

Two-mode laser with excess noise

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In this paper we investigate how excess noise behaves in a two-mode laser below and above threshold. In both cases the mode correlations of the stationary state modify the noise essentially. Our two-mode model relates directly to experimental situations.

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I. INTRODUCTION

In the usual lasers the linewidth is given by the Schawlow-Townes formula [1–3]. In certain systems, however, the noise can be enhanced by the so-called excess-noise factor. This was predicted by Petermann [4] for gain-guided semiconductor lasers. Later it was generalized for arbitrary systems within a semiclassical theory [5] and recently within a complete quantum theory [6]. Also many concrete models have been analyzed theoretically in detail [7]. On the other hand, the essential parts of the theory have been confirmed experimentally in various systems with large output coupling [8], coupled transverse polarizations [9], or inserted small apertures [10]. The measurement of an excess-noise factor up to a few hundreds for geometrically unstable laser cavities [11] is particularly remarkable.

In our earlier papers [6], we have discussed the excess noise in the linear operation regime, that is, just around the threshold for lasing. In this case of a linear attenuator or quantum amplifier, the excess noise is determined by only a few properties of the reservoirs, and the results emerging are of rather universal character. Unfortunately this general behavior breaks down when saturation sets in. In an actual laser, the noise depends on the details of the physical configuration, which greatly complicates the analysis. In this paper we treat a model case which, however, relates closely to experimentally realizable situations.

We analyze the case of a two-mode laser [12] which corresponds exactly to the experimental configuration of Ref. [9]. Due to a non-Hermitian coupling between the two polarization modes of the laser, the linewidth shows excess noise. We consider the two transverse polarizations. Because of the geometry of the cavity and of the reservoir, we can neglect all other modes and the many-mode treatment of Ref. [6] reduces to two discrete modes. Using the Faraday effect, the frequencies of the two circularly polarized modes are split by an external magnetic field. Due to anisotropic losses, the two circularly polarized modes are coupled in a non-Hermitian way. This leads to nonorthogonal eigenmodes—the so-called quasimodes. In the theory of excess noise, so far, the enhancement factor only depends on the properties of the quasimodes, in particular their scalar products or overlaps. We show that the amount of excess noise is modified by the correlations between the quasimodes. These correlations are determined by the stationary state and may vary strongly even in cases when the usual excess noise is sup-

posed to remain constant. We first analyze the linear case below threshold. There the spectrum can be calculated analytically. In the nonlinear case above threshold, we only treat the case of small nonlinearities. We can make a prediction for the maximally possible excess noise and suggest how to increase or decrease the noise in the corresponding experiment of Ref. [9].

II. LINEAR AMPLIFIER

The general case of the linear amplifier is described by the master equation [6]

$$\begin{aligned} \frac{d}{dt}\hat{\rho} = & \frac{1}{2} \sum_{n,m} L_{m,n} \{ 2\hat{a}_n^\dagger \hat{\rho} \hat{a}_m - \hat{a}_m \hat{a}_n^\dagger \hat{\rho} - \hat{\rho} \hat{a}_m \hat{a}_n^\dagger \} \\ & + \frac{1}{2} \sum_{n,m} \Gamma_{m,n} \{ 2\hat{a}_n \hat{\rho} \hat{a}_m^\dagger - \hat{a}_m^\dagger \hat{a}_n \hat{\rho} - \hat{\rho} \hat{a}_m^\dagger \hat{a}_n \} \\ & - i \sum_n \omega_n [\hat{a}_n^\dagger \hat{a}_n, \hat{\rho}] \end{aligned} \quad (1)$$

for the density operator $\hat{\rho}$ where \hat{a}_n and \hat{a}_n^\dagger denote the annihilation and creation operators of the field modes. The matrices

$$L_{m,n} = \frac{1}{V} \int d^3x R_L(x) [u_n(x) d_L(x)] [u_m(x) d_L(x)] \quad (2)$$

and

$$\Gamma_{m,n} = \frac{1}{V} \int d^3x R_\Gamma(x) [u_n(x) d_\Gamma(x)] [u_m(x) d_\Gamma(x)] \quad (3)$$

are determined by the parameters of the reservoirs: the amplification rate $R_L(x)$, the loss rate $R_\Gamma(x)$, and the directions of the dipole moments $d_L(x)$ and $d_\Gamma(x)$. In contrast to the treatment in Ref. [6], we have here made the assumption that the spread between the frequencies ω_n is small relative to their mean value.

We consider the two transverse polarizations

$$\begin{pmatrix} 1 \\ 0 \end{pmatrix} \text{ and } \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

of a cavity mode $u(x)$. Using the Faraday effect, we assume that we can control the frequency splitting $\pm\Omega$ between the two circularly polarized modes,

$$u_{\pm}(x) = \begin{pmatrix} 1 \\ \pm i \end{pmatrix} u(x)/\sqrt{2},$$

by applying a magnetic field to the medium. In addition, the field is assumed to experience an isotropic gain A and anisotropic losses with the rates C_h and C_v in the directions of the horizontal and vertical polarizations, respectively.

