AMPLITUDE-SQUARED SQUEEZING IN TWO-ATOM RESONANCE FLUORESCENCE ☆

Z. FICEK, R. TANAŚ and S. KIELICH

Nonlinear Optics Division, Institute of Physics, A. Mickiewicz University, 60-780 Poznań, Poland

Amplitude-squared squeezing (ASS), defined recently by Hillery, is extended to atomic dipole operators and considered for resonance fluorescence from two interacting atoms. It is shown that amplitude-squared squeezing, which does not appear in the one-atom case, can occur in two-atom resonance fluorescence. A considerable amount of ASS is obtained for small detuning and large interatomic separations. As interatomic separation decreases, dipole–dipole interaction between the atoms increases and ASS shifts to the region of larger detuning but its value strongly decreases. For very strong dipole–dipole interaction or for very large detuning, the ASS tends to zero. Moreover, it is found that the ASS is directly connected with photon antibunching and can be measured by correlation techniques. The connection between ASS in the atomic variables and ASS in a fluorescent field is also discussed.

1. Introduction

The nonclassical nature of squeezed states in an electromagnetic field is now well understood theoretically [1] and their successful experimental observation is a fact [2–5]. Squeezed states exhibit reduced fluctuations in one of the two quadrature components, the in-phase $E_1 = E^{(+)} + E^{(-)}$ or the out-of-phase component $E_2 = -i(E^{(+)} - E^{(-)})$ of the electromagnetic field, below the vacuum limit, or below that achievable in completely coherent field.

With the development of techniques for the measurement of higher-order correlation functions it is possible to define higher-order squeezing effect. Hong and Mandel [6] defined a state to be squeezed to $2N$th order if the expectation value of the $2N$th power of the difference between a field quadrature component and its average value is less than it would be in a coherent state. Braunstein and McLachlan [7] defined generalized squeezed states which are higher-order analogs of the squeezing operator [8]. Hillery [9] defined amplitude-squared squeezing, which corresponds to the squeezing of the variables which describe the real and imaginary parts of the square of the complex amplitude of the electromagnetic field.

He found that this type of squeezing is a relatively common feature of two-photon processes and appears in a number of nonlinear optical processes.

The idea of amplitude-squared squeezing (ASS) may be extended to other quantum systems. In particular, to atomic two-level systems, where radiation properties are described by atomic dipole operators. However, ASS cannot be formed for a single atom because the square of the dipole raising and lowering operators is zero. Physically, this is a consequence of the fact that one two-level atom cannot emit two photons simultaneously. For a multiatom system the square of the dipole raising and lowering operators is different from zero and squared amplitudes of atomic operators can be defined. As stated by Hillery, the squared amplitude of the operators in a given system describes two-photon processes. Therefore, such a process should occur in multi-atom systems, and has been found in two-atom resonance fluorescence power spectra [10,11]. This is easy to explain within the framework of collective states of a two-level system [12]. The hamiltonian for a two-atom system can be diagonalized including the dipole–dipole interaction, giving the eigenstates $|0\rangle = |1\rangle_1 |1\rangle_2$, $|\pm\rangle = (1/\sqrt{2})$ $(|2\rangle_1 |1\rangle_2 \pm |1\rangle_1 |2\rangle_2)$ and $|2\rangle = |2\rangle_1 |2\rangle_2$ with energies $E_0 = 0$, $E_\pm = \hbar(\omega_0 \pm \Omega_{12})$ and $E_2 = 2\hbar\omega_0$, where ω_0 is the

transition frequency between the lower state $|1\rangle_i$ and the excited state $|2\rangle_i$ of the ith atom ($i=1,2$) and Ω_{12} is the dipole–dipole interaction potential. In fact, the two-atom system is equivalent to a single four-level system with one upper state $|2\rangle$, one ground state $|0\rangle$ and two intermediate states $|\pm\rangle$. The population of the upper state $|2\rangle$ can be transferred to the states $|+\rangle$, $|-\rangle$ and $|0\rangle$ of frequencies (energies) $\omega_0-\Omega_{12}$, $\omega_0+\Omega_{12}$ and $2\omega_0$, respectively. This means that the transition $|2\rangle\rightarrow|0\rangle$ with frequency $2\omega_0$ is allowed in two-photon process.

