Xun-Li Feng, Zhao-Yang Chen<sup>a</sup>, and Zhi-Zhan Xu

CCAST (World Laboratory), P.O.Box 8730, Beijing 100080, and Laboratory for High Intensity Optics, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai, 201800, P.R. China

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**Abstract.** The nondegenerate two-photon interaction of a two-level atom with a broadband multimode squeezed vacuum is investigated. We find that in the two-photon process the squeezed vacuum has a driving effect on the atom which can lead to an ac Stark effect when the average photon number of the squeezed vacuum is larger than a critical value.

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### 1 Introduction

In recent years, squeezed light sources have become available in the laboratory and a great deal of attention has been given to the interactions between the squeezed light and the atomic systems. The pioneer work performed by Gardiner in 1986 suggested that the atomic phase decay can be inhibited by a squeezed vacuum as compared to the normal vacuum, thus leading to a subnatural linewidth in the atomic fluorescence spectrum [1]. The subnatural linewidth phenomena in the resonance fluorescence [2] and the absorption spectrum [3] for such systems were reported in the further studies by others. Many new features of the squeezed light interactions, such as atomic level shifts [4] and the generalized Bloch-Siegert shifts [5], were also found in the one-photon processes. Because of the strong two-photon correlations present in squeezed light, it is natural to extend such studies to the two-photon interactions between the atom and the squeezed light. However, in most of the early studies, the two-photon processes are almost about three-level atomic systems [6]. To our knowledge, the investigations of the nondegenerate two-photon interactions between a two-level atom and a broadband squeezed vacuum have not been reported up to now. The purpose of this paper is to solve this problem. We find that in the two-photon process the squeezed vacuum has a driving effect on the atom which can lead to an ac Stark effect when the average photon number of the squeezed vacuum is larger than a critical value. That is, the frequency shifts can occur in the oscillation of the atomic dipole moment, thus the atomic two-photon fluorescence spectrum can exhibit a three-peaked structure.

#### 2 Model and master equation

The interactions between a single two-level atom and a multimode light field can be described by a Dicke Hamiltonian for one-photon [7] and multiphoton [8] processes. Recently, He *et al.* studied the nondegenerate two-photon interactions between a two-level atom and the multimode light field in a normal vacuum state [9] and in a squeezed vacuum state [10], respectively. Following He *et al.*, we consider that a single atom, having two levels of the same parity, is coupled to a broadband multimode squeezed vacuum via a nondegenerate two-photon process. One-photon transition is assumed forbidden. The Hamiltonian of the system in the rotating-wave approximation is of the form

$$H = H_{\rm A} + H_{\rm F} + H_{\rm I},\tag{1}$$

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where  $H_{\rm A}$  and  $H_{\rm F}$  are the free Hamiltonians of the atom and the light field, respectively,  $H_{\rm I}$  is the interaction Hamiltonian,

$$H_{\rm A} = \frac{1}{2} \hbar \Omega \sigma_z,$$
  

$$H_{\rm F} = \hbar \int d\omega b^{\dagger}(\omega) b(\omega) \omega,$$
  

$$H_{\rm I} = \hbar \int d\omega d\omega' g(\omega, \omega') [b(\omega_{\rm c} + \omega) b(\omega_{\rm c} + \omega') \sigma^{+} + b^{\dagger}(\omega_{\rm c} + \omega) b^{\dagger}(\omega_{\rm c} + \omega') \sigma],$$
(2)

where  $\Omega$  is the frequency of the atomic transition,  $\sigma$ ,  $\sigma^+$ and  $\sigma_z$  are the pseudospin atomic opertors,  $b(\omega)$  ( $b^{\dagger}(\omega)$ ) is the photon annihilation (creation) operator with frequency  $\omega$  of the squeezed vacuum,  $\omega_c$  is the central frequency of the squeezed vacuum,  $g(\omega, \omega')$  is the coupling constant.