A. Fokker-Planck equation

Starting from the master equation, we arrive at the following Fokker-Planck equation for the P function:

$$\begin{aligned} \dot{P} = & \frac{1}{2} \sum_{n,m=\pm} L_{n,m} \left\{ 2 \frac{\partial^2}{\partial \alpha_n \partial \alpha_m^*} - \frac{\partial}{\partial \alpha_m^*} \alpha_n^* - \frac{\partial}{\partial \alpha_n} \alpha_m \right\} P \\ & + \frac{1}{2} \sum_{n,m=\pm} \Gamma_{n,m} \left\{ \frac{\partial}{\partial \alpha_m^*} \alpha_n^* + \frac{\partial}{\partial \alpha_n} \alpha_m \right\} P \\ & - i \sum_{n,m=\pm} \omega_{n,m} \left\{ \frac{\partial}{\partial \alpha_m^*} \alpha_n^* - \frac{\partial}{\partial \alpha_n} \alpha_m \right\} P, \end{aligned} \quad (4)$$

with the matrices defined as

$$L = \frac{A}{2} \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}, \quad (5)$$

$$\Gamma = \frac{1}{2} \begin{bmatrix} C_h + C_v & C_h - C_v \\ C_h - C_v & C_h + C_v \end{bmatrix}, \quad (6)$$

and

$$\omega = \begin{bmatrix} \Omega & 0 \\ 0 & -\Omega \end{bmatrix}. \quad (7)$$

The indices \pm denote the two circularly polarized modes. Here we are working in a frame rotating with the mean frequency of the two modes.

To solve the Fokker-Planck equation, we make the ansatz

$$\begin{aligned} P(\alpha_+, \alpha_-, \alpha_+^*, \alpha_-^*, t) \\ = N(t) \exp \left\{ -\frac{1}{2} \sum_{k,l} [\alpha_l^* - \bar{\alpha}_l^*(t)] G_{l,k}(t) [\alpha_k - \bar{\alpha}_k(t)] \right\}, \end{aligned} \quad (8)$$

with the mean values of the amplitudes $\bar{\alpha}_k(t) = \langle \hat{a}_k \rangle$, the Hermitian matrix $G(t)$, and the normalization constant $N(t) = \det\{G(t)\}/(2\pi)^2$. The inverse of the matrix G ,

$$2(G^{-1})_{k,l} = \langle \hat{a}_l^\dagger \hat{a}_k \rangle - \langle \hat{a}_l^\dagger \rangle \langle \hat{a}_k \rangle, \quad (9)$$

is the normally ordered correlation. Knowing the equations of motion for $\langle \hat{a}_k \rangle$ and $\langle \hat{a}_l^\dagger \hat{a}_k \rangle$ [6], we find the equations of motion for $\bar{\alpha}_k$ and G^{-1} to be given by

$$\frac{d}{dt} \begin{pmatrix} \bar{\alpha}_+ \\ \bar{\alpha}_- \end{pmatrix} = D \begin{pmatrix} \bar{\alpha}_+ \\ \bar{\alpha}_- \end{pmatrix} \quad (10)$$

and

$$\frac{d}{dt} G^{-1} = D G^{-1} + G^{-1} D^\dagger + \frac{1}{2} L, \quad (11)$$

with $D = (L - \Gamma)/2 - i\omega$.

The general solutions of Eqs. (10) and (11) are given by

$$\begin{pmatrix} \bar{\alpha}_+(t) \\ \bar{\alpha}_-(t) \end{pmatrix} = e^{Dt} \begin{pmatrix} \bar{\alpha}_+(0) \\ \bar{\alpha}_-(0) \end{pmatrix} \quad (12)$$

and

$$G^{-1}(t) = e^{Dt} [G^{-1}(0) - G_s^{-1}] e^{D^\dagger t} + G_s^{-1}, \quad (13)$$

where G_s^{-1} is the stationary solution of Eq. (11) which is unique as long as $\Gamma \neq L$. For the Green function \mathcal{G} of the Fokker-Planck equation (4) we use the initial condition $G^{-1}(0) = 0$ so that $\mathcal{G}(\alpha_+, \alpha_-, \alpha_+(0), \alpha_-(0), t=0) = \delta^{(2)}(\alpha_+ - \alpha_+(0)) \delta^{(2)}(\alpha_- - \alpha_-(0))$. Here $\delta^{(2)}$ denotes the two-dimensional delta function.

For our particular example, we find

$$G_s^{-1} = \frac{1}{2} \frac{A}{C_h + C_v - A} \begin{bmatrix} (C_h + C_v - A)^2 + 16\Omega^2 & (C_h - C_v)(C_h + C_v - A + 4i\Omega) \\ (C_h - C_v)(C_h + C_v - A - 4i\Omega) & (C_h + C_v - A)^2 + 16\Omega^2 \end{bmatrix} \quad (14)$$

and

$$e^{Dt} = e^{(C_h + C_v - A)t/4} \begin{pmatrix} \frac{\sin(\sqrt{\Omega^2 - (C_h - C_v)^2/16}t)}{\sqrt{\Omega^2 - (C_h - C_v)^2/16}} \begin{bmatrix} -i\Omega & (C_h - C_v)/4 \\ (C_h - C_v)/4 & i\Omega \end{bmatrix} + \cos[\sqrt{\Omega^2 - (C_h - C_v)^2/16}t] \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \end{pmatrix}. \quad (15)$$

This stationary state is valid for the case below threshold $A < C_h + C_v$.

