Atomic two-photon excitation by an injected squeezed vacuum in a cavity

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We study the two-photon excitation rate and the spectrum of a three-level atom in the Ξ configuration interacting with a resonant cavity mode which is coupled to a broadband squeezed vacuum through its input-output mirror. We show that the optical Bloch equations in the cavity environment are formally identical to these in free space but, in the bad-cavity limit, with cavity-modified parameters describing the injected squeezed vacuum. It is shown that the two-photon excitation rate (which is proportional to the upper-level population) has two components, one depending linearly and the other quadratically on the squeezed photon number N, in excellent agreement with experiment [Georgiades *et al.*, Phys. Rev. Lett. **75**, 3426 (1995)]. The presence of the term quadratic in N is entirely due to the effectively imperfect correlation of the squeezed photon pairs in the bad cavity. We also find that the two-photon excitation spectrum (as measured by the fluorescent emission from the upper level to the intermediate level) exhibits an extremely narrow peak at line center, under appropriate choices of the bad-cavity parameters. All these results are independent of the squeezed phase. [S1050-2947(96)10909-4]

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I. INTRODUCTION

The interaction of quadrature squeezed light with atoms has been a topic of considerable theoretical and experimental interest since the first successful generation of squeezed light about a decade ago [1]. This field of research was initiated by Gardiner [2], who showed that broadband quadrature squeezed light can in principle inhibit the phase decay of a two-level atom, thus resulting in a subnatural linewidth in the atomic fluorescence spectrum. The modifications of the resonance fluorescence spectrum of such a system were examined by Carmichael, Lane, and Walls [3], who found that, for strong driving fields, the relative heights and widths of the peaks in the spectrum are very sensitive to the squeezed phase. A number of other investigations of the effect of quadrature squeezing on resonance fluorescence and probe absorption of a coherently driven two-level atom have been also reported [4–10], with particularly distinctive spectral features, for example, hole burning and dispersive profiles at line center [4], suppression of the fluorescence radiation [5], Fano-like profiles [6], and gain without population inversion [7], being found. These departures from the usual spectral features of a driven atom in an ordinary vacuum are due to the fundamental alteration of atomic radiative processes by the nonclassical field.

Because of the strong two-photon correlations present in squeezed light, it is quite natural to extend the studies of atom–squeezed-light interactions to three-level schemes where the behavior of atomic two-photon transitions will be influenced in a significant way by squeezed light. Recently, great attention has been paid to the effect of a squeezed vacuum on the two-photon transition rate in a three-level Ξ system [11,12], where the dependence of the two-photon transition rate on excitation intensity in the weak field limit is predicted to be linear, instead of the quadratic dependence observed when coherent or thermal light is employed as the excitation field [13]. Buzek *et al.* [14] showed that pure states can be achieved when a three-level atom (in the Ξ, V, or Λ configuration) is damped by a single, squeezed vacuum reservoir. Other related problems, such as population trapping [15] of V- and Λ-type atoms in the presence of an additional coherent driving field, the optical double-resonance spectrum [16] in a Ξ -type atomic system, and the resonance fluorescence spectra [17] of a three-level atom (in all possible configurations) which interacts with two conventional laser sources and two independent squeezed vacua, have been also investigated extensively. A variety of interesting phenomena have been predicted.

All the effects described above take place in free space. However, as emphasized by Gardiner [2] and Carmichael *et al.* [3], such experiments would be difficult to perform since they require *all* the modes with which the atom interacts to be squeezed. That is, the squeezed modes must occupy the whole 4π solid angle of space. In order to avoid this difficulty, the cavity situation is a natural alternative to consider, since the atom interacts strongly only with the privileged cavity mode. Only the modes within a small solid angle about this cavity mode need be squeezed. Parkins and Gardiner [18] have considered a single atom inside a microcavity which has squeezed light incident upon the output mirror, and demonstrated that the inhibition of atomic phase decays should still be observable under appropriate conditions. The resonance fluorescence and absorption spectra of a two-level atom in an optical cavity with injected squeezed vacuum have been also reported in the bad cavity limit [19], where unusual spectral features, for instance, hole-burning and dispersion profiles at line center, have been shown to persist, although they are less pronounced than in free space due to a reduction of the degree of two-photon correlation of the squeezing in the bad optical cavity. However, they remain detectable within the cavity environment. The two-photon population inversion of a three-level atom in the Ξ-type structure excited by a squeezed vacuum in a Fabry-Pérot microcavity has recently been reported [20]. Gardiner and Parkins [21] have also evaluated the two-photon absorption rate of a Ξ-type atom by squeezed light in an alternative cavity configuration where the nonlinear optical parametric oscillator medium generating the squeezed vacuum is put inside a cavity together with the atom, and the cavity mode and atom are damped by ordinary vacua. They have shown via a numerical solution of the master equation of the system that the linear dependence of the absorption rate on the excitation intensity for weak excitation still holds.

Recently, Georgiades *et al.* [22], have carried out the first experimental investigation of the modification of the fundamental atomic radiative processes brought about by illumination with nonclassical light. More specifically, they observed the rate of the two-photon transition $6S_{1/2}\rightarrow 6D_{5/2}$ for trapped atomic cesium excited by a squeezed vacuum light generated via nondegenerate parametric down conversion. The rate observed shows both linear and quadratic growths with the light intensity for weak excitation, in contrast with the purely quadratic dependence produced by classical light sources [13], in qualitative agreement with theoretical studies of a three-level Ξ-type atom interacting with a single squeezed vacuum [11,12,21].

In this paper we study the two-photon transitions of a Ξ-type atom excited by a squeezed vacuum injected through the input-output cavity mirror inside an optical cavity. This system is somewhat in accordance with the experimental configuration set up by Georgiades *et al.* [22] . We present a simple and physically appealing theory of this situation using the cavity renormalized squeezing parameters that we have recently introduced [19]. We also point out that interesting features can be seen in the resonance fluorescence spectrum of this system.

The current paper is organized as follows: the physical system under consideration is described in Sec. II, where we also derive the optical Bloch equations in the bad-cavity limit. We show that these are formally identical to those in free space, the only difference being a redefinition of the parameters describing spontaneous decay and the squeezing. In Sec. III we investigate the two-photon transition rate by determining the upper-level population as a function of the squeezed photon number (excitation intensity). We find that the rate of two-photon excitation consists of a linear and a quadratic dependence on the squeezed photon number, which is in excellent agreement with the experimental observation [22]. We calculate the fluorescence emission spectrum of such an atomic system under the two-photon excitation by squeezed light inside a cavity in Sec. IV, where a distinctive narrow spectral feature is found in the bad-cavity limit. This spectral feature does not occur in free space [11], nor in a cavity with an injected black-body radiation field. A summary is contained in Sec. V.