In the present paper we consider the possibility of obtaining amplitude-squared squeezing in steady-state resonance fluorescence from two interacting atoms. We define the amplitude-squared atomic dipole operators and, from Lehmberg's [13] master equation, we obtain analytical expressions for fluctuations in the amplitude-squared operators. We discuss the conditions for the appearence and the possibility of measuring such squeezing.

2. Amplitude-squared squeezing in the atomic operators.

In order to discuss ASS in multi-atom resonance fluorescence we define operators which represent the real and imaginary parts of the square of the collective atomic dipole raising and lowering operators

$$S_1 = \tfrac{1}{2}(S^{+2}+S^{-2}),$$

$$S_2 = (1/2i)(S^{+2}-S^{-2}), \tag{1}$$

where

$$S^{\pm} = \sum_{i=1}^{N} S_i^{\pm} \exp(\pm i\mathbf{k}\cdot\mathbf{r}_i),$$

with the operators S_i^+ and $S_i^- = (S_i^+)^\dagger$ which raise and lower the energy of the ith atom. These operators satisfy the well-known commutation relations for Pauli spin-half operators:

$$[S_i^+, S_j^-] = 2S_i^z\delta_{ij}, \quad [S_i^z, S_j^\pm] = \pm S_i^\pm\delta_{ij},$$

$$[S_i^+, S_j^-]_+ = \delta_{ij}, \tag{2}$$

with S_i^z describing the energy of the ith atom.

From the commutation relations (2), the corresponding commutator of S_1 and S_2 is given by

$$[S_1, S_2] = 2(S^+S^-+S^-S^+-1)S^z, \tag{3}$$

which vanishes for one-atom resonance fluorescence. For two atoms ($N=2$), the commutator (3) has the simple form

$$[S_1, S_2] = iS^z, \tag{4}$$

with $S_1 = S_1^+S_2^+ + S_1^-S_2^-$ and $S_2 = -i(S_1^+S_2^+ - S_1^-S_2^-)$. Squeezing in one of the components S_1 or S_2 of the square of the atomic operators may by observed if one of the quantities $p_{1,2}$, defined as

$$p_{1,2} = (\langle(\Delta S_{1,2})^2\rangle - |\langle S^z\rangle|)/|\langle S^z\rangle|, \tag{5}$$

is negative, in other words, if the ASS variance of one of the two noncommuting observables (S_1 or S_2) is less than one-half of the absolute value of their commutator, i.e., $\langle(\Delta S_{1,2})^2\rangle < |\langle S^z\rangle|$. The parameter p_i describes the degree of squeezing [14] and takes its maximum negative values -1 for perfect squeezing. It is obvious from (5) that the parameters p_1 and p_2 are dependent on the expectation values of the atomic operators which, on the other hand, are readily obtained from Lehmberg's [13] master equation. This master equation for the slowly varying parts of the operators $S_i^\pm$ and S_i^z reads

$$(d/d\tau)S_i^\pm = -\tfrac{1}{2}(1\mp i\Delta)S_i^\pm$$

$$+ [\pm 2i\beta+(a\mp ib)S_j^\pm]S_i^z, \tag{6a}$$

$$(d/d\tau)S_i^z = -(\tfrac{1}{2}+S_i^z)+i\beta(S_i^+-S_i^-)$$

$$-\tfrac{1}{2}[(a+ib)S_i^+S_j^-+\text{h.c.}], \quad i\neq j, \tag{6b}$$

where

$$\tau = 2\gamma t, \quad \beta = \Omega/4\gamma, \quad a = \gamma_{12}/\gamma,$$

$$b = \Omega_{12}/\gamma, \quad \Delta = (\omega_0-\omega_L)/\gamma. \tag{6c}$$

In eqs. (6), Ω is the Rabi frequency, Δ the detuning of the driving laser field frequency ω_L from the atomic transition frequency ω_0, and 2γ the Einstein A coefficient for spontaneous emission. The collective parameters Ω_{12} and γ_{12} are dependent on the interatomic distance r_{12} and reflect the retarded dipole-dipole and radiative interaction between the atoms [12,13]. When deriving eqs. (6) we choose the reference frame so that the atoms are at the positions $r_1 = (-\tfrac{1}{2}r_{12}, 0, 0)$ and $r_2 = (\tfrac{1}{2}r_{12}, 0, 0)$, the phase of the field is choosen as zero, and the Rabi frequency Ω is real and the same for both atoms. In this case,