B. Spectrum

The spectrum of the electromagnetic field is defined as the Fourier transform of the two-time correlation function $\langle E^{(-)}(t+\tau)E^{(+)}(t) \rangle_s$ in the stationary state ($t \rightarrow \infty$). In order to obtain the spectrum for a certain polarization orientation, we have first to project the field onto the normalized vector v of this orientation. To do so, it is convenient to calculate first

$$M(\tau) = \begin{bmatrix} \langle \hat{a}_+^\dagger(t+\tau)\hat{a}_+(t) \rangle_s & \langle \hat{a}_+^\dagger(t+\tau)\hat{a}_-(t) \rangle_s \\ \langle \hat{a}_-^\dagger(t+\tau)\hat{a}_+(t) \rangle_s & \langle \hat{a}_-^\dagger(t+\tau)\hat{a}_-(t) \rangle_s \end{bmatrix} \quad (16)$$

and then to project onto the particular direction

$$v = \begin{pmatrix} v_+ \\ v_- \end{pmatrix}$$

using

$$M^{(\pm)}(\tau) = e^{\tau d^{(\pm)*}} v^{(\pm)\dagger} G_s^{-1} * v^{(\pm)}$$

$$= \frac{A}{C_h + C_v - A} \frac{[(C_h + C_v - A)^2 + 16\Omega^2 + (C_h - C_v)^2]}{(C_h + C_v - A)^2 + 16\Omega^2 - (C_h - C_v)^2} e^{-(C_h + C_v - A)\tau/4 \mp i\sqrt{\Omega^2 - (C_h - C_v)^2}/16\tau}. \quad (19)$$

Here

$$d^{(\pm)} = -(C_h + C_v - A)/4 \pm i\sqrt{\Omega^2 - (C_h - C_v)^2}/16 \quad (20)$$

are the eigenvalues of D . This result is for the case below threshold, $A < C_h + C_v$, and outside the locking region, $4|\Omega| \geq |C_h - C_v|$, when the two eigenvalues $d^{(\pm)}$ have different imaginary parts. Taking the real part of the Fourier transform we obtain a Lorentzian spectral line with the width

$$\Delta\omega = \frac{1}{2}(C_h + C_v - A). \quad (21)$$

Hence, the linewidth itself does not show any excess width. However, considering that the quasi mode experiences a loss rate $(C_h + C_v)/2$ and an amplification rate $A/2$, the stationary state intensity given by $I_s = M(0)$ shows an excess of intensity by a factor

$$K = \frac{(C_h + C_v - A)^2 + 16\Omega^2 + (C_h - C_v)^2}{(C_h + C_v - A)^2 + 16\Omega^2 - (C_h - C_v)^2} \quad (22)$$

in comparison with intensity $A/(C_h + C_v - A)$ given by the single-mode theory. Note that in the linear regime the intensity of the field is entirely due to the field uncertainty (noise). However, a similar argument will be seen to apply for the

$$v^\dagger M(\tau)v = \langle [v_+^* \hat{a}_+^\dagger(t+\tau) + v_-^* \hat{a}_-^\dagger(t+\tau)][v_+ \hat{a}_+(t) + v_- \hat{a}_-(t)] \rangle_s. \quad (17)$$

According to the quantum regression theorem [13], $M(\tau)$ is given by

$$M(\tau) = \int d^4\alpha \int d^4\alpha' \begin{pmatrix} \alpha_+^* \\ \alpha_-^* \end{pmatrix} \times (\alpha'_+, \alpha'_-) \mathcal{G}(\alpha_+, \alpha_-, \alpha'_+, \alpha'_-, \tau) P_s(\alpha') = 2(G_s^{-1} e^{D^\dagger \tau})^T, \quad (18)$$

where $d^4\alpha$ denotes the integration over the whole two-mode phase space and P_s is the stationary state P function.

Particularly interesting are now the polarization directions of the quasimodes which are given by the normalized eigenvectors $v^{(\pm)}$ of the matrix D . Those can be selected by adjusting the polarization of the detector accordingly. For the case $4|\Omega| > |C_h - C_v|$, we find

regime above threshold in the next section. As we see from Eq. (19), the excess noise factor depends on the stationary state through $v^{(\pm)\dagger} G_s^{-1} * v^{(\pm)}$. If G_s^{-1} is proportional to the unit matrix, then $K=1$ since then the two quasimodes are correlated in such a way that their individual excess noises cancel each other. This is the case for $(C_h + C_v - A) \gg 4|\Omega|$. The two quasimodes are uncorrelated only if $(C_h + C_v - A) \ll 4|\Omega|$, which means that G_s^{-1} is proportional to the incoherent superposition $v^{(+)} \otimes v^{(+)\dagger} + v^{(-)} \otimes v^{(-)\dagger}$; i.e., we then have

$$K \approx \frac{16\Omega^2 + (C_h - C_v)^2}{16\Omega^2 - (C_h - C_v)^2}. \quad (23)$$

This still differs from the usual result

$$K = \frac{16\Omega^2}{16\Omega^2 - (C_h - C_v)^2} \quad (24)$$

approximately by a factor of 2 for large excess noise. The maximal excess noise is given by

$$K_{max} = \frac{(C_h + C_v - A)^2 + 2(C_h - C_v)^2}{(C_h + C_v - A)^2}, \quad (25)$$

which depends only on the ratio of the isotropic and the anisotropic contributions of the reservoir as can be seen from $\Delta\omega$, Eq. (21), and G_s^{-1} , Eq. (14). It is interesting to note that in the two directions perpendicular to the two quasimodes there is no excess noise.

We can equivalently write the linewidth as

$$\Delta\omega = \frac{1}{2}(C_h + C_v - A) = K \frac{A}{I_s}. \quad (26)$$

What counts in the Shawlow-Townes linewidth formula is the ratio between the damping and amplification rates and the stationary intensity. We have chosen here the normal order to calculate the spectrum. Using symmetric or antinormal order, the K factor is slightly different. For instance, for the antinormal order, G_s^{-1} has to be replaced by $G_s^{-1} + 1$ in Eq. (19). However, in the regime of large excess noise, $(C_h + C_v - A) \ll 4|\Omega|$, the different orderings agree.

