Two-photon absorption and nonclassical states of light

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We investigate the dynamical evolution of nonclassical states of light undergoing a two-photon absorption process. We consider two distinct cases of initial states, a squeezed coherent state and an eigenstate of the two-photon annihilation operator (a superposition of macroscopically distinct coherent states). We analyze the fluctuations in the photon-number operator and in the quadrature components of the field. Whereas one-photon linear damping rapidly destroys quantum features such as squeezing, we demonstrate that substantial coherence is retained when such light interacts with a two-photon-absorbing reservoir. This surviving coherence is responsible for the preservation of squeezing in the steady state despite the effect of dissipation. We relate the origin of squeezing of initially unsqueezed light interacting with two-photon absorbers with the squeezing generated by simple superposition states of light.

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I. INTRODUCTION

Quantum features of light are usually very sensitive to dissipation. Squeezing, oscillations in the photon-number distribution, and interference effects are dramatically reduce when the physical system is coupled to a macroscopic environment [1]. Yet we know of methods to produce squeezed light through a two-photon absorption mechanism [2]. How is it that nonlinear damping, far from removing quantum features, actually creates them?

In this paper we analyze the effects of two-photon absorption. Because of the two-photon nature of the process, all the even-photon-number state components cascade down to the vacuum and the odd numbers to the one-photon state. Remarkably, the photon distribution during its decay becomes narrower than the Poisson distribution, revealing a specific quantum feature of the field generated by the damping mechanism. One might expect this to be the end of the matter, with a final state which is a statistical mixture of these two states $|0\rangle$ and $|1\rangle$, characteristic of equilibrium. But the most striking result is that this is not the case at all. As shown by Simaan and Loudon [3], there is an additional constant of motion leading to a nonvanishing degree of coherence between the two relevant states. It is precisely the existence of this coherence, preserved by the damping process, which is responsible for squeezing, and as we will show, this squeezing is identical to that described by Wódkiewicz *et al.* [4] in *superpositions* of quantum field states.

II. THE MODEL

We consider a model of two-photon absorption by a reservoir of two-level atoms from a single mode of the electromagnetic field. We follow the treatment of Tornau and Bach [5]. The reservoir consists of an ensemble of independent two-level atoms and is characterized by the operators $\hat{\Gamma}$ and $\hat{\Gamma}^\dagger$. The total Hamiltonian can be written as

$$\hat{H}=\hat{H}_0+\hat{H}_I \ , \tag{1}$$

$$\hat{H}_0=\hbar\omega\hat{a}^\dagger\hat{a}+\hat{H}_R \ , \tag{2}$$

$$\hat{H}_I=\hbar\lambda(\hat{a}^{\dagger 2}\hat{\Gamma}+\hat{a}^2\hat{\Gamma}^\dagger) \ , \tag{3}$$

$$\hat{H}_R=\hbar\sum_i\omega\hat{\sigma}_i^z \ , \tag{4}$$

$$\hat{\Gamma}=\sum_i\hat{\sigma}_i^- \ . \tag{5}$$

The boson operators $\hat{a},\hat{a}^\dagger$ and the two-level atom annihilation and creation operators $\hat{\sigma}^-,\hat{\sigma}^+$ satisfy the canonical commutation and anticommutation relations, respectively. We have assumed exact two-photon resonance between the atoms and the field. The atom-field density operator $\hat{\chi}$ satisfies the Liouville equation of motion:

$$i\hbar\frac{\partial\hat{\chi}}{\partial t}=[\hat{H}_I(t),\hat{\chi}(t)] \ , \tag{6}$$

where the operators are expressed in the interaction picture. We also assume that at the initial time the photon field and the atomic system are decoupled:

$$\hat{\chi}(0)=\hat{\rho}(0)\otimes\hat{\rho}_A(0) \ , \tag{7}$$

$$\hat{\rho}_A(0)=\prod_i\hat{\rho}_i(0) \ , \tag{8}$$

where $\hat{\rho}_i(0)$ is the thermal-equilibrium density operator for the ith atom. Using standard Born-Markov techniques [6], we obtain the following master equation for the radiation field:

$$\frac{\partial\hat{\rho}}{\partial t}=\mathcal{L}_1\hat{\rho}+\mathcal{L}_2\hat{\rho} \ , \tag{9}$$

where

$$\mathcal{L}_1\hat{\rho}=\kappa_1([\hat{a}^2\hat{\rho},\hat{a}^{\dagger 2}]+[\hat{a}^2,\hat{\rho}\hat{a}^{\dagger 2}]) \ , \tag{10}$$

and

$$\mathcal{L}_2\hat{\rho}=\kappa_2([\hat{a}^{\dagger 2}\hat{\rho},\hat{a}^2]+[\hat{a}^{\dagger 2},\hat{\rho}\hat{a}^2]) \ . \tag{11}$$

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The two Liouville operators $\mathcal{L}_1\hat{\rho}$ and $\mathcal{L}_2\hat{\rho}$ describe, respectively, the absorption and the emission parts of the two-photon damping process. In this paper we consider only the absorption part, i.e., we suppose the reservoir is at zero temperature ($\kappa_2=0$). We may show then easily from (9) that

$$\frac{\partial\langle\hat{a}\rangle}{\partial t}=-2\kappa_1\langle\hat{a}^\dagger\hat{a}^2\rangle\ , \tag{12}$$

$$\frac{\partial\langle\hat{a}^\dagger\hat{a}\rangle}{\partial t}=-4\kappa_1\langle\hat{a}^{\dagger 2}\hat{a}^2\rangle\ . \tag{13}$$

We see from these two equations that the rate of change of each moment of the field operator depends on the next normally ordered higher moment, leading to an infinite set of coupled equations. This makes the evolution highly dependent on the statistical properties of the field. Several authors have presented analytical solutions to this problem using different techniques such as Laplace transform [7], density-matrix approach [8], stochastic Langevin equation [9], and more extensively a generating-function method [10]. Explicit calculations have been carried out for coherent and thermal fields. They have shown that the field loses its classical features through two-photon absorption. Obviously, a nonlinear absorber removes preferentially the large amplitude fluctuations and reduces the amplitude noise of the field. The field second-order coherence falls below unity and the fluctuations in the quadratures below the vacuum level for appropriate interaction times. It is the purpose of this paper to provide a transparent physical explanation of these effects, and to show how these results are modified when the *initial* state of the field is already nonclassical, for example, one prepared as a squeezed coherent field or as a macroscopic superposition of two coherent states out of phase. These nonclassical states display initially reduced fluctuations. We show the competition between the squeezing present initially and the squeezing effect of the two-photon absorption process. We may regard this as the first step in understanding the stability of nonclassical field states such as squeezed light and Schrödinger-cat-like states in an environment in which two-photon absorption is the dominant loss mechanism. For example, in a semiconductor whose band gap is substantially greater than the energy of a single photon such effects may be of significance. Indeed, quantum well structures of this kind have been proposed as *sources* of squeezed light through their large parametric susceptibilities. We will discuss elsewhere the combined effect of the parametric coupling and two-photon losses within a single system as a possible scheme to produce eigenstates of the two-photon annihilation operator. The paper is organized as follows: in Sec. III we briefly review the solution given by Simaan and Loudon [3] for the diagonal and off-diagonal density-matrix elements, in Sec. IV we analyze the fluctuations in the photon-number operator, and in Sec. V those of the quadrature components of the field. In both cases, we summarize first the known coherent-state results for comparison purposes.