<sup>&</sup>lt;sup>a</sup> e-mail: chen\_zhaoyang@163.net

We treat the broadband squeezed vacuum as a reservoir into which the two-level atom decays through twophoton transition. In practice, as emphasized by Gardiner [1] and Carmichael *et al.* [2], it is difficult to prepare the broadband squeezed environment, which requires all the modes that interact with the atom to be squeezed. That is, the squeezed modes must occupy the whole  $4\pi$ solid angle of space. In order to overcome this obstacle, many schemes have been put forward to mimic such an environment by means of more convenient methods [11–14] recently. Lutkenhaus, Cirac and Zoller proposed a scheme to realize such an environment by using the interference between the different spontaneous emission channels of a degenerate four-level system driven by weak fields [12]. Wiseman demonstrated that the in-loop squeezing produced by electro-optical feedback is like real squeezing to an in-loop atom [13]. Zhou and Swain employed a twolevel atom driven by a strong coherent field and a weak, amplitude-fluctuating field for mimicking a squeezed environment [14].

As a theoretical analysis, we still follow Gardiner's treatment in his original paper [1] and use the output from a degenerate parametric oscillator (DPO) operating below threshold [15] as a source of the broadband squeezed vacuum, assuming that the linewidth of the output from the DPO is much larger than the linewidth of the atomic transition. For our convenience, we define a reservoir operator as

$$F = \int d\omega d\omega' g(\omega, \omega') b(\omega_{\rm c} + \omega) b(\omega_{\rm c} + \omega').$$
(3)

By extending Gardiner's methods [1, 16] to the two-photon process, we obtain the mean values and the two-time correlation functions of the reservoir operator F(t) and its Hermitian conjugate operator  $F^{\dagger}(t)$ ,

$$\langle F(t) \rangle = \langle F^{\dagger}(t) \rangle^* = \frac{1}{2} \sqrt{\pi \Gamma} C \exp(-2i\omega_{\rm c} t),$$
 (4)

$$\langle F^{\dagger}(t)F(t')\rangle = \frac{1}{4}\pi\Gamma[|C|^{2} + \kappa N^{2}\delta(t-t')]\exp[2i\omega_{c}(t-t')], (5)$$

$$\langle F(t)F^{\dagger}(t')\rangle = \frac{1}{4}\pi\Gamma[|C|^{2} +\kappa(N+1)^{2}\delta(t-t')]\exp[-2i\omega_{c}(t-t')], \quad (6)$$

$$\langle F(t)F(t')\rangle = \frac{1}{4}\pi\Gamma \left[C^2 + \kappa M^2 \delta(t-t')\right] \exp\left[-2i\omega_{\rm c}(t+t')\right], (7)$$
  
$$\langle F^{\dagger}(t)F^{\dagger}(t')\rangle = \langle F(t)F(t')\rangle^*, \qquad (8)$$

where

$$C = \frac{\zeta^2 - \eta^2}{2} \left( \frac{1}{\zeta} + \frac{1}{\eta} \right) \exp(i\theta), N = \frac{(\zeta^2 - \eta^2)^2}{(2\zeta\eta)^2},$$
$$M = \frac{(\zeta^2 - \eta^2)(\zeta^2 + \eta^2)}{(2\zeta\eta)^2} \exp(i\theta),$$
$$\zeta = \frac{1}{2}\kappa + |\epsilon|, \quad \eta = \frac{1}{2}\kappa - |\epsilon|, \quad \epsilon = |\epsilon| \exp(i\theta),$$

 $\epsilon$  and  $\kappa$  are, respectively, the amplification constant and the bandwidth of the squeezed vacuum [1,15]. N is the

average photon number. M and C are the measures of the squeezing degree of the squeezed vacuum with the relations  $|M| = \sqrt{N(N+1)}$  and  $|C| = \kappa \sqrt{N}$ .  $\Gamma = 2\pi g^2(\omega_c) \mathcal{D}(\omega_c)$  denotes the two-photon natural linewidth, where  $\mathcal{D}(\omega_c)$  is the mode density of the multimode field.