III. NONLINEAR AMPLIFIER

In this section we are considering an amplifier with saturation. This system describes the laser above threshold with $A > C_h + C_v$. In the Appendix we derive the equations for a multimode amplifier with saturation from an atomic model for the reservoir. We restrict ourselves to the case of small saturation. Then, like in the single-mode case, there is only a fourth-order term of the type

$$\sum_{n,m,k,l} \mathcal{L}_{n,m,k,l} \{ \hat{\rho}_F \hat{a}_n \hat{a}_m^\dagger \hat{a}_k \hat{a}_l^\dagger - 4 \hat{a}_l^\dagger \hat{\rho}_F \hat{a}_n \hat{a}_m^\dagger \hat{a}_k + 6 \hat{a}_k \hat{a}_l^\dagger \hat{\rho}_F \hat{a}_n \hat{a}_m^\dagger - 4 \hat{a}_m^\dagger \hat{a}_k \hat{a}_l^\dagger \hat{\rho}_F \hat{a}_n + \hat{a}_n \hat{a}_m^\dagger \hat{a}_k \hat{a}_l^\dagger \hat{\rho}_F \} \quad (27)$$

added to the master equation (1) with the coefficients $\mathcal{L}_{n,m,k,l}$ as defined in Eq. (A20) in the Appendix. As can be seen from the definition, \mathcal{L} depends on the geometry of the reservoir. We only consider the three generic cases [2] of mode competition, mode locking, and the neutral one of coexistence of the two modes.

A. Quasimodes

The first point which we are addressing is how the quasimodes are modified by the saturation. For this purpose, we are considering the expectation values of the field amplitudes without noise. In that case, the quartic terms in the master equation can be approximately factorized in the amplitudes. Then the time evolution is covered by an equation like Eq. (10):

$$\begin{aligned} \frac{d}{dt} \begin{pmatrix} \bar{\alpha}_+ \\ \bar{\alpha}_- \end{pmatrix} &= D \begin{pmatrix} \bar{\alpha}_+ \\ \bar{\alpha}_- \end{pmatrix} - \frac{B}{2} (|\bar{\alpha}_+|^2 + |\bar{\alpha}_-|^2) \begin{pmatrix} \bar{\alpha}_+ \\ \bar{\alpha}_- \end{pmatrix} \\ &= D_{\text{eff}} \begin{pmatrix} \bar{\alpha}_+ \\ \bar{\alpha}_- \end{pmatrix}, \end{aligned} \quad (28)$$

with an effective time evolution operator given by

$$D_{\text{eff}} = \frac{1}{2}(L - \Gamma) - i\omega - \frac{B}{2} (|\bar{\alpha}_+|^2 + |\bar{\alpha}_-|^2) \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}. \quad (29)$$

The parameter B describes the strength of the saturation and is proportional to the amplification rate A . In this isotropic case, all polarization directions experience the same saturation. All possible superpositions of the two quasimodes are stable. If the initial condition excites a pure quasimode, the oscillation is perfectly harmonic as shown in Fig. 1(a). However, if both quasimodes are excited, the oscillations of both modes show higher harmonics of the quasimode frequency as shown in Fig. 1(b). The strength of the higher harmonics depends on the ratio between the quasimode frequency $\sqrt{\Omega^2 - (C_h - C_v)^2}/16$ and the time scale of the (intensity) saturation determined by $A - (C_h + C_v)$ and B . The time scale of the saturation can be chosen independently of the stationary state intensity, which is given by $(A - C_h - C_v)/B$. If the saturation process is much slower than the quasimode frequency, then the oscillation is harmonic again as shown in Fig. 1(c). The nonlinear term is given by the sum of the intensities of the two quasimodes independently. This can be shown easily by applying the rotating wave approximation to the nonlinear term in Eq. (28). In the other extreme case, when the saturation is much faster than the quasimode frequency, the intensity of each quasimode is oscillating with the quasimode frequency which leads to higher harmonics of field oscillation. Then the real part of the field amplitude approximates a rectangular function whereas the imaginary part resembles a triangular function as shown in Fig. 1(b). Note that the distinction between real and imaginary parts depends on the choice of the initial phase.

When the reservoir has its dipole moments oriented in the direction of the horizontal and of the vertical polarization, then we have a case with

$$D_{\text{eff}} = (L - \Gamma)/2 - i\omega - \frac{B}{2} \begin{bmatrix} |\bar{\alpha}_+|^2 + |\bar{\alpha}_-|^2 & \bar{\alpha}_+ \bar{\alpha}_-^* + \bar{\alpha}_+^* \bar{\alpha}_- \\ \bar{\alpha}_+ \bar{\alpha}_-^* + \bar{\alpha}_+^* \bar{\alpha}_- & |\bar{\alpha}_+|^2 + |\bar{\alpha}_-|^2 \end{bmatrix}. \quad (30)$$

Here the quasimodes are competing. Depending on the initial condition either one or the other is excited and suppresses the other as shown in Fig. 2. When the reservoir has its dipole moments oriented uniformly over all transverse directions of the polarization, we have a case similar to the isotropic one with

$$D_{\text{eff}} = \frac{1}{2}(L - \Gamma) - i\omega - \frac{B}{4} \times \begin{bmatrix} |\bar{\alpha}_+|^2 + 2|\bar{\alpha}_-|^2 & 0 \\ 0 & 2|\bar{\alpha}_+|^2 + |\bar{\alpha}_-|^2 \end{bmatrix}. \quad (31)$$