our model is similar to that of one atom in a resonant field in the presence of a conducting metallic surface [15,16]. Such a model has been realized experimentally [17]. Eqs. (6) enable us to obtain analytical solutions for the parameters $p_{1,2}$. For two atoms they lead to a closed system of fifteen equations describing the evolution of the atomic variables [18]. By setting the left-hand side of eqs. (6) equal to zero this system of equations gives the steady-state solution. For the correlation functions needed to calculate $p_{1,2}$ we have the following solutions

$$\langle S_1^+ S_2^+ + S_1^- S_2^- \rangle = 8\beta^2 [(1+a) - \Delta(\Delta+b)]/D ,$$

$$\langle S_1^+ S_2^+ - S_1^- S_2^- \rangle = 0 , \quad \langle S_1^+ S_2^+ S_2^- S_1^- \rangle = 16\beta^4/D ,$$

$$\langle S^z \rangle = -(1+\Delta^2)[8\beta^2 + (1+a)^2 + (\Delta+b)^2]/D ,$$

$$(7)$$

with

$$D = 64\beta^4 + 16(1+\Delta^2)\beta^2$$

$$+ (1+\Delta^2)[(1+a)^2 + (\Delta+b)^2] .$$

The above steady-state solutions, which include the collective damping parameter a, the dipole–dipole interaction parameter b, and the detuning Δ, permit the calculation of the parameters $p_{1,2}$ describing the amplitude-squared fluctuations in steady-state two-atom resonance fluorescence.

Insertion of (7) into (5) leads to

$$p_1 = \frac{32\beta^4\{D - 2[(1+a) - \Delta(\Delta+b)]^2\}}{D(1+\Delta^2)[8\beta^2 + (1+a)^2 + (\Delta+b)^2]} , \quad (8)$$

$$p_2 = \frac{32\beta^4}{(1+\Delta^2)[8\beta^2 + (1+a)^2 + (\Delta+b)^2]} . \quad (9)$$

Our solutions (8) and (9) describe the amplitude-squared fluctuations in steady-state resonance fluorescence from two atoms coherently driven by a coherent laser field. Eqs. (8), (9) are valid for any values of the field strength and interatomic distances r_{12}. It is obvious from eq. (9) that p_2 is always positive, independently of β, Δ and b. Amplitude-squared squeezing occurs only in p_1. This parameter (p_1) is illustrated graphically in fig. 1 as a function of the detuning Δ for $\beta = 0.2$, and for different values of the interatomic separations r_{12} normalized to the resonant wavelength λ. This graph shows that p_1 strongly depends on the detuning Δ and a pro-
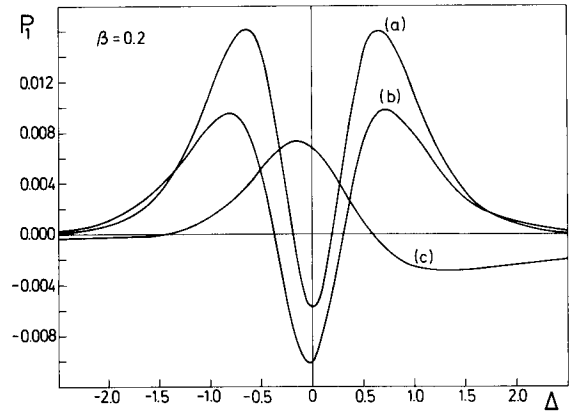

Fig. 1. Degree of amplitude-squeezing p_1 versus Δ for field strength $\beta = 0.2$ and different interatomic separations r_{12}. Curve (a) $r_{12}/\lambda \gg 1$; curve (b) $r_{12}/\lambda = 0.5$; curve (c) $r_{12}/\lambda = 0.125$. For curve (c), the parameter p_1 has been multiplied by a factor of 10.