Equation (4) implies that the mean values of the reservoir operators,  $\langle F(t) \rangle$  and  $\langle F^{\dagger}(t) \rangle$ , which are generally zero for one-photon processes both in normal vacuum and in squeezed vacuum and for two-photon processes in normal vacuum, are nonzero. The nonzero mean values of F(t) and  $F^{\dagger}(t)$  arise from the two-photon correlations of the squeezed vacuum. It is these terms that give rise to the ac Stark effect discussed below. As indicated in equations (5-8), the correlation functions have a similar form of two terms with the first term a constant and the second a delta function. The constant term appears because the reservoir operators have the nonzero mean values. If the correlation functions are those in which these mean values are subtracted, such as  $\langle (F(t) - \langle F(t) \rangle)(F(t') - \langle F(t') \rangle) \rangle$ , and these are delta correlated as usual. So the squeezed vacuum reservoir in the two-photon process is still Markovian.

The master equation for the reduced density operator  $\rho$  of the atom can be derived starting from the Hamiltonian (1), using the standard technique and the correlations (4-8). On resonance ( $\Omega = 2\omega_c$ ), the equation in a frame rotating at frequency  $\Omega$  is given by

$$\frac{\partial \rho}{\partial t} = -\frac{i}{2} \sqrt{\pi \Gamma} \left( C[\sigma^+, \rho] + C^*[\sigma, \rho] \right) + \frac{1}{8} \pi \kappa \Gamma \\
\times \left\{ (N+1)^2 \left( [\sigma\rho, \sigma^+] + [\sigma, \rho\sigma^+] \right) \\
+ N^2 \left( [\sigma^+, \rho\sigma] + [\sigma^+\rho, \sigma] \right) \\
+ M^2 \left( [\sigma^+\rho, \sigma^+] + [\sigma^+, \rho\sigma^+] \right) \\
+ M^{*2} \left( [\sigma\rho, \sigma] + [\sigma, \rho\sigma] \right) \right\}.$$
(9)

Not including the first term, the master equation (9) is similar to that of one-photon process in the squeezed vacuum [1] but with the different coefficients. The first term in equation (9) arises from the nonzero mean values of the reservoir operators, it shows a driving effect of the squeezed vacuum on the atom, because this term is the same as the contribution of an external classical driving field to the atom.

# 3 Two-photon Bloch equation and ac Stark effect

From the master equation (9), we obtain the two-photon Bloch equation

$$\frac{\mathrm{d}}{\mathrm{d}t} \begin{pmatrix} \langle \sigma \rangle \\ \langle \sigma^+ \rangle \\ \langle \sigma_z \rangle \end{pmatrix} = \begin{pmatrix} -\frac{1}{2}\gamma & B\exp(i2\theta) & iR\exp(i\theta) \\ B\exp(-i2\theta) & -\frac{1}{2}\gamma & -iR\exp(-i\theta) \\ i2R\exp(-i\theta) & -i2R\exp(i\theta) & -\gamma \end{pmatrix} \\ \times \begin{pmatrix} \langle \sigma \rangle \\ \langle \sigma^+ \rangle \\ \langle \sigma_z \rangle \end{pmatrix} - \begin{pmatrix} 0 \\ 0 \\ \gamma_0 \end{pmatrix}, \tag{10}$$

where

$$\gamma = \frac{1}{4}\pi\kappa\Gamma(2N^2 + 2N + 1),$$
  

$$B = \frac{1}{4}\pi\kappa\Gamma |M|^2,$$
  

$$R = \frac{1}{2}\sqrt{\pi\Gamma} |C|,$$
  

$$\gamma_0 = \frac{1}{4}\pi\kappa\Gamma(2N + 1).$$

The eigenequation corresponding to equation (10) is

$$\left(\lambda + \frac{1}{2}\gamma - B\right) \times \left[\lambda^2 + \left(\frac{3}{2}\gamma + B\right)\lambda + \gamma\left(\frac{1}{2}\gamma + B\right) + 4R^2\right] = 0 \quad (11)$$

with the solutions

$$\lambda_{1} = -\frac{1}{8}\pi\kappa\Gamma,$$
  

$$\lambda_{2,3} = -\frac{1}{2}\pi\kappa\Gamma\left[N\left(N+1\right) + \frac{3}{8}\right]$$
  

$$\pm\frac{1}{2}\kappa\sqrt{\left(\frac{1}{8}\pi\Gamma\right)^{2} - 4\pi\Gamma N}.$$
(12)