This type of reservoir constitutes a realistic description of an amplifier without any polarization or frequency dependence. When the quasimode frequency is much faster than the amplification and saturation processes, the situation behaves

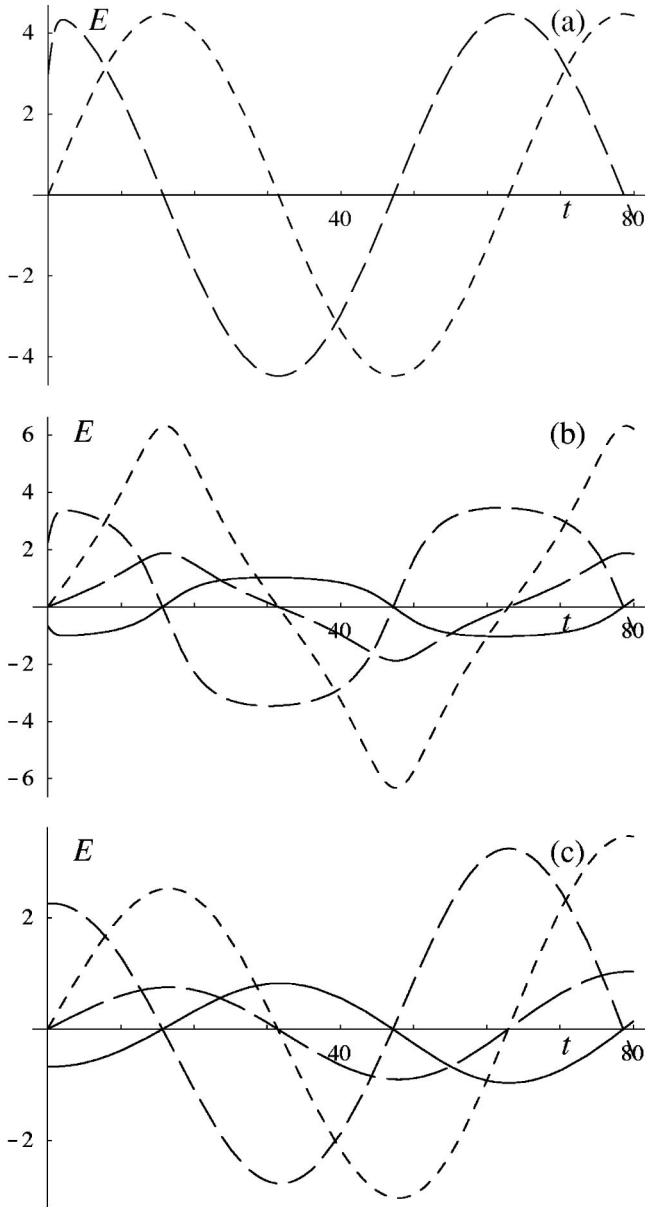


FIG. 1. Behavior of the quasimodes in the isotropic case. Exactly one quasimode is excited in (a). The oscillation of the real part (dashed line) and the imaginary part of the field amplitude E (short dashed line) are perfectly harmonic. The parameters are $(A - C_h - C_v)/2 = 2$, $(C_h - C_v)/2 = 1.99$, $B/2 = 0.1$, and $\Omega = 1$. The oscillation period and the intensity are given by $\sqrt{\Omega^2 - (C_h - C_v)^2/16}$ and $(A - C_h - C_v)/B$, respectively. In (b) also a small part of the other quasimode (real part solid line, imaginary part long dashed line) is excited by the initial condition. The oscillation shows strong nonlinearities. In (c) we show the same situation as in (b) but $(A - C_h - C_v)/2 = 0.02$ and $B/2 = 0.001$. This results in the same intensities; however, the higher harmonics are practically not visible.

like in the previous case as shown in Fig. 3(a). Otherwise it exhibits mode competition between the horizontal and vertical polarization and the oscillation frequency is given by the bare mode frequency Ω as shown in Figs. 3(b) and 3(c). The exactly isotropic case can be realized by a reservoir, as used above, in combination with a reservoir of the same strength