II. OPTICAL BLOCH EQUATIONS IN THE BAD-CAVITY LIMIT

We consider the two-photon interaction of a single Ξ-type atom with a cavity mode that is assumed to be coupled to an injected broadband squeezed vacuum via the lossy mirror of a single-ended cavity. The atom is damped by a single, standard vacuum environment. The configuration considered is shown in Fig. 1. For simplicity, the cavity resonance frequency ω_c and the center frequency ω_s of the squeezed vacuum are taken to be identical, and the atom is also assumed to be on two-photon resonance with the cavity mode, i.e., $E_3-E_1=2\omega_c$. In a frame rotating at the resonance cavity frequency ω_c, the master equation is of the form

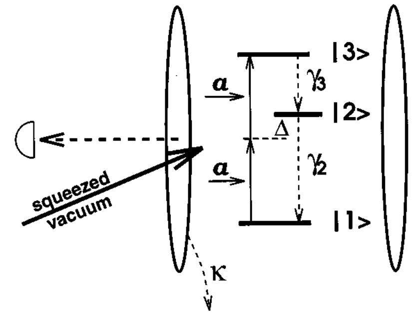

FIG. 1. Cavity configuration of the two-photon excitation of a Ξ-type atom by an injected squeezed vacuum, where a is the annihilation operator of the cavity mode, Δ is the one-photon detuning and κ,γ_3, and γ_2 are the decay constants of the cavity mode, the upper atomic level, and the intermediate atomic level, respectively.

$$\dot{\rho}=-i[H,\rho]+\mathcal{L}_c\rho+\mathcal{L}_a\rho, \tag{1}$$

with

$$H=\frac{\Delta}{2}A_{22}+ig_2(a^\dagger A_{12}-A_{21}a)+ig_3(a^\dagger A_{23}-A_{32}a), \tag{2}$$

$$\mathcal{L}_c\rho=\kappa(N+1)(2a\rho a^\dagger-a^\dagger a\rho-\rho a^\dagger a)+\kappa N(2a^\dagger\rho a-aa^\dagger\rho-\rho aa^\dagger)-\kappa Me^{-i\phi}(2a\rho a-a^2\rho-\rho a^2)-\kappa Me^{i\phi}(2a^\dagger\rho a^\dagger-a^{\dagger 2}\rho-\rho a^{\dagger 2}), \tag{3}$$

and

$$\mathcal{L}_a\rho=\frac{1}{2}\gamma_3(2A_{23}\rho A_{32}-A_{33}\rho-\rho A_{33})+\frac{1}{2}\gamma_2(2A_{12}\rho A_{21}-A_{22}\rho-\rho A_{22})+\gamma_{23}A_{23}\rho A_{21}+\gamma_{23}A_{12}\rho A_{32}, \tag{4}$$

where H describes the two-photon interaction of the atom with the cavity mode, $\Delta=\omega_c-(E_2-E_1)=(E_3-E_2)-\omega_c$ is the detuning of the intermediate state $|2\rangle$ from the exact one-photon resonance, and $A_{lk}\equiv|l\rangle\langle k|$ represents a population operator for $l=k$ and a dipole transition operator for $l\neq k$. The cavity mode has annihilation and creation operators a and $a^\dagger$. g_2 and g_3 are the coupling constants of the respective atomic transitions $|1\rangle\leftrightarrow|2\rangle$ and $|2\rangle\leftrightarrow|3\rangle$ to the cavity mode. Here transitions between the excited level $|3\rangle$ and the ground level $|1\rangle$ are dipole forbidden. $\mathcal{L}_c\rho$ and $\mathcal{L}_a\rho$, respectively describe damping of the cavity field by the injected squeezed reservoir, and atomic damping to background modes other than the privileged cavity mode. κ is the cavity decay constant, γ_2 and γ_3 are the decay constants of the levels $|2\rangle$ and $|3\rangle$ respectively, and γ_{23} is associated with the quantum correlation between the $|1\rangle\leftrightarrow|2\rangle$ and $|2\rangle\leftrightarrow|3\rangle$ transition pathways, which is strongly dependent upon the orientations of the atomic dipole polarizations. In what follows we take $\gamma_{23}=\sqrt{\gamma_2\gamma_3}$ to make the quantum interference effects maximal.

The real parameters N and M characterize the broadband squeezed reservoir through the relations

$$\langle a^\dagger(\omega)a(\omega')\rangle = N\delta(\omega-\omega'),$$

$$\langle a(\omega)a^\dagger(\omega')\rangle = (N+1)\delta(\omega-\omega'),$$

$$\langle a(\omega)a(\omega')\rangle = Me^{i\phi}\delta(2\omega_s-\omega-\omega'), \tag{5}$$

where $a(\omega)$ is the photon annihilation operator for the squeezed vacuum, ω_s is the center frequency of the squeezed vacuum (which is tuned close to the cavity frequency ω_c), N is the squeezing photon number, M measures the strength of the two-photon correlations, and ϕ is the phase of the squeezed vacuum. The squeezing is assumed to be broadband so that N and M are independent of the frequency ω. They obey the relation

$$M=\eta\sqrt{N(N+1)}, \quad (0\leqslant\eta\leqslant 1). \tag{6}$$

The quantity η measures the degree of two-photon correlations in the squeezed vacuum. Its interpretation is simple: $\eta=0$ implies no squeezing and our cavity field is then equivalently damped by a chaotic field. $\eta=1$, however, corresponds to the reservoir being in an ideal or minimum uncertainty squeezed state. We refer to this as perfect correlation, since in this instance the photon twins, characteristic of a squeezed vacuum, are maximally correlated for the particular value of N. In this paper we assume that the squeezed vacuum is injected into the cavity with perfect two-photon correlation ($\eta=1$). Theory predicts that the squeezed output of an ideal parametric oscillator is characterized by $\eta=1$ [23].