Thus, the time-dependent analytical solutions for  $\langle \sigma(t) \rangle$ ,  $\langle \sigma^+(t) \rangle$  and  $\langle \sigma_z(t) \rangle$  can be found easily for the atom initially in the upper state,

$$\langle \sigma(t) \rangle_{\text{Normal}} = i \operatorname{Re}^{i(\Omega t + \theta)} \\ \times \left[ \frac{\lambda_2 - \gamma_0}{\lambda_2(\lambda_2 - \lambda_3)} e^{\lambda_2 t} + \frac{\lambda_3 - \gamma_0}{\lambda_3(\lambda_3 - \lambda_2)} e^{\lambda_3 t} - \frac{\gamma_0}{\lambda_2 \lambda_3} \right], \quad (13) \\ \langle \sigma^+(t) \rangle_{\text{Normal}} = (\langle \sigma(t) \rangle_{\text{Normal}})^*, \qquad (14)$$

where we have transformed the rotating frame into the normal frame with the following relations:

$$\langle \sigma(t) \rangle_{\text{Normal}} = \langle \sigma(t) \rangle \exp(i\Omega t), \langle \sigma^+(t) \rangle_{\text{Normal}} = \langle \sigma^+(t) \rangle \exp(-i\Omega t),$$

the subscript index Normal stands for in the normal frame.

From equation (12), we find that there is a threshold,  $\pi\Gamma/16^2$ , for the average photon number N. When N is below the threshold, the eigenroots  $\lambda_{2,3}$  are real. In this case, equation (13) indicates that the atomic dipole moment oscillates with a single frequency  $\Omega$  only. When N is above the threshold, however, the eigenroots  $\lambda_{2,3}$  are complex numbers,

$$\lambda_{2,3} = \lambda_{\rm R} \pm i\lambda_{\rm I},$$
  

$$\lambda_{\rm R} = -\frac{1}{2}\pi\kappa\Gamma\left[N\left(N+1\right) + \frac{3}{8}\right],$$
  

$$\lambda_{\rm I} = \frac{\kappa}{2}\sqrt{4\pi\Gamma N - \left(\frac{1}{8}\pi\Gamma\right)^2}.$$
(15)

In this case, equation (13) can be rewritten as

$$\langle \sigma(t) \rangle_{\text{Normal}} = i \text{Re}^{i\theta} \left[ -\frac{(\lambda_{\text{R}} - \gamma_0) - i\lambda_{\text{I}}}{2\lambda_{\text{I}}(\lambda_{\text{I}} + i\lambda_{\text{R}})} e^{\lambda_{\text{R}}t} e^{i(\Omega - \lambda_{\text{I}})t} \right. \\ \left. -\frac{\gamma_0}{\lambda_{\text{R}}^2 + \lambda_{\text{I}}^2} e^{i\Omega t} + \frac{(\lambda_{\text{R}} - \gamma_0) + i\lambda_{\text{I}}}{2\lambda_{\text{I}}(-\lambda_{\text{I}} + i\lambda_{\text{R}})} e^{\lambda_{\text{R}}t} e^{i(\Omega + \lambda_{\text{I}})t} \right].$$
(16)

Equation (16) reveals that the atomic dipole moment oscillates with three different frequencies: the central frequency  $\Omega$  and the two sideband frequencies,  $\Omega + \lambda_{\rm I}$  and  $\Omega - \lambda_{\rm I}$ . The two sideband frequencies are the shifts from the central one, up and down, by the value  $\lambda_{\rm I}$ , whose magnitude depends on both the average photon number N and the bandwidth  $\kappa$  of the squeezed vacuum field. This phenomenon is generally called ac Stark effect. As analyzed above, the ac Stark effect arises from the nonzero mean values of the squeezed reservoir operators, *i.e.* from the two-photon correlations of the squeezed vacuum. The ac Stark effect can be detected by absorption and emission experiments. In the next section, we shall give the atomic two-photon fluorescence spectrum to demonstrate the ac Stark effect.