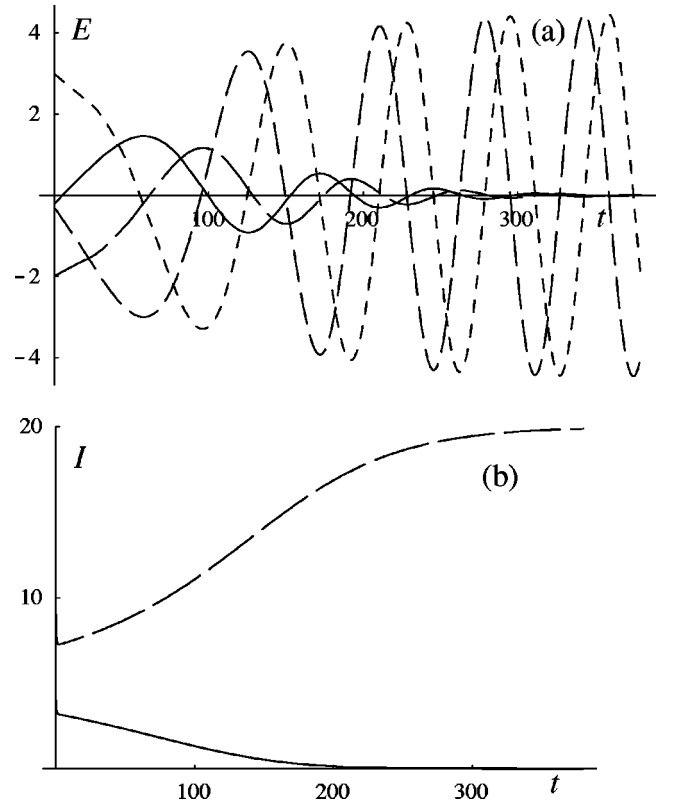


FIG. 2. Case of mode competition. The initial condition excites the two quasimodes in the ratio 3:2 of the amplitudes. We use $(A - C_h - C_v)/2 = 2$ and $B/2 = 0.1$; the remaining parameters are as in Fig. 1. In (a) we show the amplitudes E and in (b) the intensities I of the quasimodes. After a few oscillations the mode which was initially excited more strongly supersedes the other one.

which amplifies independently the right and left circularly polarizations, respectively. Note that different reservoirs can yield identical matrices L but different tensors \mathcal{L} .

For the purpose of this paper we are satisfied with this rather qualitative discussion. All we want to emphasize is that the quasimode description is not changed by the nonlinearities—at least not in the case of perfectly isotropic saturation. Only some features like stability, mode competition, and higher harmonics of the quasimodes depend on the details of the reservoir.

B. Linewidth

In this section, we investigate the linewidth of the quasimodes in the saturated case. Then the intensity is taken to be stabilized, and the linewidth is mainly caused by the phase diffusion. In the Fokker-Planck equation (4), the (normal-order) noise originates from the diffusion term $\sum_{n,m=\pm} L_{n,m} \partial^2 / \partial \alpha_n \partial \alpha_m^*$. When we express this in the amplitudes β_ν of the quasimodes, we find

$$\begin{aligned} & \sum_{\nu,\mu=\pm} \left(\sum_{n,m=\pm} L_{n,m} c_n^{(\mu)*} c_n^{(\nu)} \right) \frac{\partial^2}{\partial \beta_\nu \partial \beta_\mu^*} \\ &= \frac{A}{2} \sum_{\nu,\mu=\pm} \left(\sum_{n=\pm} c_n^{(\mu)*} c_n^{(\nu)} \right) \frac{\partial^2}{\partial \beta_\nu \partial \beta_\mu^*}. \end{aligned} \quad (32)$$

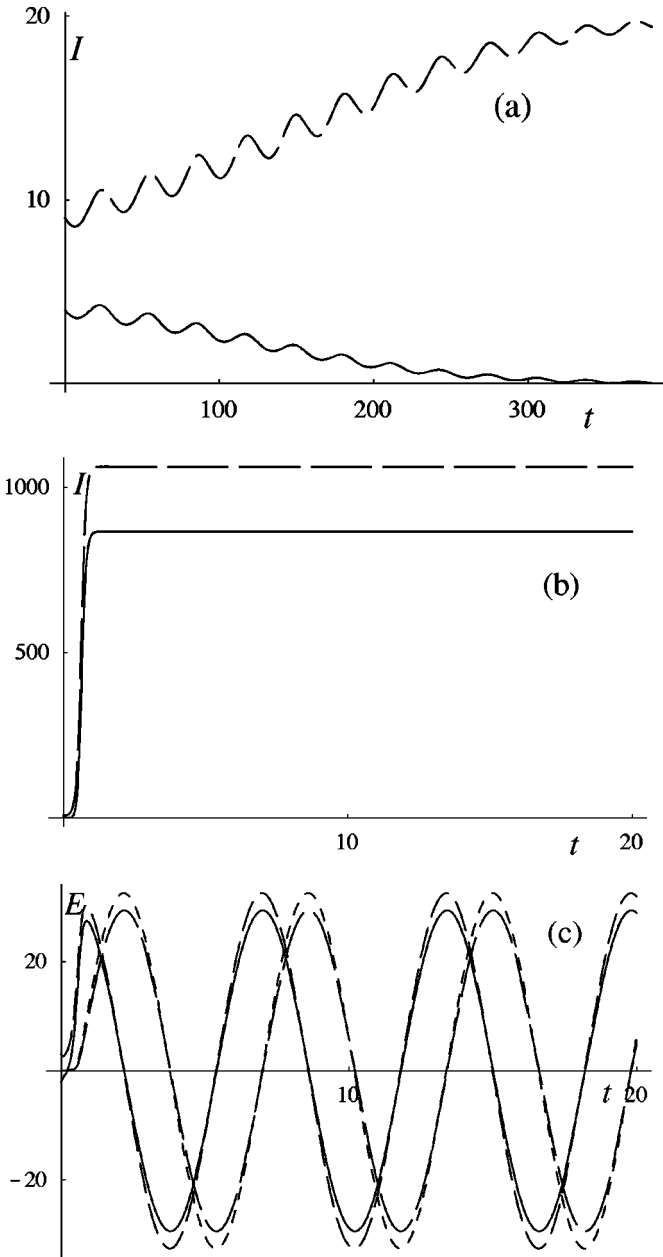


FIG. 3. Realistic reservoir competition. The initial condition excites the two quasimodes in the ratio 3:2 of the amplitudes; the remaining parameters are as in Fig. 1. In (a) we show the two intensities I for $(A - C_h - C_v)/2 = 0.02$ and $B/2 = 0.001$ and in (b) for $(A - C_h - C_v)/2 = 20$ and $B/2 = 1$. Whereas (a) exhibits mode competition, in (b) the quasimodes are locked and the field amplitudes E oscillate with the bare mode frequency Ω as shown in (c); the (stable) stationary state is approximately given by the vertical polarization which experiences the smallest loss.