If one performs the squeeze transformation on the master equation (1) by

$$\tilde{\rho}=S\rho S^\dagger, \tag{7}$$

where S is the usual squeeze operator [1] that transforms the annihilation and creation operators as

$$SaS^\dagger=\mu a+\nu a^\dagger,$$

$$Sa^\dagger S^\dagger=\nu^* a+\mu a^\dagger, \tag{8}$$

with $\mu=\sqrt{N+1}$ and $\nu=\sqrt{N}e^{i\phi}$, in the squeezed picture the master equation (1) takes the form

$$\dot{\tilde{\rho}}=-i[\tilde{H},\tilde{\rho}]+\mathcal{L}_a\tilde{\rho}+\mathcal{L}_v\tilde{\rho}, \tag{9}$$

where

$$\tilde{H}=\frac{\Delta}{2}A_{22}+ig_2[(\mu a^\dagger+\nu^* a)A_{12}-A_{21}(\nu a^\dagger+\mu a)]$$
$$+ig_3[(\mu a^\dagger+\nu^* a)A_{23}-A_{32}(\nu a^\dagger+\mu a)],$$
$$\mathcal{L}_v\tilde{\rho}=\kappa(2a\tilde{\rho}a^\dagger-\tilde{\rho}a^\dagger a-a^\dagger a\tilde{\rho}). \tag{10}$$

The transformed cavity mode is damped by a standard vacuum, and the effect of the squeezed reservoir is transferred to the atom-cavity interaction. The alternative viewpoint is that the atom couples to the transformed cavity mode.

Accordingly, the time evolution of the expectation values of the atom is governed by

$$\langle\dot{A}_{33}\rangle=-\gamma_3\langle A_{33}\rangle-g_3(\nu\langle a^\dagger A_{32}\rangle+\mu\langle a^\dagger A_{23}\rangle+\mu\langle A_{32}a\rangle$$
$$+\nu^*\langle A_{23}a\rangle),$$

$$\langle\dot{A}_{22}\rangle=-\gamma_2\langle A_{22}\rangle+\gamma_3\langle A_{33}\rangle-g_2(\nu\langle a^\dagger A_{21}\rangle+\mu\langle a^\dagger A_{12}\rangle$$
$$+\mu\langle A_{21}a\rangle+\nu^*\langle A_{12}a\rangle)+g_3(\nu\langle a^\dagger A_{32}\rangle+\mu\langle a^\dagger A_{23}\rangle$$
$$+\mu\langle A_{32}a\rangle+\nu^*\langle A_{23}a\rangle),$$

$$\langle\dot{A}_{32}\rangle=-\frac{1}{2}(\gamma_2+\gamma_3+i\Delta)\langle A_{32}\rangle-g_2(\nu\langle a^\dagger A_{31}\rangle+\mu\langle A_{31}a\rangle)$$
$$+g_3(\mu\langle a^\dagger A_{33}\rangle-\mu\langle a^\dagger A_{22}\rangle+\nu^*\langle A_{33}a\rangle$$
$$-\nu^*\langle A_{22}a\rangle),$$

$$\langle\dot{A}_{21}\rangle=-\frac{1}{2}(\gamma_2-i\Delta)\langle A_{21}\rangle+\gamma_{23}\langle A_{32}\rangle+g_3(\nu\langle a^\dagger A_{31}\rangle$$
$$+\mu\langle A_{31}a\rangle)-g_2(\mu\langle a^\dagger A_{22}\rangle-\mu\langle a^\dagger A_{11}\rangle+\nu^*\langle A_{22}a\rangle$$
$$-\nu^*\langle A_{11}a\rangle),$$

$$\langle\dot{A}_{31}\rangle=-\frac{1}{2}\gamma_3\langle A_{31}\rangle+g_2(\mu\langle a^\dagger A_{32}\rangle+\nu^*\langle A_{32}a\rangle)$$
$$-g_3(\mu\langle a^\dagger A_{21}\rangle+\nu^*\langle A_{21}a\rangle), \tag{11}$$

where $\langle\ \rangle$ denotes the averages in the squeezed picture.

These equations contain expectation values of higher-order operators, which lead to a series of coupled equations for the atomic and cavity mode variables. In what follows, we are interested in the bad-cavity limit [19,24]; that is,

$$\kappa\gg\gamma_l,\ g_l, \quad \text{with } g_l\gg\gamma_l, \quad (l=2,3), \tag{12}$$

but with $C=g_l^2/\kappa\gamma_l$, $(l=2,3)$ [25] finite, where C is the effective cooperativity parameter of a single atom familiar from optical bistability. Also, to ensure the validity of the broadband squeezing assumption, the bandwidth of squeezing would need to be large compared to κ. These conditions imply that the cavity-mode response to the squeezed vacuum reservoir is much faster than that produced by its interaction with the atom. Then the atom always experiences the cavity mode in the state induced by the squeezed reservoir, which permits one to adiabatically eliminate the cavity-mode variables. This gives rise to a set of equations for the atomic variables only, which can be exactly solved. To do this we consider the Heisenberg-Langevin equation of the transformed cavity mode, which is expressed as

$$\dot{a}=-\kappa a+g_2(\mu A_{12}-\nu A_{21})+g_3(\mu A_{23}-\nu A_{32})+L(t), \tag{13}$$

whose formal solution is given by

$$a(t)=a(0)e^{-\kappa t}+g_2\int_0^t[\mu A_{12}(t-\tau)-\nu A_{21}(t-\tau)]e^{-\kappa\tau}d\tau$$
$$+g_3\int_0^t[\mu A_{23}(t-\tau)-\nu A_{32}(t-\tau)]e^{-\kappa\tau}d\tau$$
$$+\int_0^t L(t-\tau)e^{-\kappa\tau}d\tau, \quad (14)$$

where $L(t)$ indicates the Langevin operator associated with the standard vacuum [24]. In the bad-cavity limit (12), the transformed cavity mode $a(t)$ is reduced to

$$a(t)=\frac{g_2}{\kappa}[\mu A_{12}(t)-\nu A_{21}(t)]+\frac{g_3}{\kappa}[\mu A_{23}(t)-\nu A_{32}(t)]$$
$$+\frac{1}{\kappa}L(t). \quad (15)$$