nounced ASS ($p_1 < 0$) can be obtained in such a two-atom system for certain values of Δ. For large interatomic separations r_{12} this occurs for small Δ ($|\Delta| \ll 1$) with a maximum at $\Delta = 0$. As the interatomic separation r_{12} decreases, ASS shifts to the region of larger Δ but its values are very small compared to the case of large r_{12}. The largest values of ASS are obtained for large r_{12} and Δ near zero. In fig. 2, p_1 is plotted for a fixed atomic distance and different laser field intensities. For small β squeezing first increases with β and next decreases for larger β and does not appear for $\beta > 0.25$. Here, it is interesting to compare the behaviour of the ASS with the standard squeezing discussed for two-atom resonance fluorescence
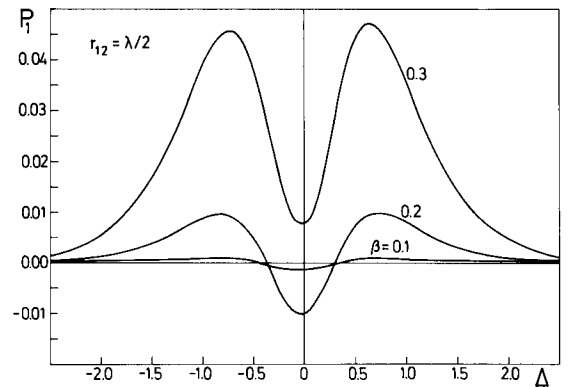

Fig. 2. p_1 versus Δ for the separation $r_{12} = \lambda/2$ and for different field strengths β.

in ref. [18]. As it has been shown [18], the standard squeezing is very sensitive to the dipole–dipole interaction between the atoms. As the latter increases, squeezing shifts to the region of finite Δ, and maximum squeezing appears for $\Delta = -b$, i.e., when the dipole–dipole interaction b and the detuning Δ cancel out mutually. This takes place for the one-photon transitions

$$|+\rangle \to |0\rangle \text{ or } |-\rangle \to |0\rangle.$$

Here however, ASS is also sensitive to the dipole–dipole interaction but its maximum appears for Δ considerably smaller than b. This means that ASS is now connected with the two-photon transition $|2\rangle \to |0\rangle$. This can be explained within the framework of collective atomic states. For not too strong laser field ($\beta < 1$) and large r_{12} (i.e., for small dipole–dipole interaction values b), the atomic levels are not shifted and the two-photon transition $|2\rangle \to |0\rangle$ is not modified. As r_{12} decreases (dipole–dipole interaction increases) the atomic levels become shifted and the transition $|2\rangle \to |0\rangle$ becomes destroyed leading to a decrease in ASS. As r_{12} becomes very small, a particular pair of energy levels of the two-atom system are strongly shifted by the dipol–dipole interaction, the laser field can be tuned to resonance with the respective pair of levels (greater Δ), and ASS appears again. This is shown in fig. 3, where p_1 is plotted versus r_{12}/λ for $\beta = 0.2$ and different detunings Δ. It is evident that for larger Δ squeezing appears only for small r_{12}. According to eqs. (8) and (9), for a not too strong field, p_1 and
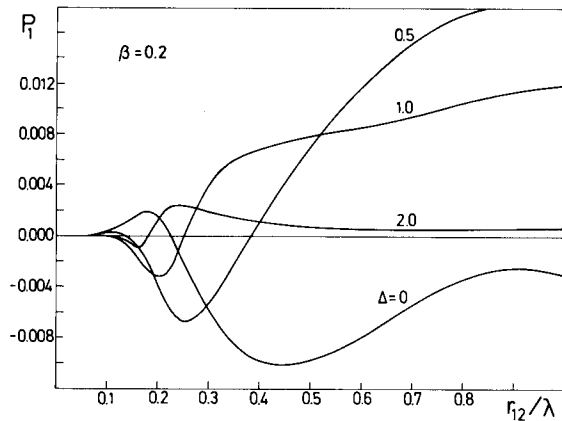

Fig. 3. p_1 versus interatomic separation for $\beta = 0.2$ and for different detunings Δ.

p_2 tend to zero as $|b| \to \infty$ (see fig. 3) or as $|\Delta| \to \infty$, i.e., when we have atomic transitions far from the resonance.