III. GENERAL SOLUTION OF THE MASTER EQUATION

We solve the master equation (9) at zero temperature with $\kappa_2=0$ using the generating-function approach developed by Simaan and Loudon [3]. We calculate the matrix elements of the field density operator between Fock states $\langle n|$ and $|n+\mu\rangle$ ($\mu=0,1,2,\dots$) and define a normalized time $\tau=2\kappa_1 t$. It is also convenient to define transformed matrix elements as follows:

$$\begin{aligned}\langle n|\hat{\rho}(\tau)|n+\mu\rangle&=\rho_{n,n+\mu}(\tau)\\&=[n!/(n+\mu)!]^{1/2}\psi_n(\mu,\tau)\ .\end{aligned} \tag{14}$$

The transformed matrix elements ψ_n satisfy then the equation

$$\begin{aligned}\frac{\partial\psi_n}{\partial\tau}&=(n+1)(n+2)\psi_{n+2}\\&\quad-[n(n-1)+\mu n+\tfrac{1}{2}\mu(\mu-2)]\psi_n\ .\end{aligned} \tag{15}$$

The system of differential equations (15) couples only elements with the same off-diagonality parameter μ, which means that the flow of change is propagating along parallels to the main diagonal. In particular, we obtain the equation of motion for the photon-number distribution $P_n\equiv\langle n|\hat{\rho}(\tau)|n\rangle$ when $\mu=0$:

$$\frac{\partial P_n}{\partial\tau}=(n+2)(n+1)P_{n+2}-n(n-1)P_n\ . \tag{16}$$

Because the two-photon absorption cannot empty the Fock states $|0\rangle$ and $|1\rangle$, we expect a nonvanishing value in the steady-state limit ($\tau\to\infty$) for $\rho_{0,0}$ and $\rho_{1,1}$:

$$\rho_{0,0}(\infty)=\sum_{n=0}^{\infty}P_{2n}(\tau)=\sum_{n=0}^{\infty}P_{2n}(0)\equiv\gamma_0\ , \tag{17}$$

$$\rho_{1,1}(\infty)=\sum_{n=0}^{\infty}P_{2n+1}(\tau)=\sum_{n=0}^{\infty}P_{2n+1}(0)\equiv\gamma_1\ . \tag{18}$$

The constants of motion γ_0 and γ_1 are related only through the normalization relation

$$\gamma_0+\gamma_1=1\ . \tag{19}$$

Dissipative interactions of the above kind might be expected to generate merely a statistical mixture of the one-photon and vacuum state with no interesting statistical properties. Surprisingly, this is not the case. Simaan and Loudon have pointed out the existence of an additional constant of motion for the elements on the first off-diagonal, leading to a non-vanishing value in the steady state for $\rho_{0,1}$:

$$\rho_{0,1}(\infty)=\psi_0(1,\infty)=\sum_{n=0}^{\infty}\frac{(2n-1)!!}{(2n)!!}\psi_{2n}(1,0)\equiv\gamma_{01}\ . \tag{20}$$

We observe indeed in the master equation (9) that the second normally ordered moment of the number operator acts on both sides of the density operator, allowing coherence between the one-photon and the vacuum state to survive in the steady-state solution. Thus the steady-state density matrix reduces to a 2×2 nondiagonal matrix defined by the three constants of motion γ_0, γ_1, and γ_{01}, which depend only on the initial conditions:

$$\hat{\rho}(\infty)=\begin{bmatrix}\gamma_0 & \gamma_{01}\\ \gamma_{01} & \gamma_1\end{bmatrix} . \tag{21}$$

The positivity of the density operator gives a relation between the three constants:

$$\gamma_0\gamma_1-\gamma_{01}^2\geq 0 , \tag{22}$$

where the lower bound is reached for a pure state. This remarkable result can be generalized to an arbitrary k-photon absorption process. In this case the only nonvanishing elements $\psi_n(\mu)$ in the steady state are such that $(n+\mu)!/(n+\mu-k)!=n!/(n-k)!$. They form a $k\times k$ nondiagonal density matrix. This shows that nonlinear dissipative processes preserve a certain degree of coherence. The actual value of γ_{01} depends only on the initial state. We see from (20) that for an initial thermal field $\gamma_{01}=0$. In Fig. 1 we plot the off-diagonal constant of motion as a function of the square of the displacement parameter for initial coherent and squeezed states defined in (43). In the limit of a large mean photon number, the amount of preserved coherence saturates to a common value for both fields.

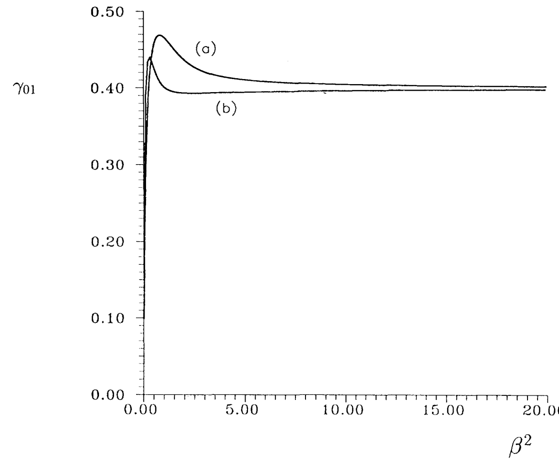

FIG. 1. Off-diagonal constant of motion γ_{01} for initial (a) coherent states, (b) squeezed states, vs the square of the displacement parameter. For (b) the squeezing parameter $r=0.5$. The maximal degree of asymptotic coherence is smaller for squeezed states and is independent of the squeezing parameter when the coherent component is dominant and tends to the value of 0.4.

In order to solve the system of coupled differential equations (15) we define a generating function by

$$\mathcal{G}(x,\mu,\tau)=\sum_{n=0}^{\infty}x^n\psi_n(\mu,\tau) , \tag{23}$$

where $|x|\leq 1$. The inverse transform is given by

$$\psi_n(\mu,\tau)=(n!)^{-1}\frac{\partial^n\mathcal{G}}{\partial x^n}\bigg|_{x=0} . \tag{24}$$

Multiplying Eq. (15) by x^n and summing over n, we obtain the following equation for the generating function:

$$\frac{\partial\mathcal{G}}{\partial\tau}=(1-x^2)\left[\frac{\partial^2\mathcal{G}}{\partial x^2}\right]-\mu x\left[\frac{\partial\mathcal{G}}{\partial x}\right]-\frac{1}{2}\mu(\mu-1)\mathcal{G} . \tag{25}$$

The infinite set (15) of coupled equations for $\psi_n(\mu,\tau)$ reduces to a single linear differential equation for the generating function. The two-photon-order process leads to a second-order equation. We solve this equation by separation of variables and write the generating function in a factorized expansion form as

$$\mathcal{G}(x,\mu,\tau)=\sum_{k=0}^{\infty}A_k^\sigma F_k^\sigma(x)\exp(-\lambda_k\tau) . \tag{26}$$

The set of functions $F_k^\sigma(x)$ then satisfies the differential equation for the Gegenbauer polynomials [11]:

$$F_k^\sigma(x)=C_k^\sigma(x) , \tag{27}$$

with

$$\sigma=\tfrac{1}{2}(\mu-1) , \tag{28}$$

and

$$\lambda_k=k(k+\mu-1)+\tfrac{1}{2}\mu(\mu-1) . \tag{29}$$

The coefficients A_k^σ of the superposition are determined by the initial conditions. Using the orthogonality relations for the Gegenbauer polynomials we can express them as

$$A_k^\sigma=\frac{(k+\sigma)\Gamma(\sigma)}{2^k\pi^{/2}}\sum_{\substack{m=k\\(m-k\text{ even})}}^{\infty}\frac{m!\Gamma(\frac{1}{2}m-\frac{1}{2}k+\frac{1}{2})}{(m-k)!\Gamma(\frac{1}{2}m+\frac{1}{2}k+\sigma+1)}\times\psi_m(\mu,0),\quad \sigma\neq 0 . \tag{30}$$

Using Eq. (24), we find the following corresponding expression for the normalized density-matrix elements:

$$\psi_n(\mu,\tau)=\sum_{\substack{k=n\\(k-n\text{ even})}}^{\infty}\frac{(-1)^{(1/2)k-(1/2)n}2^n\Gamma(\frac{1}{2}k+\frac{1}{2}n+\sigma)}{n!\Gamma(\sigma)\Gamma(\frac{1}{2}k-\frac{1}{2}n+1)}A_k^\sigma\exp(-\lambda_k\tau),\quad \sigma\neq 0 . \tag{31}$$