Substituting $a(t)$ into (11) and utilizing the property $\langle L(t)\rangle=0$, we obtain the following cavity modified Bloch equations:

$$\langle\dot{A}_{33}\rangle=2\gamma_3 CN\langle A_{22}\rangle-\gamma_3(1+2C+2CN)\langle A_{33}\rangle$$
$$+\gamma_{23}CMe^{i\phi}\langle A_{31}\rangle+\gamma_{23}CMe^{-i\phi}\langle A_{13}\rangle,$$

$$\langle\dot{A}_{22}\rangle=2\gamma_2 CN\langle A_{11}\rangle-[\gamma_2(1+2C+2CN)+2\gamma_3 CN]\langle A_{22}\rangle$$
$$+\gamma_3(1+2C+2CN)\langle A_{33}\rangle-2\gamma_{23}CMe^{i\phi}\langle A_{31}\rangle$$
$$-2\gamma_{23}CMe^{-i\phi}\langle A_{13}\rangle,$$

$$\langle\dot{A}_{32}\rangle=-\frac{1}{2}[\gamma_2(1+2C+2CN)$$
$$+\gamma_3(1+2C+4CN)+i\Delta]\langle A_{32}\rangle$$
$$-2\gamma_3 CMe^{-i\phi}\langle A_{23}\rangle+2\gamma_{23}CN\langle A_{21}\rangle$$
$$+\gamma_{23}CMe^{-i\phi}\langle A_{12}\rangle,$$

$$\langle\dot{A}_{21}\rangle=-\frac{1}{2}[\gamma_2(1+2C+4CN)+2\gamma_3 CN-i\Delta]\langle A_{21}\rangle$$
$$-2\gamma_2 CMe^{-i\phi}\langle A_{12}\rangle+\gamma_{23}(1+2C+2CN)\langle A_{32}\rangle$$
$$+\gamma_{23}CMe^{-i\phi}\langle A_{23}\rangle,$$

$$\langle\dot{A}_{31}\rangle=-\frac{1}{2}[\gamma_3(1+2C+2CN)+2\gamma_2 CN]\langle A_{31}\rangle$$
$$+\gamma_{23}CMe^{-i\phi}(\langle A_{11}\rangle-2\langle A_{22}\rangle+\langle A_{33}\rangle). \quad (16)$$

If we define the following scaled parameters,

$$\beta=\frac{2C}{1+2C},\quad N_c=\beta N,\quad M_c=\beta M, \quad (17)$$

$$\Gamma_l=\gamma_l(1+2C),\quad \xi_l=\Gamma_l M_c e^{i\phi},\quad (l=2,\,3,\,23), \quad (18)$$

then the modified Bloch equations in the bad-cavity limit can be expressed as

$$\langle\dot{A}_{33}\rangle=\Gamma_3 N_c\langle A_{22}\rangle-\Gamma_3(1+N_c)\langle A_{33}\rangle+\frac{1}{2}\xi_{23}\langle A_{31}\rangle$$
$$+\frac{1}{2}\xi_{23}^*\langle A_{13}\rangle,$$

$$\langle\dot{A}_{22}\rangle=\Gamma_2 N_c\langle A_{11}\rangle-[\Gamma_2(1+N_c)+\Gamma_3 N_c]\langle A_{22}\rangle$$
$$+\Gamma_3(1+N_c)\langle A_{33}\rangle-\xi_{23}\langle A_{31}\rangle-\xi_{23}^*\langle A_{13}\rangle,$$

$$\langle\dot{A}_{32}\rangle=-\frac{1}{2}[\Gamma_2(1+N_c)+\Gamma_3(1+2N_c)+i\Delta]\langle A_{32}\rangle$$
$$-\xi_3^*\langle A_{23}\rangle+\Gamma_{23}N_c\langle A_{21}\rangle+\frac{1}{2}\xi_{23}^*\langle A_{12}\rangle,$$

$$\langle\dot{A}_{21}\rangle=-\frac{1}{2}[\Gamma_2(1+2N_c)+\Gamma_3 N_c-i\Delta]\langle A_{21}\rangle$$
$$-\xi_2^*\langle A_{12}\rangle+\Gamma_{23}(1+N_c)\langle A_{32}\rangle+\frac{1}{2}\xi_{23}^*\langle A_{23}\rangle,$$

$$\langle\dot{A}_{31}\rangle=-\frac{1}{2}[\Gamma_2 N_c+\Gamma_3(1+N_c)]\langle A_{31}\rangle+\frac{1}{2}\xi_{23}^*(\langle A_{11}\rangle$$
$$-2\langle A_{22}\rangle+\langle A_{33}\rangle). \quad (19)$$

The quantity Γ_l is the cavity enhanced spontaneous decay rate of the level $|l\rangle$, and N_c and M_c are the effective values of the squeezing parameters for the atom in the cavity environment. Equations (19) are formally identical to the optical Bloch equations in free space [12,14]. It is as though we were considering a free atom damped by a broadband squeezed vacuum but that the squeezing in the reservoir was now described by N_c and M_c. The only difference between the equations in the bad cavity and free space lies in the values of the parameters.

From the definitions of the parameters N_c and M_c (17), in the bad cavity we have

$$M_c=\beta M=\beta\sqrt{N(N+1)}=\sqrt{N_c(N_c+\beta)}$$
$$<\sqrt{N_c(N_c+1)}<\sqrt{N(N+1)} \quad (20)$$

so that the effect of the cavity is to degrade the strength of the two-photon correlations of the squeezed vacuum. Even if the squeezed field has perfect two-photon correlations in free space as we assume ($\eta=1$), within the cavity it necessarily behaves like a squeezed field with imperfect two-photon correlations, $\eta_c\equiv M_c/N_c<1$. Only in the limit $C\to\infty$, (i.e., $\beta\to1$) are the correlations the same strength in free space and in the bad cavity. Therefore, we must now interpret N_c and M_c as the *effective* squeezing parameters experienced by the atom in the bad-cavity limit.

The renormalization of the parameters has very important consequences. We shall see that the two-photon absorption rate has not only a term linearly dependent on the excitation intensity, but also a quadratically dependent term, as observed experimentally [22]. The latter is due to the reduction of the two-photon correlation strength in the injected squeezed vacuum inside the bad cavity. The extremely narrow spectral features in the two-photon fluorescence spectrum are also due to this reduction: they do not arise in the corresponding free space situation with $\eta=1$.