## 4 Two-photon fluorescence spectrum of the atom

The fluorescent light field consists of the elastically scattered field and the inelastically scattered field, the latter arises from radiation from the atom. In this section, we shall calculate the spectrum of the inelastically scattered field, *i.e.*, the incoherent spectrum. In the two-photon process, the incoherent spectrum is given by [9]

$$S(\omega) = \frac{1}{\pi} \operatorname{Re} \left\{ \int_0^\infty d\tau \langle \delta \sigma^{\dagger}(0) \delta \sigma(\tau) \rangle_{\rm ss} \exp \left[ i \left( \omega - \frac{1}{2} \Omega \right) \tau \right] \right\}, (17)$$

where Re denotes the real part and  $\delta\sigma = \sigma - \langle \sigma \rangle$ ,  $\delta\sigma^+ = \sigma^+ - \langle \sigma \rangle^+$ . The steady-state correlation function appearing in equation (17) can be calculated by means of the quantum regression theorem from the two-photon Bloch equation (10) under the steady-state conditions

$$\langle \sigma \rangle_{\rm ss} = (\langle \sigma^+ \rangle_{\rm ss})^* = -\frac{iR\gamma_0 \exp(i\theta)}{\lambda_2\lambda_3},$$
  
$$\langle \sigma_z \rangle_{\rm ss} = -\frac{\gamma_0(\gamma/2 + B)}{\lambda_2\lambda_3}.$$
 (18)

We find that

$$\langle \delta \sigma^{\dagger}(0) \delta \sigma(\tau) \rangle = C_1 \exp(\lambda_1 \tau) + C_2 \exp(\lambda_2 \tau) + C_3 \exp(\lambda_3 \tau), \quad (19)$$



Fig. 1. Incoherent fluorescent spectra when N is below the threshold for  $\Gamma = 100.0$  and different N: N = 0.1 (dotted line), N = 0.2 (solid line), and N = 0.5 (dashed line).

where

$$C_{1} = \frac{(\gamma/2 + B)(\gamma - \gamma_{0}) + 4R^{2}}{4\lambda_{2}\lambda_{3}},$$

$$C_{2} = \frac{\lambda_{3} + \gamma/2 + B}{4(\lambda_{2} - \lambda_{3})} \left[ \frac{\gamma_{0}(\gamma/2 + B)}{\lambda_{2}\lambda_{3}} - 1 \right]$$

$$+ \frac{\gamma_{0}R^{2}(\lambda_{2} + \gamma_{0})}{\lambda_{2}^{2}\lambda_{3}(\lambda_{2} - \lambda_{3})},$$

$$C_{3} = \frac{\lambda_{2} + \gamma/2 + B}{4(\lambda_{3} - \lambda_{2})} \left[ \frac{\gamma_{0}(\gamma/2 + B)}{\lambda_{2}\lambda_{3}} - 1 \right]$$

$$+ \frac{\gamma_{0}R^{2}(\lambda_{3} + \gamma_{0})}{\lambda_{2}\lambda_{3}^{2}(\lambda_{3} - \lambda_{2})}.$$
(20)

It is not difficult to prove that the coefficients  $C_i$  (i = 1, 2, 3) are independent of  $\kappa$ .