The case $\sigma=0$ must be treated separately because the orthogonality relation used to derive (30) does not hold in that case. One way to overcome this problem is to use Chebyshev polynomials of the first kind which are related to the Gegenbauer polynomials of order zero by

$$C_n^0(x)=\frac{2}{n}T_n(x),\quad n\neq 0 . \tag{32}$$

The resulting expression for $\psi_n(1,\tau)$ is given by

$$\psi_n(1,\tau)=\sum_{\substack{k=n\\(k-n\text{ even})}}^{\infty}\frac{(-1)^{(1/2)k-(1/2)n}2^{n-1}k\,\Gamma(\frac{1}{2}k+\frac{1}{2}n)}{n!\,\Gamma(\frac{1}{2}k-\frac{1}{2}n+1)}B_k\exp(-k^2\tau)\ , \tag{33}$$

where

$$B_k=\sum_{\substack{m=k\\(m-k\text{ even})}}^{\infty}\frac{m!}{2^{m-\delta(k)}(\frac{1}{2}m+\frac{1}{2}k)!(\frac{1}{2}m-\frac{1}{2}k)!}\times\psi_m(1,0)\ , \tag{34}$$

and

$$\delta(k)=\begin{cases}1 & \text{if } k>0\\ 0 & \text{if } k=0\ .\end{cases} \tag{35}$$

Equations (30), (31), (33), and (34) provide a complete time evolution for the transformed matrix elements $\psi_n(\mu,\tau)$ which are related to the matrix elements $\rho_{n,n+\mu}(\tau)$ through the relation (14). The exponential decay of the series (31) and (33) provides a cutoff in the expansion. In particular, we note that in the steady-state limit the only contribution to the two series arises when $\lambda_k=0$. From Eq. (29) we see that this condition is satisfied only for $\mu=0$ when $k=0$ or 1 and for $\mu=1$ when $k=0$. The first solution leads to the two constants of motion (17) and (18) for the diagonal elements:

$$A_0^{-1/2}=\gamma_0\ , \tag{36}$$

$$A_1^{-1/2}=-\gamma_1\ , \tag{37}$$

while the second solution gives the off-diagonal constant of motion (20):

$$B_0=\gamma_{01}\ . \tag{38}$$

The existence of these constants of motion induces a parity selection rule in the expression of the relevant series.

So far we have established general equations which govern the two-photon absorption. We have seen that the nonlinearity of the process makes the evolution of field observables highly dependent on the field statistics. We next first briefly summarize the results for a coherent state and then investigate the cases of an initial squeezed state and of eigenstates of the two-photon annihilation operator, namely, the even and odd coherent states. We present numerical evaluations, and compare a squeezed vacuum field to a squeezed field containing a coherent amplitude, at equal mean energy for the number fluctuations and at equal squeezing for the quadrature fluctuations. We start with an analysis of the fluctuations in the photon-number operator in the next section.

IV. FLUCTUATIONS IN THE NUMBER OPERATOR

In this section we analyze the evolution of the fluctuations of the diagonal part of the density operator. From the equation of motion for the photon-number distribution (16) we note two opposite contributions to the change of P_n: one proportional to $n(n-1)$ if the field contains n photons and the other proportional to $(n+2)(n+1)$ if the field contains $(n+2)$ photons. The two-photon absorption nonlinearity appears through the quadratic dependence on n. The rate of change for the first moment of the number operator is easily obtained from (16):

$$\frac{\partial\langle\hat{n}\rangle}{\partial\tau}=-2\langle\hat{n}(\hat{n}-1)\rangle=-2\langle\hat{a}^{\dagger 2}\hat{a}^2\rangle\ . \tag{39}$$

In terms of the Glauber second-order correlation function [12] defined as

$$g^{(2)}(0)=\frac{\langle\hat{a}^{\dagger 2}\hat{a}^2\rangle}{\langle\hat{a}^{\dagger}\hat{a}\rangle^2}\ , \tag{40}$$

we can rewrite (39) as

$$\frac{\partial\langle\hat{n}\rangle}{\partial\tau}=-2\langle\hat{n}\rangle^2 g^{(2)}(0)\ . \tag{41}$$

This equation makes the absorption rate highly dependent on the field statistics. It is proportional to the second normally ordered correlation function of the field. In a similar way, the rate of change of the second moment depends on the next-order factorial moment:

$$\frac{\partial\langle\hat{n}^2\rangle}{\partial\tau}=-4[\langle\hat{n}(\hat{n}-1)(\hat{n}-2)\rangle+\langle\hat{n}(\hat{n}-1)\rangle]=-4(\langle\hat{a}^{\dagger 3}\hat{a}^3\rangle+\langle\hat{a}^{\dagger 2}\hat{a}^2\rangle)\ . \tag{42}$$

Thus, for equal initial mean photon numbers, a super-Poissonian field $[g^2(0)>1]$ is more rapidly absorbed than a sub-Poissonian one $[g^2(0)<1]$, at an enhanced rate in direct ratio of their second-order coherence functions. In particular the absorption rate for chaotic light is twice that for coherent light. This result has of course been known for some time [13].

A. Coherent-state results

If the initial state of the field is a coherent state, we find from the previous analysis that, surprisingly, the photon distribution becomes narrower than its initial Poisson width during the evolution. This means that the two-photon absorption influences the photon statistics to such a degree that it loses its classical characteristics. The variance relative to the mean photon number falls below unity. The more intense is the initial field, the faster is the absorption rate and the greater the deviation from Poissonian statistics. The second-order Glauber coherence function [12] decreases, causing a decrease in the absorption rate, and tends to zero in the steady state.

B. Squeezed coherent-state analysis

We now examine how these general principles apply to an initial squeezed coherent state. We follow the definition given by Stoler [14]. In this definition the vacuum field $|0\rangle$ is first squeezed through a squeezing operator $\hat{S}(r)$ then displaced through a displacement operator $\hat{D}(\beta)$. For simplicity we consider the case of squeezing along the displacement and take a real squeezing parameter. This initial field state can then be written as

$$|\psi\rangle=\hat{D}(\beta)\hat{S}(r)|0\rangle \; . \tag{43}$$

The squeezing operator is

$$\hat{S}(r)=\exp[\tfrac{1}{2}r(\hat{a}^2-\hat{a}^{\dagger 2})] \; , \tag{44}$$

and the operator $\hat{D}(\beta)$ is the Glauber displacement operator [12]:

$$\hat{D}(\beta)=\exp[\beta(\hat{a}^{\dagger}-\hat{a})] \; . \tag{45}$$

Expanding the squeezed state (43) in the number basis, we get the following expression for the transformed matrix elements defined in (14):

$$\begin{aligned}\psi_n(\mu,0)=&\frac{(\frac{1}{2}\tanh r)^{n+(1/2)\mu}}{n!\cosh r}\exp[-\tilde{\beta}^2+\tilde{\beta}^2\tanh(r)]\\ &\times H_n(\tilde{\beta}[2\sinh r\cosh r]^{-1/2})\\ &\times H_{n+\mu}(\tilde{\beta}[2\sinh r\cosh r]^{-1/2}) \; ,\end{aligned} \tag{46}$$

where $H_n(x)$ are the Hermite polynomials of order n [15], and

$$\tilde{\beta}=\beta(\sinh r+\cosh r) \; . \tag{47}$$

Squeezed states have been studied in detail [16] because of their unique nonclassical properties. Examples include oscillations in the photon-number distribution, sub-Poissonian and super-Poissonian statistics, and reduced quadrature fluctuations. In particular we obtain from (46) the number distribution for a squeezed vacuum state ($\beta=0$):

$$\begin{aligned}P_{\mathrm{SV}}(2n)&=\frac{(\frac{1}{2}\tanh r)^{2n}(2n)!}{(\cosh r)(n!)^2} \; ,\\ P_{\mathrm{SV}}(2n+1)&=0 \; .\end{aligned} \tag{48}$$

Equation (48) illustrates a maximal quantum interference: the squeezed vacuum contains only even photon numbers leading to the maximal value of 1 for γ_0 defined in (17). We show in Fig. 2 the time evolution of a squeezed vacuum photon distribution under the influence of the dissipative master equation (9). We see that the two-photon absorption preserves the two-photon nature of the field as expected. Indeed, the coefficients A_k^{σ} are given in Eq. (30) by a sum over even numbers for even indices k and by a sum over odd numbers for the odd indices. Similarly, the expression for the field matrix elements $\psi_n(\mu,\tau)$ in Eq. (31) contains the even coefficients A_k^{σ} for n even and the odd coefficients for n odd. Thus the initial oscillations in the photon distribution are retained. We calculate easily from Eq. (46) the initial mean photon number:

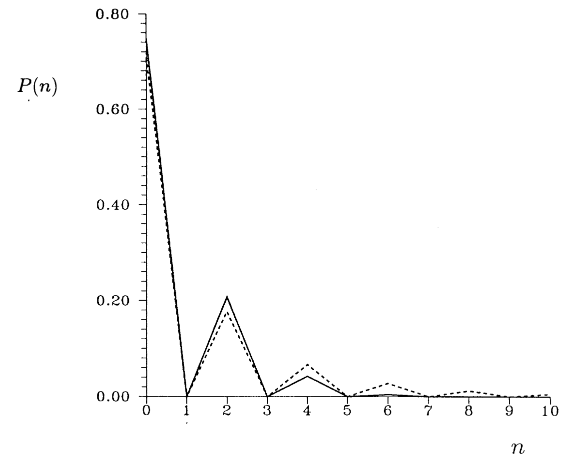

FIG. 2. Photon distribution for an initial squeezed vacuum with mean photon number equal to 1. The dashed and solid lines correspond, respectively, to the initial state ($\tau=0$) and the state after an interaction time $\tau=0.1$.

$$\langle\hat{n}(0)\rangle=\beta^2+\sinh^2 r \; , \tag{49}$$

and the fluctuation in the mean number:

$$\langle[\Delta\hat{n}(0)]^2\rangle=\beta^2\exp(-2r)+2\sinh^2 r\cosh^2 r \; . \tag{50}$$

We see from (49) and (50) that the squeezed vacuum always displays super-Poissonian statistics and satisfies

$$\langle(\Delta\hat{n})^2\rangle_{\mathrm{SV}}=2\langle\hat{n}\rangle_{\mathrm{SV}}(\langle\hat{n}\rangle_{\mathrm{SV}}+1) \; , \tag{51}$$

whereas sub-Poissonian statistics can be found for a squeezed coherent state, if the coherent component is dominant in the mean number. In Fig. 3 we compare the time evolution of the mean number for a squeezed vacu-

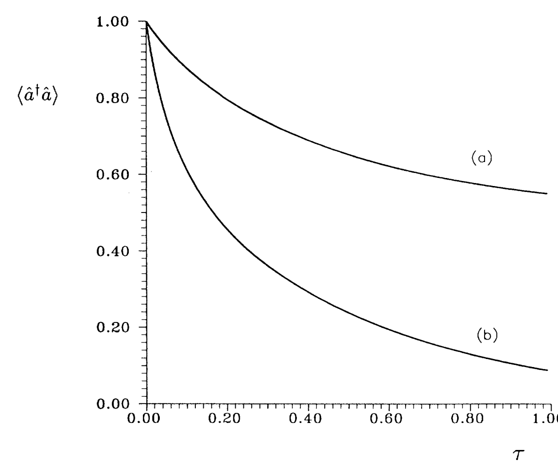

FIG. 3. Time evolution of the mean photon number. The initial value is equal to one. The curve (a) refers to a squeezed field with a coherent component equal to $\beta^2=0.8$, whereas the curve (b) refers to the squeezed vacuum. We observe a much faster absorption rate for the squeezed vacuum due to super-Poissonian statistics. The relative absorption rate is equal to the ratio of the second factorial moments. From Fig. 4, we see that it is equal to 5 at the initial time.

um and a squeezed state with a coherent amplitude. We observe a faster decay of the squeezed vacuum excitation number. Its absorption rate on a short-time scale is, according to Eq. (39), roughly five times higher than for the sub-Poissonian squeezed coherent field. The asymptotic value ($\tau \rightarrow \infty$) is equal to γ_1 defined in Eq. (18). Figure 4 shows the evolution of the second factorial moment which controls the mean number evolution. We note the enhanced absorption rate ratio due to third-order moment dependence. In Fig. 5 we display the variance relative to the mean number which measures the deviation from Poissonian statistics $[\langle(\Delta\hat{n})^2\rangle / \langle\hat{n}\rangle = 1]$. The squeezed vacuum remains super-Poissonian. The asymptotic relative variance is equal to two, although the steady state is the vacuum state. This is due to the unique nature of the squeezed vacuum photon distribution. Indeed, we approach the asymptotic state when the contribution of the two-photon Fock state tends to zero. Since the squeezed vacuum does not contain a one-photon state contribution, this leads to

$$\frac{\langle[\Delta\hat{n}(\infty)]^2\rangle}{\langle\hat{n}(\infty)\rangle} = \lim_{\rho_{22}\rightarrow 0} \frac{4\rho_{22}(1-\rho_{22})}{2\rho_{22}} = 2 \ . \tag{52}$$

The two-photon contribution cancels out in the asymptotic relative variance. In contrast, the asymptotic relative variance for the squeezed coherent field, due to a one-photon contribution, is equal to γ_0 defined in (17):

$$\frac{\langle[\Delta\hat{n}(\infty)]^2\rangle}{\langle\hat{n}(\infty)\rangle} = \frac{\gamma_1(1-\gamma_1)}{\gamma_1} = \gamma_0 \ . \tag{53}$$

In Fig. 6, the initial relative number fluctuation is equal to its value in the steady state. The intermediate evolution presents first a sharp increase followed by a smooth decrease on a long-time scale. We compare in Fig. 7 the relative variance for two fields which have the same degree of sub-Poissonicity initially. Their markedly different behavior during the interaction illustrates how

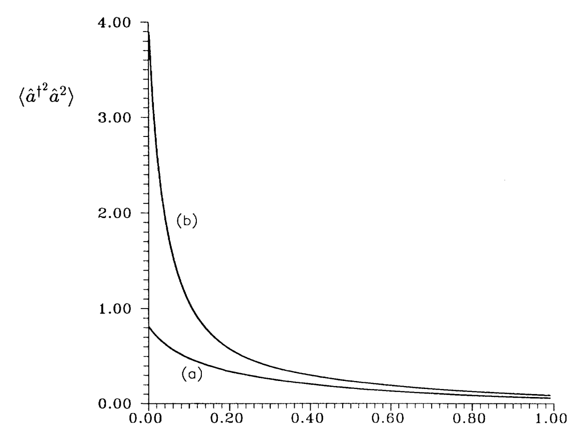

FIG. 4. Time evolution of the second factorial moment of the number operator, for the same initial condition as in Fig. 3. (a) Squeezed coherent field, (b) squeezed vacuum. Because the evolution of this quantity is related to the higher-order moments, the relative decay rate is initially enhanced.

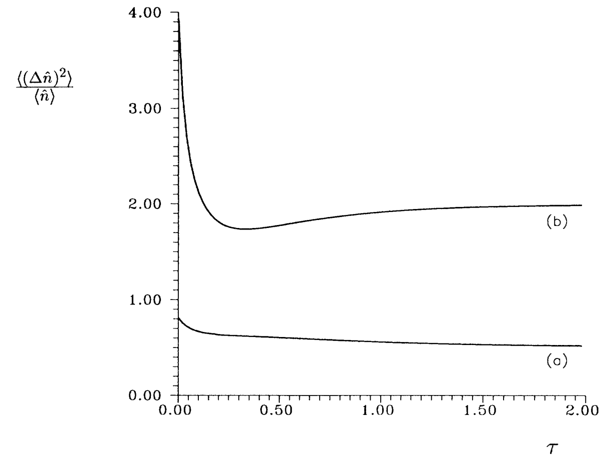

FIG. 5. Time evolution of the number variance relative to the mean number. The initial conditions are the same as in Fig. 3. In particular, the initial number variance is equal to the second factorial moment of the number operator since the mean number is equal to one. (a) refers to the squeezed coherent field. The asymptotic value is equal to γ_0 defined in (17). (b) Squeezed vacuum. The asymptotic value is equal to 2. We note the discontinuity between the two asymptotic values due to the particular nature of the squeezed vacuum state.

the dynamical evolution depends on the field statistics as mentioned earlier.