Here, the

$$\begin{pmatrix} c_+^{(\nu)} \\ c_-^{(\nu)} \end{pmatrix}$$

are the (unnormalized) eigenvectors of D which define the quasimode operators $\hat{A}_\nu = c_+^{(\nu)} \hat{a}_+ + c_-^{(\nu)} \hat{a}_-$ together with the quasimode function

$$\frac{1}{(c_+^{(\nu)2} + c_-^{(\nu)2})} \begin{pmatrix} c_+^{(\nu)} \\ c_-^{(\nu)} \end{pmatrix} \quad (33)$$

expressed on the basis of the circular polarization modes.

Now we will consider only the diagonal term $\partial^2/\partial\beta_+ \partial\beta_+^*$. When the two quasimodes are orthogonal the cross term is zero. When they are almost identical, the cross term leads to a mixing of the two modes; however, this does not contribute significantly to the noise. Transforming to a polar representation of the field amplitude $\beta_+ = R_+ e^{i\varphi_+}$, we get

$$\frac{\partial^2}{\partial\beta_+ \partial\beta_+^*} = \frac{1}{4} \left(\frac{1}{R_+^2} \frac{\partial^2}{\partial\varphi_+^2} + \frac{1}{R_+} \frac{\partial}{\partial r} + \frac{\partial^2}{\partial r^2} \right). \quad (34)$$

Assuming that the modulus R_+ of the quasimode amplitude is constant because of the saturation, the diffusion term

$$(|c_+^{(+)}|^2 + |c_-^{(+)}|^2) \frac{A}{2} \frac{1}{4R_+^2} \frac{\partial^2}{\partial\varphi_+^2} \quad (35)$$

only describes diffusion of the phase φ_+ .

The interesting point is to see how the quasimode amplitude R_+ connects to the intensity of the field in the polarization direction of the quasimode. Using the field operator expressed in terms of quasimodes,

$$\hat{E} = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \hat{a}_+ + \begin{pmatrix} 0 \\ 1 \end{pmatrix} \hat{a}_- = \sum_{\nu=\pm} \frac{1}{c_+^{(\nu)2} + c_-^{(\nu)2}} \begin{pmatrix} c_+^{(\nu)} \\ c_-^{(\nu)} \end{pmatrix} \hat{A}_\nu, \quad (36)$$

we find the field in the direction of the quasimode by projecting onto the normalized eigenvectors

$$\begin{pmatrix} v_+^{(+)} \\ v_-^{(+)} \end{pmatrix} = \frac{1}{\sqrt{|c_+^{(+)}|^2 + |c_-^{(+)}|^2}} \begin{pmatrix} c_+^{(+)} \\ c_-^{(+)} \end{pmatrix}. \quad (37)$$

Hence the intensity is given by

$$I_+ = \frac{1}{|c_+^{(+)}|^2 + |c_-^{(+)}|^2} \times \sum_{\nu, \mu=\pm} \frac{\begin{pmatrix} c_+^{(\mu)*} \\ c_-^{(\mu)*} \end{pmatrix} \begin{pmatrix} c_+^{(+)} \\ c_-^{(+)} \end{pmatrix} \begin{pmatrix} c_+^{(+)*} \\ c_-^{(+)*} \end{pmatrix} \begin{pmatrix} c_+^{(\nu)} \\ c_-^{(\nu)} \end{pmatrix}}{c_+^{(\mu)2} + c_-^{(\mu)2} \quad c_+^{(\nu)2} + c_-^{(\nu)2}} \langle \hat{A}_\mu^\dagger \hat{A}_\nu \rangle. \quad (38)$$

The (usual) excess noise factor $K = (|c_+^{(\nu)}|^2 + |c_-^{(\nu)}|^2)^2 / (c_+^{(\nu)2} + c_-^{(\nu)2})^2$ is large when the two quasimodes become degenerate. In that limit the intensity can be written as

$$I_+ \approx \frac{K}{|c_+^{(+)}|^2 + |c_-^{(+)}|^2} (\langle \hat{A}_+^\dagger \hat{A}_+ \rangle + \langle \hat{A}_+^\dagger \hat{A}_- \rangle + \langle \hat{A}_-^\dagger \hat{A}_+ \rangle + \langle \hat{A}_-^\dagger \hat{A}_- \rangle). \quad (39)$$

With $\langle \hat{A}_+^\dagger \hat{A}_+ \rangle = R_+^2$, we see that the relation between the intensity I_+ in the polarization direction of the quasimode and the intensity R_+^2 of the quasimode itself depends strongly on the correlation of the two quasimodes in the stationary state. In the case of perfect competition of the quasimodes, we have $\langle \hat{A}_+^\dagger \hat{A}_- \rangle = \langle \hat{A}_-^\dagger \hat{A}_+ \rangle = 0$ and the phase diffusion constant, Eq. (35), is given by

$$\Delta\omega = (|c_+^{(+)}|^2 + |c_-^{(+)}|^2) \frac{A}{2} \frac{1}{4R_+^2} = K \frac{A}{8I_+}. \quad (40)$$

Such a stationary state can be constructed with states like

$$|\psi_+\rangle = |\alpha_+ = c_+^{(+)}\alpha_0\rangle |\alpha_- = c_-^{(+)}\alpha_0\rangle \quad (41)$$

obeying $\hat{A}_+|\psi_+\rangle = (c_+^{(+)} + c_-^{(+)}\alpha_0/\alpha_+)|\psi_+\rangle$ and $\hat{A}_-|\psi_+\rangle = 0$.