When the average photon number N is below the threshold, the incoherent spectrum is given by

$$S(\omega) = -\frac{1}{\pi} \left[ \frac{C_1 \lambda_1}{\lambda_1^2 + (\omega - \frac{1}{2}\Omega)^2} + \frac{C_2 \lambda_2}{\lambda_2^2 + (\omega - \frac{1}{2}\Omega)^2} + \frac{C_3 \lambda_3}{\lambda_3^2 + (\omega - \frac{1}{2}\Omega)^2} \right].$$
 (21)

This is the sum of three Lorentzians all centered on  $\Omega/2$ , with widths  $2\lambda_1$ ,  $2\lambda_2$  and  $2\lambda_3$ , respectively. The lineshape is shown in Figure 1.

When N is above the threshold, the incoherent spectrum is of the form

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$$S(\omega) = -\frac{1}{\pi} \left[ \frac{C_{\rm R}\lambda_{\rm R} - C_{\rm I} \left(\omega - \frac{1}{2}\Omega - \lambda_{\rm I}\right)}{\lambda_{\rm R}^2 + \left(\omega - \frac{1}{2}\Omega - \lambda_{\rm I}\right)^2} + \frac{C_{\rm I}\lambda_{\rm I}}{\lambda_{\rm I}^2 + \left(\omega - \frac{1}{2}\Omega\right)^2} + \frac{C_{\rm R}\lambda_{\rm R} + C_{\rm I} \left(\omega - \frac{1}{2}\Omega + \lambda_{\rm I}\right)}{\lambda_{\rm R}^2 + \left(\omega - \frac{1}{2}\Omega + \lambda_{\rm I}\right)^2} \right].$$
(22)



Fig. 2. Incoherent fluorescent spectra when N is above the threshold for  $\Gamma = 0.01$  and different N: N = 0.1 (dotted line), N = 0.2 (solid line), and N = 0.8 (dashed line).

Here, we have set  $C_{2,3} = C_{\rm R} \pm i C_{\rm I}$ , with

$$C_{\rm R} = \frac{1}{8} \left[ 1 - \frac{\gamma_0(\gamma/2 + B)}{\lambda_{\rm R}^2 + \lambda_{\rm I}^2} \right] - \frac{\gamma_0^2 R^2}{2(\lambda_{\rm R}^2 + \lambda_{\rm I}^2)^2},$$

$$C_{\rm I} = \frac{1}{8} \left[ 1 - \frac{\gamma_0(\gamma/2 + B)}{\lambda_{\rm R}^2 + \lambda_{\rm I}^2} \right] \frac{\lambda_{\rm R} + \gamma/2 + B}{\lambda_{\rm I}}$$

$$- \frac{\gamma_0 R^2 (\lambda_{\rm R}^2 + \lambda_{\rm I}^2 + \lambda_{\rm R} \gamma_0)}{2\lambda_{\rm I} (\lambda_{\rm R}^2 + \lambda_{\rm I}^2)^2}.$$
(23)

Equation (23) indicates the incoherent spectrum has a three-peaked structure, with the central peak at  $\Omega/2$ and two sidebands at  $\Omega/2 - \lambda_{\rm I}$  and  $\Omega/2 + \lambda_{\rm I}$ , respectively. The two sidebands just correspond to the ac Stark shifts discussed in Section 3. The two sidebands are of the same width  $2\lambda_{\rm R}$ , but the width of the central peak is  $2\lambda_{\rm I}$ . Figure 2 illustrates the three-peaked spectrum. From Figure 2 we can also find that the two sidebands move away from the central peak and become wide as the average photon number N increases. So if N is sufficiently large, the two sidebands will disappear due to their too big widths.

#### 5 Summary

In this paper, we have studied the nondegenerate twophoton interaction of a two-level atom with a broadband squeezed vacuum. We find that in the two-photon process the squeezed vacuum has a driving effect on the atom as an external classical field does. The driving effect can give rise to an ac Stark effect when the average photon number of the squeezed vacuum is larger than a threshold. That is, the oscillating frequency of the atomic dipole moment can be splitted from one to three, thus the atomic two-photon fluorescence spectrum can exhibit three-peaked structure.

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