C. Even and odd coherent-state analysis

We now analyze the effect of two-photon absorption on another kind of nonclassical state of light, a superposition of two coherent states out of phase. We choose for sim-

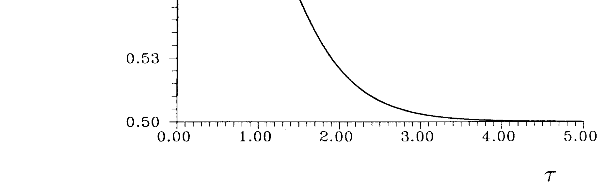

FIG. 6. Relative number variance for an initial sub-Poissonian field. The fluctuations in the photon number are initially reduced to half the mean number. The squeezing parameter r is equal to 0.4 and $\beta^2 = 5.45$. We note a rapid increase of the fluctuations on a short-time scale until a saturation time is reached followed by a smooth decrease in the fluctuations until the steady state is reached. During the overall evolution the field remains within the sub-Poissonian boundary.

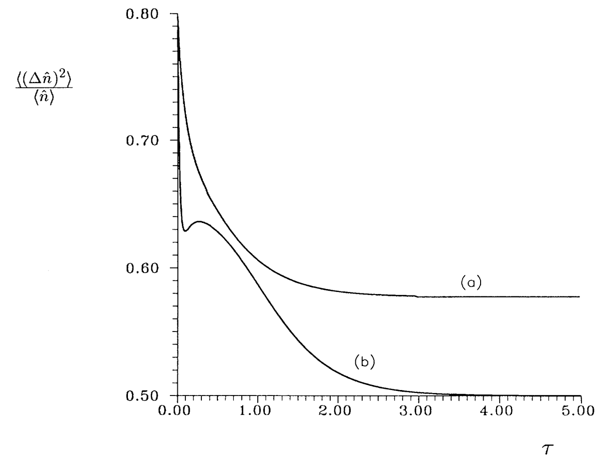

FIG. 7. Comparison between the relative number variance of two sub-Poissonian fields whose distributions have initially the same degree of sub-Poissonicity equal to 0.8. The curve (a) corresponds to a squeezing parameter $r=0.3$ and $\beta^2=0.512$ while (b) corresponds to $r=0.7$ and $\beta^2=2.44$. (b) displays a strong oscillation pattern in the photon distribution leading to a very different evolution on a short-time scale. The rate of change is first negative and very high in absolute value, then is positive during a significant time compared to the initial decay time, and ends negative relaxing slowly to the steady state. (a) shows only a negative rate of change. The photon distribution does not display an oscillation pattern.

plicity the two coherent states with real displacement parameter. Various methods have been proposed to generate these so-called even and odd coherent states [17]. These states have been investigated mainly by Janszky and Vinogradov [18] and Bužek, Vidiella-Barranco, and Knight [19]. They are described by

$$|\beta\rangle_e = N_e^{1/2}(|\beta\rangle + |-\beta\rangle) , \tag{54}$$

$$|\beta\rangle_o = N_o^{-1/2}(|\beta\rangle - |-\beta\rangle) , \tag{55}$$

where the normalization constants are given by

$$N_e = \{2[1+\exp(-2\beta^2)]\}^{-1} , \tag{56}$$

$$N_o = \{2[1-\exp(-2\beta^2)]\}^{-1} . \tag{57}$$

They are eigenstates of the square of the annihilation operator:

$$\hat{a}^2|\beta\rangle_{e,o} = \beta^2|\beta\rangle_{e,o} . \tag{58}$$

The interference between the coherent states $|\beta\rangle$ and $|-\beta\rangle$ is responsible for pairwise oscillations in the photon-number distribution. The even coherent state contains only even photon numbers, while the odd coherent state is a superposition of odd Fock states. Their photon statistics are, respectively,

$$P_e(2n) = 4N_e P_{\text{coh}}(2n) , \tag{59}$$

$$P_e(2n+1) = 0 , \tag{60}$$

$$P_o(2n+1) = 4N_o P_{\text{coh}}(2n+1) , \tag{61}$$

$$P_o(2n) = 0 , \tag{62}$$

where $P_{\text{coh}}(n)$ is the Poissonian distribution

$$P_{\text{coh}}(n) = \frac{\exp(-\beta^2)(\beta)^{2n}}{n!} . \tag{63}$$

In Fig. 8 we plot the photon statistics evolution for an initial even coherent state. Akin to the squeezed vacuum, the two-photon absorption perfectly preserves the oscillatory pattern of the initial distribution. The even coherent state exhibits super-Poissonian field statistics according to

$$\frac{\langle(\Delta\hat{n})^2\rangle_e}{\langle\hat{n}\rangle_e} = 1 + \frac{4\beta^2\exp(-2\beta^2)}{1-\exp(-4\beta^2)} > 1 , \tag{64}$$

whereas the odd coherent state displays sub-Poissonian statistics according to

$$\frac{\langle(\Delta\hat{n})^2\rangle_o}{\langle\hat{n}\rangle_o} = 2 - \langle(\Delta\hat{n})^2\rangle/\langle\hat{n}\rangle < 1 . \tag{65}$$

In Fig. 9 we plot the time evolution of the variance in the number operator relative to the mean number for both states. We observe deviation from super-Poissonian statistics for the even coherent state when the coherent amplitude is large enough. For $\beta=2$ the initial even coherent state is nearly Poissonian and at $\tau \simeq 0.13$ the field is 26% below the coherent level. It becomes then super-Poissonian again and relaxes to the vacuum state. Because the two-photon nature of the field is conserved during the damping, the steady-state value is 2, as for the squeezed vacuum. The odd coherent state presents a totally different behavior. The field remains sub-Poissonian. The relative number variance reaches its

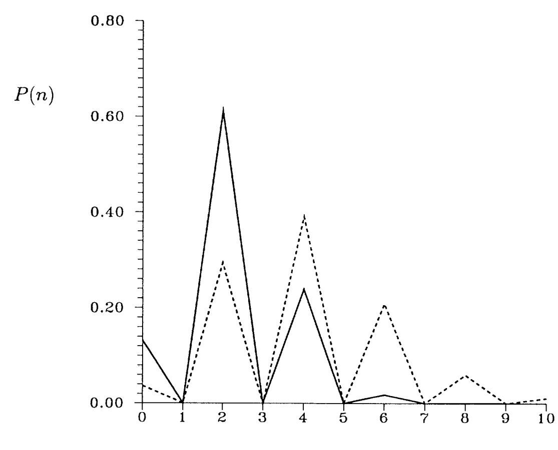

FIG. 8. Photon distribution evolution for an initial even coherent state with $\beta=2$. The dashed line represents the initial distribution and the solid line the distribution after an interaction time $\tau=0.1$ with the two-photon absorber. We observe perfect preservation of the pairwise oscillations as for the squeezed vacuum (Fig. 2) case. This shows that the dissipative mechanism does not remove strong quantum features of the initial field.

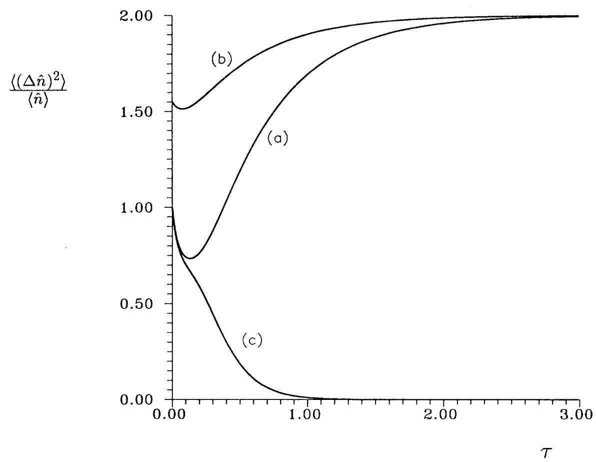

FIG. 9. Photon-number variance relative to mean number for (a) initial even coherent state with $\beta=2$, (b) initial even coherent state with $\beta=1$, and (c) initial odd coherent state with $\beta=2$. The two-photon dissipation reduces the fluctuations in the field operator below unity for even coherent states with large amplitude component as shown in (a). When the damping proceeds, the initial coherent state relaxes to the vacuum characterized by a super-Poissonian value 2. This was found also for an initial squeezed vacuum. The odd coherent state on the contrary keeps reduced fluctuations relaxing to a one-photon state with maximal sub-Poissonicity.

minimal possible value of zero. The steady state is a *pure* state containing one photon.

