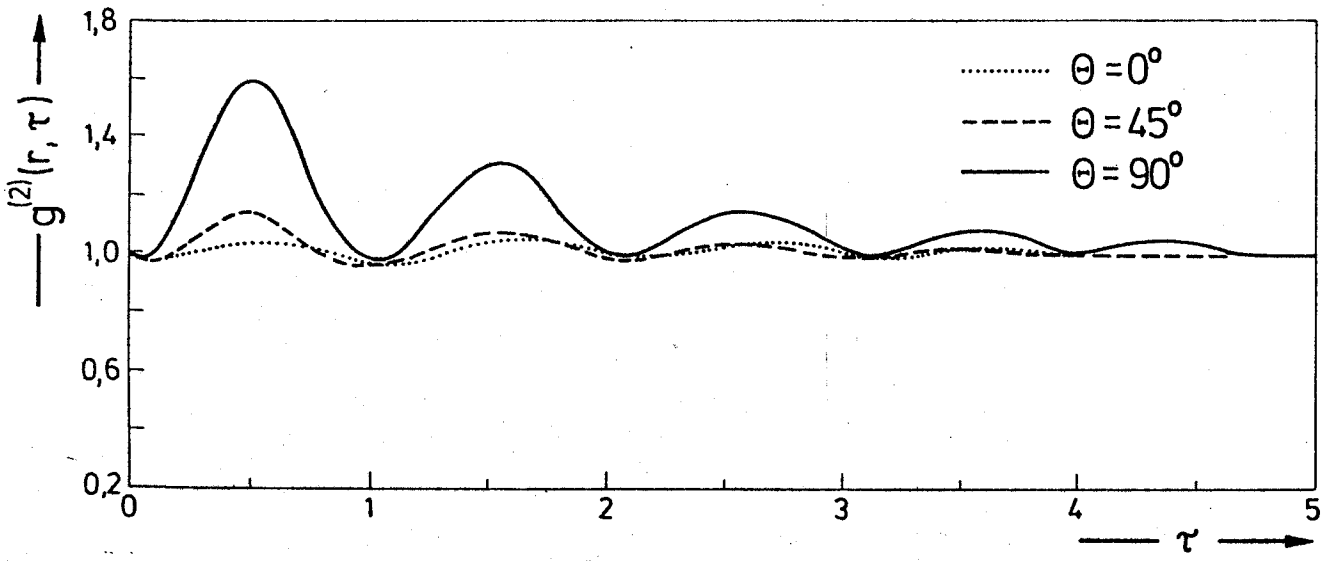

Fig.2. The normalized second-order correlation function $g^{(2)}(\vec{R},\vec{R},\tau)$ for S^2-breaking system plotted versus the dimensionless time τ with $\beta = 3$, $|\vec{r}_{12}| = \frac{1}{4}\lambda$ and various directions against the line connecting the two atoms.

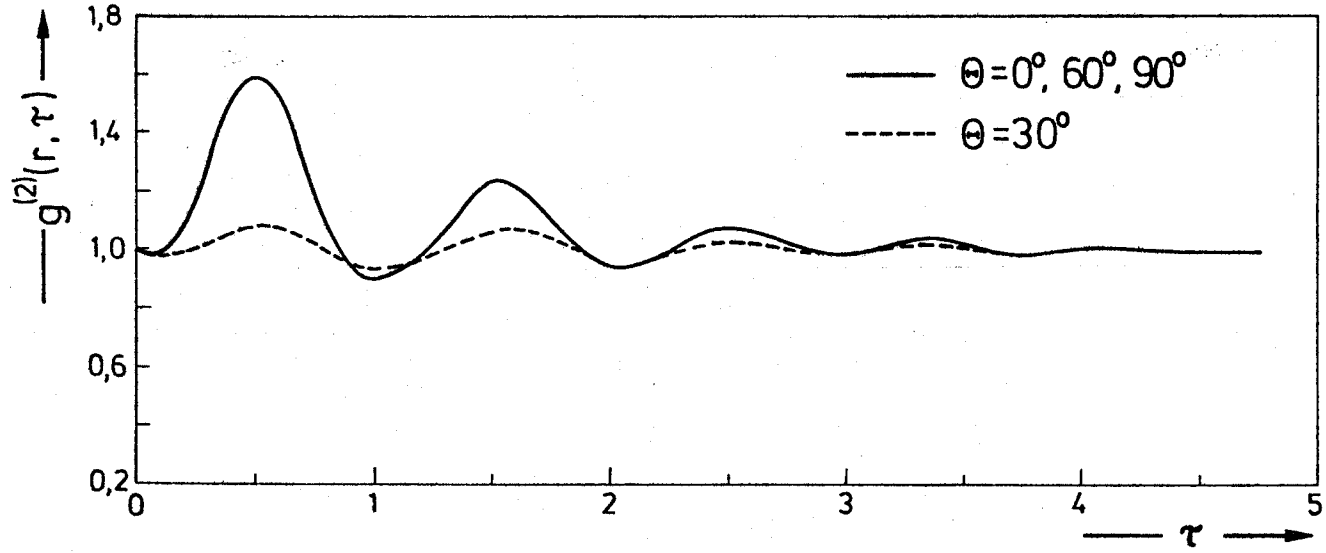

Fig.3. The same as Fig.2 but with $|\vec{r}_{12}| = 2\lambda$.