III. TWO-PHOTON EXCITATION RATE

Experimentally, the two-photon excitation probability is usually measured by recording the atomic fluorescence intensity from the levels $|3\rangle$ to $|2\rangle$, that is, by measuring the upper-level population $\langle A_{33}\rangle$ [22]. For our model the steady-state populations of the excited and intermediate levels and the steady-state two-photon coherence are

$$\langle A_{33}\rangle=\frac{N_c^2(\Gamma_2 N_c+\Gamma_3 N_c+\Gamma_3)-M_c^2(\Gamma_2 N_c+\Gamma_3 N_c-\Gamma_2)}{(3N_c^2+3N_c+1-3M_c^2)(\Gamma_2 N_c+\Gamma_3 N_c+\Gamma_3)},$$

$$\langle A_{22}\rangle=\frac{N_c(1+N_c)-M_c^2}{3N_c^2+3N_c+1-3M_c^2},$$

$$\langle A_{13}\rangle=\frac{\Gamma_{23}M_c e^{i\phi}}{(3N_c^2+3N_c+1-3M_c^2)(\Gamma_2 N_c+\Gamma_3 N_c+\Gamma_3)}. \qquad (21)$$

The steady-state, single-photon coherences are zero: $\langle A_{12}\rangle=\langle A_{23}\rangle=0$. The nonzero two-photon coherence $\langle A_{13}\rangle$, proportional to the two-photon correlation strength of the injected squeezed vacuum inside the bad cavity, reflects the fact that the correlations in the excitation field induce a stationary correlation between levels $|1\rangle$ and $|3\rangle$. Note that $\langle A_{13}\rangle$ is also proportional to Γ_{23}.

In free space, $\beta=1$, we see that the population in the intermediate level is zero: $\langle A_{22}\rangle=0$, and the population transfers from the ground level to the upper level via a single-step, two-photon process. That is, one photon of the pairs promotes the atom from the ground level to the virtual intermediate level, while its twin immediately (in a time less than the virtual level lifetime) completes the two-photon transition, due to the highly correlated photon pairs in the squeezed vacuum. The resulting two-photon absorption rate (proportional to the upper level population $\langle A_{33}\rangle$) is linearly dependent on the excitation intensity N [11,12] when $M=\sqrt{N(N+1)}$.

However, in the bad-cavity situation, $\beta<1$ so that $M_c<\sqrt{N_c(N_c+1)}$, $\langle A_{22}\rangle\neq 0$ even in the case when a perfectly correlated squeezed vacuum is injected into the cavity. The effective degree of correlation η_c is now less than 1, which means that some of the photon pairs in the squeezed field are not correlated, due to the cavity effect. The corresponding two-photon excitation is a mixed, two-photon process of one-step transitions and two-step transitions. Therefore, the two-step transition process makes a quadratic intensity-dependent contribution to the two-photon absorption rate (for low excitation strengths), as shown by Mollow [27].

We may rewrite the expression for $\langle A_{33}\rangle$ in Eq. (21) in the abbreviated form

$$\langle A_{33}\rangle=\frac{\beta N_c+(1-\beta)(\alpha+1)N_c^2}{(\alpha+\alpha N_c+N_c)[3N_c(1-\beta)+1]}, \qquad (22)$$

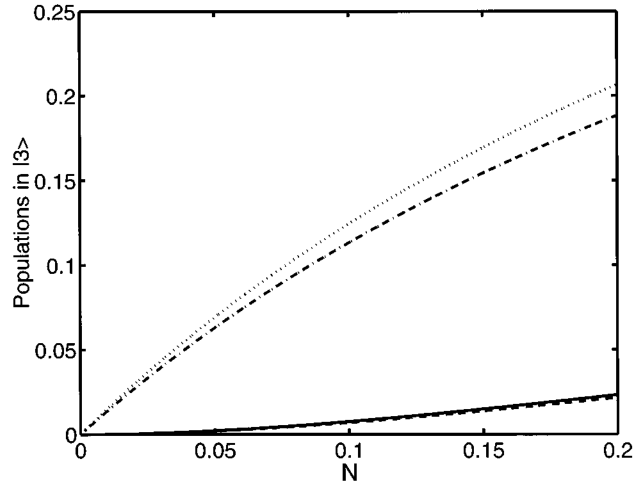

FIG. 2. Two-photon transition rate (proportional to the population in the upper level) as a function of N in regions of weak excitation, for $\alpha=0.64$, and in the bad-cavity limit with $C=10$, $M=0$ (dashed line) and $C=10$, $M^2=N(N+1)$ (dash-dotted line), and in the free space with $M=0$ (solid line) and $M^2=N(N+1)$ (dotted line).

with $\alpha=\gamma_3/\gamma_2=\Gamma_3/\Gamma_2$. The population consists of not only a linear dependence term on the squeezed photon number N_c inside the cavity, but also a quadratic dependence term on N_c, which is in excellent agreement with the experimental result [22]. The former is always associated with the factor β, the latter with $(1-\beta)$. This result may be alternatively understood as follows. As shown above, the two-photon correlation strength of the injected squeezed vacuum is reduced due to the cavity effect. We may imagine separating the squeezed vacuum inside the cavity into a squeezed (nonclassical-field) part, indicated by the factor β, and a nonsqueezed (classical-field) part, presented by $(1-\beta)$. The nonclassical-field excitation (consisting of perfectly correlated photon pairs) contributes a linear component to the rate, whilst the classical-field excitation (consisting of uncorrelated photons) contributes a quadratic component. The presence of the term quadratic in N_c in Eq. (22) is entirely due to the effectively imperfect correlation of the squeezed photon pairs in the bad cavity.

We display the upper-level population $\langle A_{33}\rangle$ in free space and in the bad-cavity configuration ($C=10$) for weak excitation in Fig. 2, where the value $\alpha=0.64$ is set according to the linewidths of the intermediate level ($6P_{3/2},F'=5$) and the upper level ($6D_{5/2},F''=6$) of the cesium atom employed in recent experiments [22,26]. One finds that for thermal (classical-) field excitation ($M=0$), the two-photon transition rates in free space and in a cavity are almost identical, whilst for squeezed (nonclassical) light excitation, the rates are significantly different, with the rate in free space being greater than that in a cavity (in the bad-cavity limit). However, the rate of nonclassical excitation is larger than that of classical excitation.