In the next section we analyze fluctuations in the quadratures of the field which involve off-diagonal elements of the density operator.

V. FLUCTUATIONS IN THE QUADRATURE OPERATORS

We decompose the annihilation and creation operators into their Hermitian components $\hat{X}_1$ and $\hat{X}_2$:

$$\hat{a}=\hat{X}_1+i\hat{X}_2 \ , \quad \hat{a}^\dagger=\hat{X}_1-i\hat{X}_2 \ . \tag{66}$$

The quadratures $\hat{X}_1,\hat{X}_2$ then satisfy the canonical commutation relation

$$[\hat{X}_1,\hat{X}_2]=i/2 \ , \tag{67}$$

leading to the Heisenberg uncertainty principle

$$\langle \Delta\hat{X}_1\rangle\langle \Delta\hat{X}_2\rangle \geq \tfrac{1}{4} \ . \tag{68}$$

A. Coherent-state results

It has been shown [2] that the two-photon absorption leads to the development of squeezing. Maximal squeezing increases in magnitude and occurs at shorter times as the mean number in the initial coherent field increases. The maximal value of squeezing which can be obtained is 33%. Squeezing is found to persist in the steady state only for initial mean photon numbers less than 1.

B. Squeezed coherent-state analysis

The initial squeezed state (46) is a minimum uncertainty squeezed state and satisfies

$$\langle \Delta\hat{X}_1(0)\rangle\langle \Delta\hat{X}_2(0)\rangle=\tfrac{1}{4} \ , \tag{69}$$

and the initial uncertainty in the two quadratures is given by

$$\langle \Delta\hat{X}_1(0)\rangle=\tfrac{1}{2}\exp(-r) \ , \quad \langle \Delta\hat{X}_2(0)\rangle=\tfrac{1}{2}\exp(r) \ . \tag{70}$$

These are independent of the coherent amplitude β. So, it is natural to compare fields which differ only through their squeezing parameter r. The variance of the quadrature operators is expressed as

$$\langle (\Delta\hat{X}_{1,2})^2\rangle=\tfrac{1}{4}+\tfrac{1}{2}(\langle \hat{a}^\dagger\hat{a}\rangle\pm\langle \hat{a}^2\rangle)\mp\langle \hat{a}\rangle^2 \ , \tag{71}$$

where we used the fact that we have real squeezing and displacement parameters. We calculate the averages using (31) and (33):

$$\langle \hat{a}^\dagger\hat{a}\rangle=\sum_{n=0}^{\infty} n\psi_n(0,\tau) \ , \quad \langle \hat{a}\rangle=\sum_{n=0}^{\infty}\psi_n(1,\tau) \ , \quad \langle \hat{a}^2\rangle=\sum_{n=0}^{\infty}\psi_n(2,\tau) \ . \tag{72}$$

In the steady state, the field density matrix is given by Eq. (21), hence we obtain

$$\langle \hat{a}^\dagger\hat{a}(\infty)\rangle=\gamma_1 \ , \quad \langle \hat{a}(\infty)\rangle=\gamma_{01} \ , \quad \langle \hat{a}^2(\infty)\rangle=0 \ . \tag{73}$$

The steady-state variances of the quadratures can then be written as

$$\langle [\Delta\hat{X}_{1,2}(\infty)]^2\rangle=\tfrac{1}{4}+\tfrac{1}{2}\gamma_1\mp\gamma_{01}^2 \ , \tag{74}$$

and squeezing in $\hat{X}_1$ is found if

$$\tfrac{1}{2}\gamma_1-\gamma_{01}^2<0 \ . \tag{75}$$

We plot in Fig. 10 the fluctuations in $\hat{X}_1$ in the steady state as a function of the square of the displacement parameter for initial coherent and squeezed states. Squeezing is found only for very small values of β^2. Maximal squeezing occurs for an initial coherent state. In Fig. 11 we plot the dynamical evolution of the uncertainty in $\hat{X}_1$ for three different values of displacement parameter. The squeezed vacuum shows a transient saturation region followed by a slow increase leading to the isotropic vacuum fluctuation level. We note the existence of squeezing in the steady state for small displacement parameters.

C. Link with superposition-state squeezing

Wódkiewicz *et al.* [4] have studied squeezing in superposition states of the field and found squeezing for a simi-

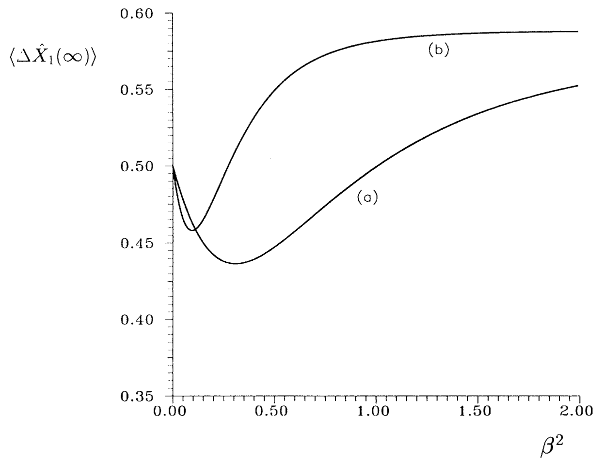

FIG. 10. Fluctuations in $\hat{X}_1$ in the steady state as a function of β^2, where β is the displacement parameter of the initial field. (a) initial coherent state, (b) initial squeezed state and we choose the squeezing parameter to be $r=0.5$. Maximal squeezing occurs for an initial coherent state. We find squeezing in the steady state when β^2 is less than 1 for the coherent-state case and this value is reduced when the initial field is squeezed.

lar range of parameters to those found above. When the coherent part of the field in our present case increases in size, extra noise is added in the steady state. In Fig. 12 we plot the uncertainty in the conjugate quadrature $\hat{X}_2$. We note the asymmetry in the evolution compared to the previous figure. Comparing the rate of change of both quadratures using Eq. (71), we see that noise introduced by the damping mechanism changes the fluctuations at a lower rate in the squeezed component $\hat{X}_1$. The three different fields in Fig. 12 display the same fluctuations

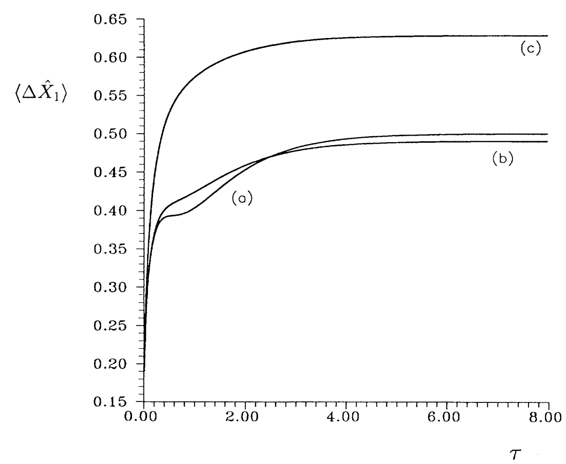

FIG. 11. Fluctuations in $\hat{X}_1$ for various displacement parameters. The squeezing parameter $r=1$. In (a) $\beta^2=0$, in (b) $\beta^2=0.01$, and in (c) $\beta^2=0.5$. We note that the squeezing persists in the steady state when the displacement parameter is not too large. There is a change in the dynamical evolution around $\tau \simeq 0.2$ for the three fields, but the squeezed vacuum state shows the most notable change, displaying a plateau which does not exist when the field contains a coherent component.