Now let us discuss a possible experimental configuration for the detection of ASS in two-atom resonance fluorescence. For this we use the one-time normalized intensity correlation function

$$g^{(2)}(R_1, t; R_2, t)$$
$$= G^{(2)}(R_1, t; R_2, t)/G^{(1)}(R_1, t) \, G^{(1)}(R_2, t),$$

which, for $R_1 = R_2 \perp r_{12}$ and two atoms, reads

$$g^{(2)}(R, t; R; t)$$
$$= 4\langle S_1^{+j} S_2^+ S_2^- S_1^- \rangle / \langle S^+ S^- \rangle^2. \qquad (10)$$

Inserting (10) into (5) we obtain

$$p_1 = (\langle S^+ S^- \rangle^2/2|\langle S^z \rangle|) \{g^{(2)}(R, t; R, t)$$
$$- [(1+a) - \Delta(\Delta+b)]^2/2[(1+\Delta^2) + 4\beta^2]^2\}. \qquad (11)$$

This equation shows that ASS is related with the photon correlations. For $\Delta = 0$ and an extremely weak field ($\beta \ll 1$) the condition required for ASS is

$$g^{(2)}(R, t; R, t) < \tfrac{1}{2}(1+a)^2. \qquad (12)$$

Since $|a| \leqslant 1$, photon antibunching implies ASS for large interatomic separations ($a \approx 0$), whereas for small interatomic separations ($a \approx 1$) the condition $g^{(2)}(R, t, R, t) < 2$ is required for ASS. However, for $|\Delta| \geqslant 1$ the second term in eq. (11) is always smaller than unity, meaning that p_1 is negative only if the proton antibunching is present. These calculations show that the techniques used for measuring photon antibunching [19,20] can be used to obtain information about ASS.

Finally, we shall show that the ASS calculated above for the atomic dipole operators can be easily generalized to the field operators. On defining the in-phase component E_1 and out-of-phase component E_2 of the amplitude-squared electromagnetic field with the squared positive (negative) frequency parts $E^{(+)2}$ ($E^{(-)2}$)

$$E_1 = E^{(+)2} + E^{(-)2},$$
$$E_2 = -i(E^{(+)2} - E^{(-)2}), \qquad (13)$$

we have the commutation rule

$$[E_1, E_2] = 8iC(E^{(-)}E^{(+)} + \tfrac{1}{2}C) , \qquad (14)$$

where C is a positive c-number. Amplitude-squared squeezing of the fluorescent field is defined by the requirement that the variance of one of the two non-commuting observables (E_1 or E_2) shall be less than $4C(\langle E^{(-)}E^{(+)}\rangle + \tfrac{1}{2}C)$. Since the following relations hold for the field operators:

$$\langle (\Delta E_{1,2})^2 \rangle = 4C(\langle E^{(-)}E^{(+)}\rangle + \tfrac{1}{2}C)$$

$$+ \langle :(\Delta E_{1,2})^2: \rangle , \qquad (15)$$

where the colon stands for normal ordering of the operators, the squeezing conditions imply negative values of the normally ordered variance of the squeezed amplitude-squared field component. The normally ordered variance of the fluorescent field can be derived using the following relation between the radiation field and the atomic operators in the far field limit [12,13]:

$$E^{(+)}(R, t) = E_0^{(+)}(R, t)$$

$$+ \tfrac{1}{2}\Psi(R) \sum_{i=1}^{2} S_i^-(t - R/c) , \qquad (16)$$

where $E_0^{(+)}(R, t)$ is the amplitude of the incident field and $\Psi(R)$ is the geometrical factor. According to eq. (16), the normally ordered variance of the amplitude-squared fluorescent field in any direction other than that of the incident beam is given by

$$\langle :(\Delta E_{1,2})^2: \rangle$$

$$= \tfrac{1}{4}\Psi^4(R) \; [\langle (\Delta S_{1,2})^2 \rangle + \langle S^z \rangle] . \qquad (17)$$

In the steady-state $\langle S^z \rangle$ is negative (see eqs. (7)) i.e., $\langle S^z \rangle = -|\langle S^z \rangle|$, and ASS in atomic variables implies a negative value of the normally ordered variance of the corresponding component of the am-

plitude-squared fluorescence field. This means that ASS in atomic dipoles for steady-state resonance fluorescence leads directly to ASS in the fluorescence field.

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