In general, the upper-level population $\langle A_{33}\rangle$ is also determined by the ratio α of the upper- and intermediate-level linewidths and the effective atomic cooperativity parameter C. In the limit $\alpha\ll 1$ (i.e., $\gamma_3\ll\gamma_2$), the upper-state population (22) reduces approximately to

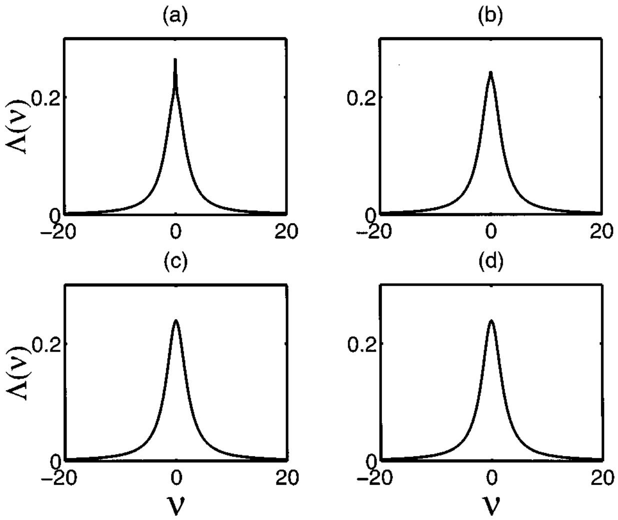

FIG. 3. Fluorescence emission spectrum from the upper level to the intermediate one, with $\Delta=0$, $\alpha=0.64$, $N=2$ and various values of C: (a) $C=10$, (b) $C=100$, (c) $C=1000$, and (d) in the free space. We assume that a perfect squeezed vacuum is injected into the cavity in the following graphs.

$$\langle A_{33}\rangle \simeq \frac{\beta+\beta(1-\beta)(\alpha+1)N}{3N\beta(1-\beta)+1}. \tag{23}$$

In free space $\langle A_{33}\rangle \simeq 1$, i.e., the atom is highly populated in the excited level $|3\rangle$ due to nonclassical two-photon excitation by a squeezed light. However, in the cavity configuration, $\beta<1$, the two-photon population inversion is reduced depending upon the atomic cooperativity parameter C. When the parameter C is very small, which corresponds to the field's inside cavity being more chaotic, the population (23) in the upper level is approximately reduced to $\frac{1}{3}$. This is the value found in two-photon excitation by black-body radiation [14].

IV. TWO-PHOTON EXCITATION SPECTRUM

When the atom experiences a two-photon excitation, it will be excited to the upper level. The two-photon excitation spectrum is represented by the fluorescence emission spectrum from the upper level to the intermediate level, which can, in principle, be measured experimentally by monitoring the fluorescence from the decay of the excited level $|3\rangle$ to the intermediate level $|2\rangle$ at frequency ω_{32} as a function of a reference frequency scanning near ω_c [22,26]. The spectrum is proportional to the Fourier transform of the stationary average value of the two-time average of the atomic polarization operators involved [27]:

$$\Lambda(\nu)=\mathrm{Re}\int_0^\infty d\tau e^{-i\nu\tau}\langle A_{32}(\tau)A_{23}(0)\rangle=\mathrm{Re}\left[\frac{E(z)}{D(z)}\right]_{z=i\nu}, \tag{24}$$

where $\nu=\omega-\omega_c$ is the frequency measured by the cavity frequency ω_c. $E(z)/D(z)$ is the Laplace transform of the two-time correlation function $\langle A_{32}(\tau)A_{23}(0)\rangle$, which can be evaluated by invoking the quantum regression theorem [28] together with the optical Bloch equations (19), as

$$E(z)=[z+\Lambda_2^*-\Gamma_{23}(1+N_c)B_1+\xi_2B_2]\langle A_{33}\rangle - \left[\xi_3^*B_1-\Gamma_{23}N_cB_2-\frac{1}{2}\xi_{23}^*\right]\langle A_{13}\rangle,$$

$$D(z)=[z+\Lambda_2^*-\Gamma_{23}(1+N_c)B_1+\xi_2B_2][z+\Lambda_3+\xi_3^*A_1 - \Gamma_{23}N_cA_2]-\left[\xi_2A_2-\Gamma_{23}(1+N_c)A_1-\frac{1}{2}\xi_{23}\right] \times\left[\xi_3^*B_1-\Gamma_{23}N_cB_2-\frac{1}{2}\xi_{23}^*\right], \tag{25}$$

where

$$A_1=\frac{-\xi_3(z+\Lambda_2)+\frac{1}{2}\xi_{23}\Gamma_{23}(1+N_c)}{(z+\Lambda_2)(z+\Lambda_3^*)-\frac{1}{4}|\xi_{23}|^2},$$

$$A_2=\frac{\Gamma_{23}(1+N_c)(z+\Lambda_3^*)-\frac{1}{2}\xi_3\xi_{23}^*}{(z+\Lambda_2)(z+\Lambda_3^*)-\frac{1}{4}|\xi_{23}|^2},$$

$$B_1=\frac{\Gamma_{23}N_c(z+\Lambda_2)-\frac{1}{2}\xi_2^*\xi_{23}}{(z+\Lambda_2)(z+\Lambda_3^*)-\frac{1}{4}|\xi_{23}|^2},$$

$$B_2=\frac{-\xi_2^*(z+\Lambda_3^*)+\frac{1}{2}\xi_{23}^*\Gamma_{23}N_c}{(z+\Lambda_2)(z+\Lambda_3^*)-\frac{1}{4}|\xi_{23}|^2},$$

$$\Lambda_2=\frac{1}{2}[\Gamma_2(1+2N_c)+\Gamma_3N_c-i\Delta],$$

$$\Lambda_3=\frac{1}{2}[\Gamma_2(1+N_c)+\Gamma_3(1+2N_c)+i\Delta]. \tag{26}$$

The fluorescence spectrum takes a complicated form. However, it is easy to see that it is independent of the squeezed phase.

We show the numerical results of the fluorescence emission spectrum $\Lambda(\nu)$ for $\Delta=0$, $\alpha=0.64$, $N=2$ and various cooperativity parameters C in Fig. 3. It is clearly seen that a significant narrow peak is superimposed on the broad resonance for $C=10$ in Fig. 3(a), while, as the cooperativity parameter increases, for instance, $C=100$ in Fig. 3(b), the height of the narrow peak is reduced. For large values of C the narrow peak disappears and only a broad component is exhibited; see, for example, $C=1000$ in the frame 3(c). The latter is almost identical to Fig. 3(d), the fluorescence spectrum in free space.