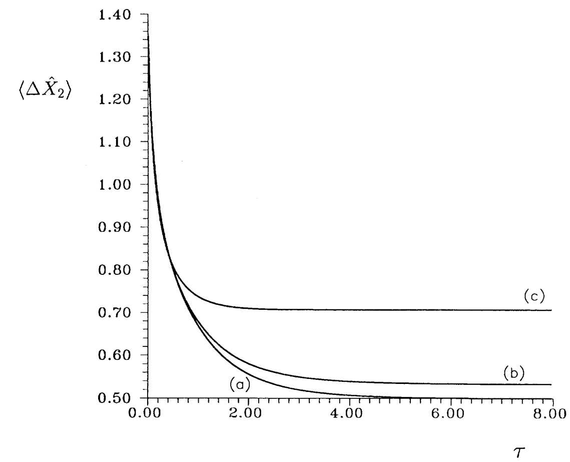

FIG. 12. Fluctuations in $\hat{X}_2$ for the same initial states as in Fig. 11. We note the greater rate of change in this quadrature.

over a longer time than in Fig. 11. Then they evolve apart, reaching their steady-state values given in Eq. (74). We show the product of the uncertainties in the canonical quadrature operators $\langle \Delta \hat{X}_1(0) \rangle \langle \Delta \hat{X}_2(0) \rangle$ in Fig. 13. We observe a general departure from the initial minimum uncertainty state value $\frac{1}{4}$. The loss of minimum uncertainty due to the fluctuations inherent in the irreversible absorption process illustrates the noninvariance of the squeezed states under two-photon absorption. Only the squeezed vacuum is reduced to a vacuum state which is trivially a minimum uncertainty state. Figures 14, 15, and 16 represent the same quantities as in Figs. 11, 12, and 13 respectively, but for different initial conditions. The squeezing is less intense and the squeezed vacuum state no longer presents the saturation region observed in Fig. 11. When the displacement parameter is different from zero the steady state is reached more rapidly. We still note the presence of squeezing in the steady state for a small coherent amplitude.

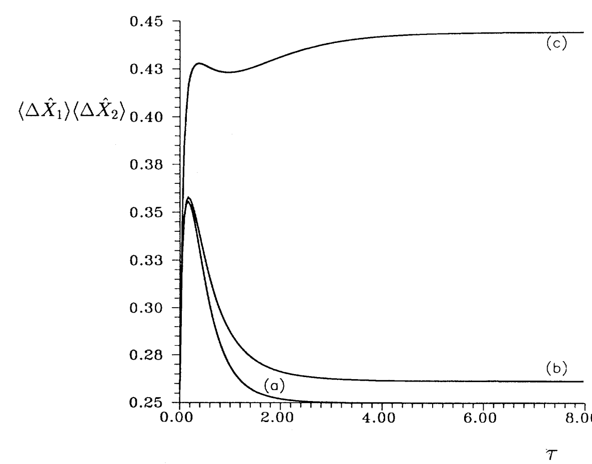

FIG. 13. Product of the fluctuations in $\hat{X}_1$ and $\hat{X}_2$ for the same initial states as in Fig. 11. General departure from the minimum uncertainty value shows that the squeezed state is not invariant under two-photon absorption.

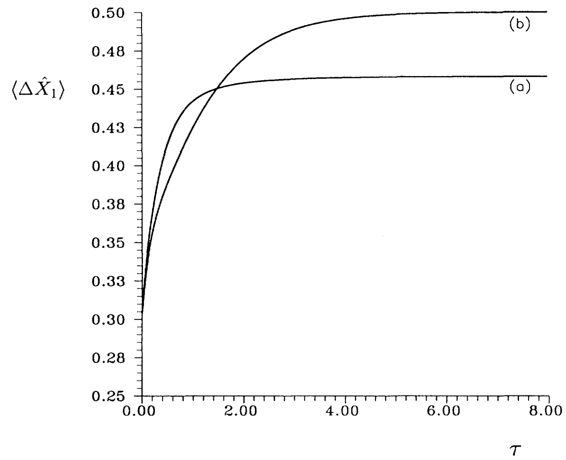

FIG. 14. Same as Fig. 11, but the squeezing parameter is now chosen to be $r=0.5$; (a) corresponds to $\beta^2=0.1$, (b) for the squeezed vacuum.

The existence of squeezing in the steady state is due to the nonvanishing value of $\langle\hat{a}(\infty)\rangle$. Contrary to linear absorption, the two-photon absorption retains phase information in the steady state. The system does not undergo a simple diagonalization, but preserves coherence and decays to a mixture of two states, $|+\rangle$ and $|-\rangle$, which are a rotation of the vacuum and the one-photon state. We calculate from Eq. (21) the eigenstates and eigenvalues of the steady-state field density matrix:

$$\hat{\rho}(\infty)=\lambda_+|+\rangle\langle+|+\lambda_-|-\rangle\langle-| \;, \tag{76}$$

where

$$\begin{aligned}
&\lambda_\pm=\tfrac{1}{2}\pm\tfrac{1}{2}[1-4(\gamma_0\gamma_1-\gamma_{01}^2)]^{1/2} \;, \\
&|+\rangle=\cos\left[\frac{\theta}{2}\right]|0\rangle+\sin\left[\frac{\theta}{2}\right]|1\rangle \;, \\
&|-\rangle=-\sin\left[\frac{\theta}{2}\right]|0\rangle+\cos\left[\frac{\theta}{2}\right]|1\rangle \;, \\
&\tan(\theta)=2\gamma_{01}(\gamma_0-\gamma_1)^{-1}, \quad 0\le\theta\le\pi \;.
\end{aligned} \tag{77}$$

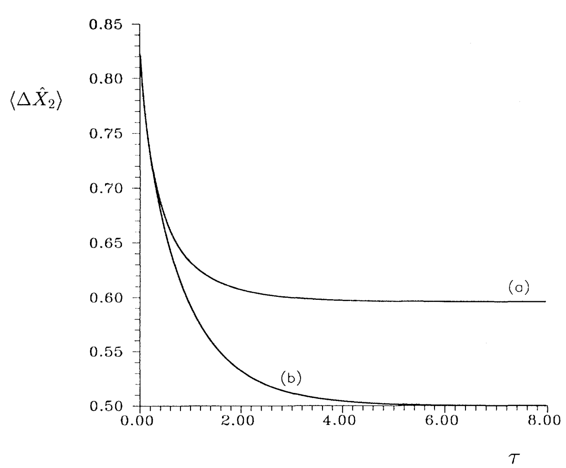

FIG. 15. Same as Fig. 12, except initial conditions are those given in Fig. 14.

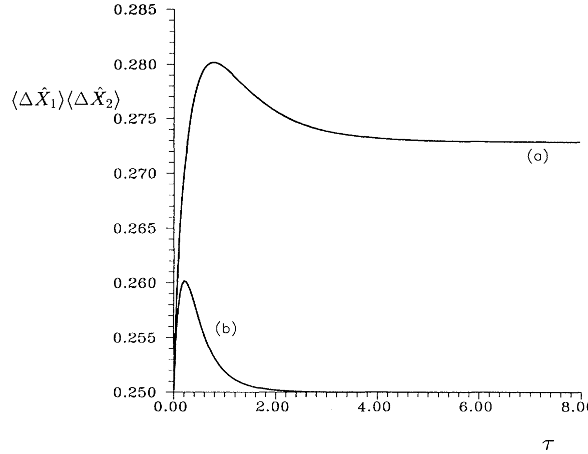

FIG. 16. Same as Fig. 13, except initial conditions given in Fig. 14.

The eigenvalues $\lambda_\pm$ satisfy

$$\begin{aligned}
&\lambda_+ +\lambda_- =\gamma_0+\gamma_1=1 \;, \\
&\lambda_+\lambda_- =\gamma_0\gamma_1-\gamma_{01}^2 \;.
\end{aligned} \tag{78}$$

Deviation from pure state values appears through the inequality

$$0\le\lambda_+\lambda_-\le\tfrac{1}{4} \;. \tag{79}$$

The lower bound corresponds to a pure state ($\hat{\rho}^2=\hat{\rho}$) and is reached by an initial squeezed vacuum, while the upper bound gives the maximal mixed state (degenerate eigenvalues, $\lambda_+=\lambda_-=0.5$), with an entropy equal to ~ 0.69. In Fig. 17 we plot the asymptotic field entropy [20]

$$S[\hat{\rho}(\infty)]=-\lambda_-\ln(\lambda_-)-\lambda_+\ln(\lambda_+) \;, \tag{80}$$

for different displacement parameters versus the squeezing parameter. Even a small coherent component leads