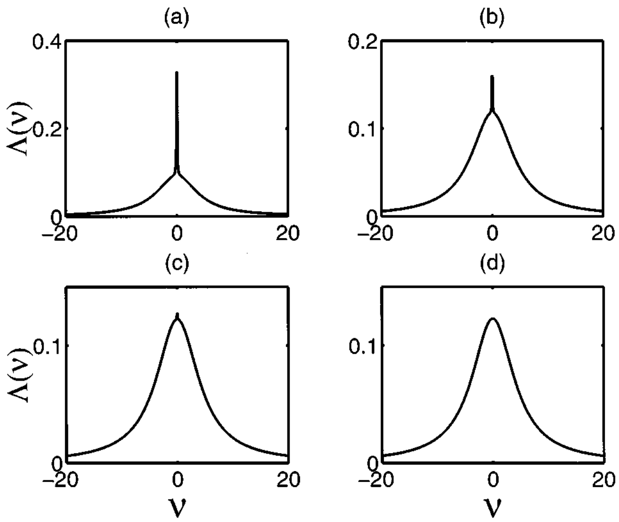

FIG. 4. Same as Fig. 3, but with $N=5$.

The additional narrow spectral feature is also dependent on the squeezed photon number N. In Fig. 4, where we set $N=5$ and keep the other parameters as in Fig. 3, we find that the narrow spectral feature is more pronounced than those in the previous figure. In the present case the narrow peak even can occur at $C=1000$. However, the background peak is wider than those for $N=2$. The sensitivity of the narrow spectral structure in the two-photon excitation spectrum to the squeezed photon number is shown more transparently in Fig. 5(a), where one sees that the larger the squeezed photon number, the more significant is the narrow peak profile for a given cooperativity parameter C. However, Fig. 5(b) exhibits that the narrow peak does not occur in the free space situation, no matter how large the squeezed photon number is taken.

All the above results are obtained assuming that a perfect squeezed vacuum is injected into the cavity. The distinctive narrow spectral feature in the two-photon excitation spectrum occurs in the bad-cavity limit, where the field inside the cavity consists of the nonclassical squeezed-field part and the classical thermal-field component. As shown above, the narrow spectral line does not occur in free space where the atom experiences only nonclassical two-photon excitation by the perfect squeezed vacuum. If we relax the condition that the injected squeezed vacuum is perfect, i.e., if we assume that η defined in Eq. (6) is less than 1, then the sharp peak can reappear even in the free space situation. For example, if we plot the spectra for same parameters as taken in the free space plot Fig. 5(b), but now assume $\eta=0.99$, we obtain a spectrum qualitatively similar to the bad-cavity result in Fig. 5(a) (where $\eta=1$). This is shown in Fig. 5(c). In the remainder of our plots we revert to the assumption that $\eta=1$.

In Fig. 6 we have plotted the fluorescence spectrum for different values of α. In the limits of $\alpha \ll 1$ ($\gamma_3 \ll \gamma_2$) or $\alpha \gg 1$ ($\gamma_3 \gg \gamma_2$) the narrow effects of the spectrum are not very evident: the extremely narrow spectral features can arise for $\alpha \sim 1$. Also, the fluorescence intensity (the area under the spectral curve) decreases as α increases. This is because the excited-state population is greater when $\alpha \ll 1$, and there are consequently more fluorescent photons emitted.

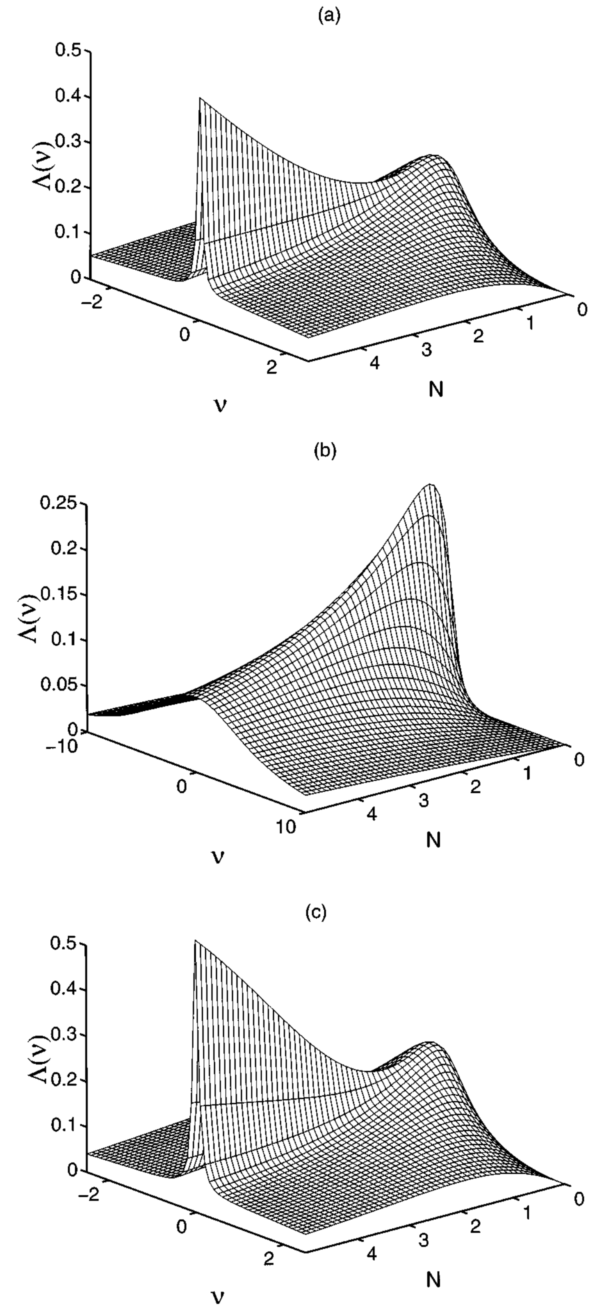

FIG. 5. Three-dimensional fluorescence spectrum for $\Delta=0$, $\alpha=1$, (a) in the bad cavity with $C=10$, and (b) in free space. In (c), we assume the same parameters as in Fig. 5(b), except that we take $\eta=0.99$ instead of $\eta=1$.

The effect of the one-photon detuning on the two-photon excitation spectrum is displayed in Fig. 7, where one finds that when the squeezed photon number $N \ll 1$, the fluorescence spectrum is centered around the Bohr frequency between the levels $|3\rangle$ and $|2\rangle$ of the fluorescent transition involved, whilst the spectrum is shifted toward line center (the cavity frequency) and broadened as N increases. For the larger values of N a remarkable narrow peak at the cavity frequency is superimposed on the wide background peak. In general, the spectrum is asymmetric. Comparing it with Fig. 5(a) we see that the detuning tends to broaden the narrow spectral feature.