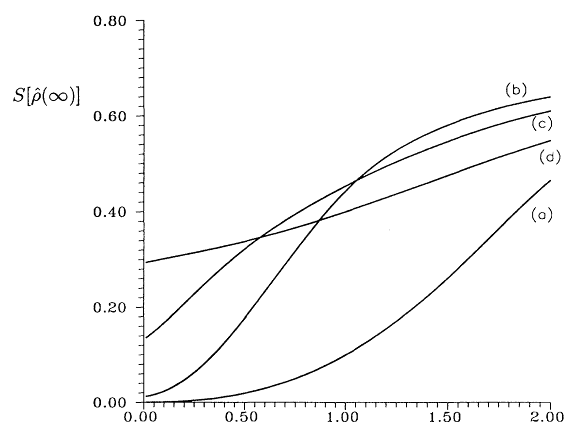

FIG. 17. Asymptotic field entropy vs the squeezing parameter. (a) $\beta^2=0.01$; (b) $\beta^2=0.25$; (c) $\beta^2=1$; (d) $\beta^2=4$. Even a small degree of coherent amplitude leads to a significant deviation from a pure state.

to a significant deviation from a pure state when the squeezing is important. When the coherent component is dominant and the squeezing not too strong ($r \leq 1$), the asymptotic entropy is nearly independent of the squeezing and equal to ~ 0.33. This can also be observed from Fig. 1, where we see that the off-diagonal elements γ_{01} of the density operator become independent of the initial parameters and equal to ~ 0.4 when the initial coherent amplitude is large, leading to a steady-state density operator which does not depend on the initial mean photon number and whose eigenvalues are $\lambda_{\pm} \simeq 0.5 \pm 0.4$.

D. Even and odd coherent-state analysis

Using the expressions (54) and (55) for the even and odd coherent states, we calculate the initial quadrature variances:

$$\langle (\Delta \hat{X}_1)^2 \rangle_{e,o} = \frac{1}{4} + \frac{\beta^2}{1 \pm \exp(-2\beta^2)} > \frac{1}{4} , \tag{81}$$

$$\langle (\Delta \hat{X}_2)^2 \rangle_{e,o} = \frac{1}{4} \mp \frac{\beta^2 \exp(-2\beta^2)}{1 \pm \exp(-2\beta^2)} . \tag{82}$$

We see from these two equations that the odd coherent state displays enhanced fluctuations, whereas the even coherent state displays reduced fluctuations in $\hat{X}_2$, but the amount of squeezing is significant only for small values of β. In Fig. 18 we plot the dynamical evolution of the squeezing in $\hat{X}_2$. An even coherent state with $\beta = 2$ exhibits initially nearly vacuum noise. Under the effect of two-photon dissipation, the fluctuations increase above the vacuum level. After an interaction time $\tau \simeq 0.23$, they have increased by 24%. We note in Fig. 9 that the same initial state develops sub-Poissonian statistics but on a shorter time scale such that sub-Poissonian statistics and squeezing never coexist; the relative number variance falls 26% below the Poissonian level at $\tau \simeq 0.13$. This clearly illustrates that the even coherent state is not invariant under two-photon absorption. When β is smaller, i.e., when the two coherent states $|\beta\rangle$ and $|-\beta\rangle$ are closer to each other, the evolution is akin to the squeezed vacuum case (Fig. 11). After this first increase in noise, the two-photon absorption reduces slowly the fluctuations in $\hat{X}_2$ and produces 12% squeezing at $\tau \simeq 1.5$. In Fig. 19 we show the evolution of the fluctuations in $\hat{X}_1$ and in Fig. 20 the product of the uncertainties. Only the

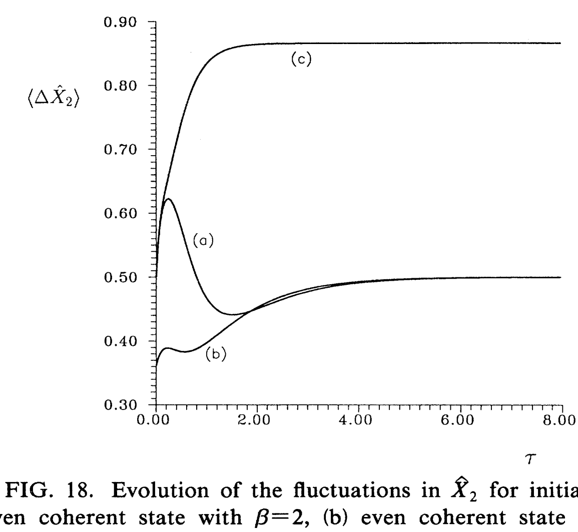

FIG. 18. Evolution of the fluctuations in $\hat{X}_2$ for initial (a) even coherent state with $\beta = 2$, (b) even coherent state with $\beta = 1$, and (c) odd coherent state with $\beta = 2$. We observe in (a) and (b) first an increase of the noise; in particular in (a), the coherent component is large and the even coherent state displays fluctuations in $\hat{X}_2$ greater than the vacuum level. This gain in noise is accompanied in a reduction of the fluctuation in the number operator below the Poissonian level, but on a shorter time scale (Fig. 9). After a maximal noise level is reached, the field recovers squeezing. Finally, it relaxes to the steady state characterized by vacuum fluctuations for the even coherent states and one-photon state fluctuations for the odd coherent state.

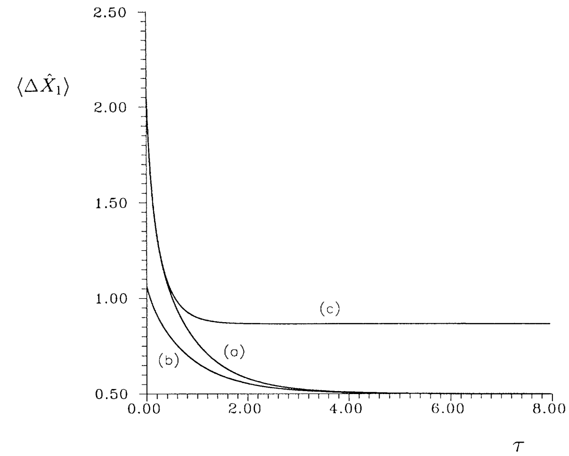

FIG. 19. Evolution of fluctuations in $\hat{X}_1$ for the same initial states as in Fig. 18. We observe a simple monotonic decrease of the noise in this quadrature. The odd and even coherent states follow initially the same evolution; then they evolve apart towards their respective steady state.

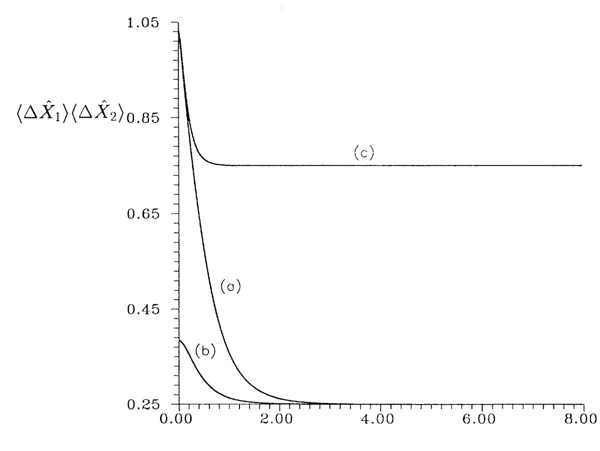

FIG. 20. Product of the uncertainties in $\hat{X}_1$ and $\hat{X}_2$ for the same initial states as in Fig. 18. The different fields remain nonminimum uncertainty states during their dissipation by the two-photon absorber.

odd coherent states which decay to a one-photon state retain extra noise.

VI. CONCLUSIONS

We have studied the dynamical evolution of two different kinds of nonclassical states undergoing a two-photon absorption process, respectively, squeezed coherent states and eigenstates of the two-photon annihilation operator, namely, the even and odd coherent states. We have shown that initial super-Poissonian fields give rise to the highest absorption rate. For appropriate initial parameters, a squeezed variable can retain reduced fluctuations in the steady state, although the excitation levels are of course very small. This effect therefore is likely to be of little importance in optical squeezing but will be of interest in micromaser systems where the mean number of photons is already small. Finally, the noninvariance of the squeezed states has been demonstrated in the uncertainty relation as a departure from the minimum uncertainty squeezed state value. Deviation from super-Poissonian statistics accompanied by extra noise in the squeezed quadrature of an initial even coherent state shows in the same way the noninvariance of eigenstates of the two-photon annihilation operator.

ACKNOWLEDGMENTS

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