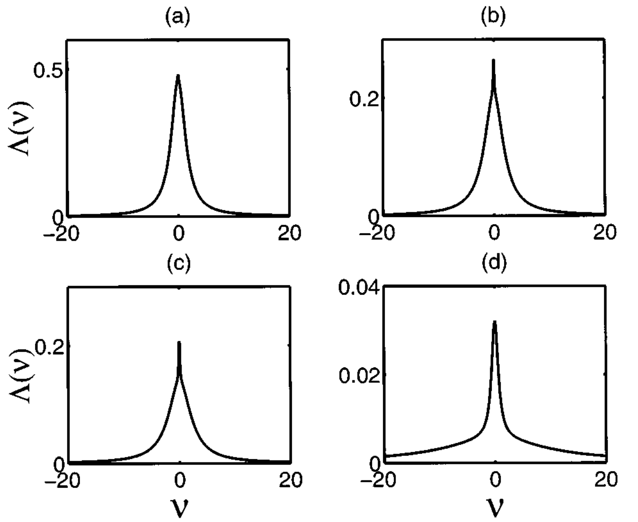

FIG. 6. Same as Fig. 3, but with $C=10$ and different values of α: (a) $\alpha=0.1$, (b) $\alpha=0.64$, (c) $\alpha=1$, and (d) $\alpha=10$.

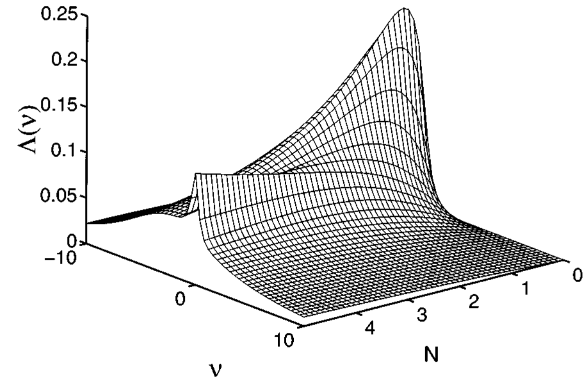

FIG. 7. Same as Fig. 5(a), but with the detuning $\Delta=5$.

Neither does the narrow spectral profile result from excitation with completely uncorrelated photons. In fact, if the atom undergoes two-photon excitation by an injected thermal field, then the fluorescence emission spectrum can be simplified to

$$\Lambda(\nu)=\frac{\langle A_{33}\rangle}{\Gamma_+-\Gamma_-}\left[-\frac{\Gamma_+(\Gamma_++\Lambda_2)}{\nu^2+\Gamma_+^2}+\frac{\Gamma_-(\Gamma_-+\Lambda_2)}{\nu^2+\Gamma_-^2}\right], \tag{27}$$

where

$$\langle A_{33}\rangle=\frac{N_c^2}{3N_c(1+N_c)+1},$$

$$\Gamma_\pm=-\frac{1}{2}(\Lambda_2+\Lambda_3)\pm\frac{1}{2}\sqrt{(\Lambda_2-\Lambda_3)^2+4\Gamma_{23}^2N_c(1+N_c)}. \tag{28}$$

The fluorescence emission spectrum is composed of two Lorentzians located at line center with linewidths $2\Gamma_\pm$. The spectrum is always very broad: no narrow spectral features are exhibited in either the free space or cavity situations.

We should emphasize that while some two-photon correlations are necessary, they do not have to be of nonclassical magnitude. Thus if we assume $M=N$, the maximum value permitted for a field that has a classical analog, it is still possible under appropriate conditions to observe the narrow line at the center of the spectrum, although it is much reduced in magnitude.

The significant spectral narrowing that occurs in the bad-cavity limit may be qualitatively understood in terms of quantum interference. Due to the mixing of the nonclassical two-photon correlated field and the black-body radiation field inside the cavity, the atom experiences two kinds of two-photon excitations: one is by the nonclassical, two-photon correlated field, which is a single-step excitation process involving the simultaneous absorption of two correlated photons, whilst the other is by the black-body radiation field, which is a two-step excitation requiring the separate absorption of two single photons. The quantum interference between these distinct excitation processes may give rise to the narrow spectral feature in the two-photon fluorescence spectrum under certain conditions. It is more significant for large squeezed photon numbers.

V. SUMMARY

We have studied the nonclassical two-photon excitation of a Ξ-type atom inside an optical cavity by an injected squeezed vacuum through the input-output cavity mirror. In the bad-cavity limit, the atomic excitation is composed of a one-step, two-photon absorption process and a two-step process involving the absorption of a single photon at each step. As a result, the steady-state two-photon transition rate (proportional to the upper-level population) consists of a linear and a quadratic dependence on the squeezed photon number, which is in agreement with experimental observation [22]. The rate is also sensitive to the ratio of the decay constants of the two excited levels. In the circumstance where $\gamma_3 \ll \gamma_2$, the upper atomic level is almost fully populated in the free space situation, while the two-photon population inversion is reduced inside the cavity to a degree that depends on the cooperativity parameter. For very small values of C, the population is reduced to $\frac{1}{3}$, reminiscent of thermal-field excitation.

We have also calculated the fluorescence emission spectrum of such an atomic system under two-photon excitation by squeezed light inside a cavity. We have found that a distinctive narrow spectral feature occurs in the bad-cavity limit for certain values of the parameters. The larger the squeezed photon number, the more pronounced is the narrow spectral feature. The remarkable spectral narrowing is a result of the quantum interference between the single-step, two-photon excitation by the nonclassical squeezed light and the two-step absorption of two single photons by the classical black-body radiation field. No spectral narrowing effect takes place in the cases of $\beta \ll 1$ or $\beta \simeq 1$. The former reduces to a cavity configuration with injected black-body radiation where the two-step excitation process is dominant, whilst the latter corresponds to the free space case where the single-step, two-photon excitation process is dominant.

It is worth pointing out that the system considered here is somewhat in accordance with the experimental configuration set up by Georgiades *et al.* [22]. We have chosen the squeezed photon number injected to be a little larger than those that they have used in the experiment, in order to make the extremely spectral narrow feature more pronounced. However, these features should be experimentally accessible. Whilst we have assumed a broadband squeezed vacuum in these calculations, it would not be surprising if the effects we have considered persist, or are even enhanced, in the narrow-band case, since they depend upon the two-photon correlations which characterize the squeezed vacuum being less than perfect.

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