The other extreme case, $K=1$, emerges when the stationary state is constructed by a incoherent superposition of states such as the two-mode coherent state $|\psi\rangle = |\alpha_+\rangle |\alpha_-\rangle$. We find then

$$I_+ = |c_+^{(+)*}\alpha_+ + c_-^{(+)*}\alpha_-|^2 / (|c_+^{(+)}|^2 + |c_-^{(+)}|^2) \quad (42)$$

and

$$R_+^2 = |c_+^{(+)}\alpha_+ + c_-^{(+)}\alpha_-|^2. \quad (43)$$

If (α_+/α_-) is real, then $|c_+^{(+)*}\alpha_+ + c_-^{(+)*}\alpha_-|^2 = |c_+^{(+)}\alpha_+ + c_-^{(+)}\alpha_-|^2$ which yields $\Delta\omega = A/(8I_+)$ and therefore no excess noise. Note that when $|C_h - C_v| = 4|\Omega|$, for the case of maximal excess noise, $(c_+^{(+)} / c_-^{(+)})$ is purely imaginary.

The remaining step is to solve for the stationary state of the Fokker-Planck equation (4) including the saturation term, Eq. (27). An ansatz consistent with the fourth-order approximation is

$$P(\alpha_+, \alpha_-) = P_0(\alpha_+, \alpha_-) \exp\{-Bf(\alpha_+, \alpha_-)\}, \quad (44)$$

where P_0 is the solution of the linear Fokker-Planck equation and f is a quartic function. We use the coordinate representation $(\alpha_+, \alpha_-) = r(\cos\theta, e^{i\varphi}\sin\theta)$ where we choose r to be real since an overall phase factor does not matter in the stationary state. The stationary state can be written as

$$P(\alpha_+, \alpha_-) = \exp\left\{f_1(\theta, \varphi)r^2 - \frac{B}{2}f_2(\theta, \varphi)r^4\right\}. \quad (45)$$

In principle, the functions f_1 and f_2 can be calculated analytically, but for most of the interesting cases the formulas become too lengthy for any interpretation, and we only show numerical plots. For a sufficiently small nonlinearity B —which means that the laser operates with high intensity—the P function is strongly localized in the r direction. Then the essential contributions in r direction are at the maximum, $r^2 = f_1/(Bf_2)$, for any fixed θ and φ . Hence $P \approx \exp\{f_1^2/(2Bf_2)\}$ where for small B the maximum of $f_1^2/(2Bf_2)$ will dominate. From the discussion above, it follows that in the case of maximal excess noise

$$K = \left| \frac{-i\cos\theta + e^{i\varphi}\sin\theta}{i\cos\theta + e^{i\varphi}\sin\theta} \right|^2. \quad (46)$$

In Fig. 4 we show how the function $f_1^2/(2Bf_2)$ behaves for the case of isotropic saturation, when, for symmetry reasons, the maximum is at $\theta = \pi/2$. For a small net amplification $(A - C_h - C_v)/2$, the stationary state is given by states such as $|\psi_+\rangle$, and hence we expect the usual excess noise which can be expressed as $K = \tan^2(\varphi/2 - \pi/4)$. For a large net amplification, the two circular modes lock in such a way as to yield the vertical polarization which experiences a smaller loss than the horizontal one. Then no excess noise is present. In both the two extreme cases, the stationary state is highly localized which justifies our assumptions. In the parameter region between the extremes, we expect our treatment to be only quantitatively right for small saturation B . The other types of reservoirs discussed above show a similar behavior.

IV. CONCLUSIONS

The usual value of the excess noise factor is modified by the correlations of the quasimodes in the stationary state. This argument holds both below and above threshold. The only difference is that, in the earlier case, the mean amplitude of the field is zero and only the uncertainties of the fields are correlated. In the latter case, the important part of the correlation originates from the amplitudes. Writing the mode correlation as $\langle \hat{a}_n^\dagger \hat{a}_m \rangle = \bar{\alpha}_n^* \bar{\alpha}_m + \langle (\hat{a}_n^\dagger - \bar{\alpha}_n^*)(\hat{a}_m - \bar{\alpha}_m) \rangle$, this means that we can neglect $\bar{\alpha}_n^* \bar{\alpha}_m$ in the case below and $\langle (\hat{a}_n^\dagger - \bar{\alpha}_n^*)(\hat{a}_m - \bar{\alpha}_m) \rangle$ in the case above threshold. Above threshold, our approximation works best with large amplitudes, in particular since the (excess) noise of the modes may be large. Since we are, however, using only the first order of the nonlinearities, the amplitudes should not be too large. Nevertheless, our analysis explains the principles of the dependence of the excess noise on the mode correlations.

We find that the excess noise is the largest when the isotropic parts of the net amplification are small in comparison with the anisotropic parts of the losses. This could be verified in the experiment [9]. By choosing a more anisotropic loss and by adjusting the frequency splitting using the magnetic field accordingly, higher excess noise could be achieved. Changing the anisotropic part of the losses may also mean a change of the isotropic part of the losses. This could easily be compensated by changing the isotropic gain correspondingly, since only the net amplification counts.

The usual excess-noise predictions are completely independent of the isotropic parts of the reservoir, since these parts do not change the nonorthogonal eigenmodes. Hence, the excess noise is assumed to be maximal (formally infinite) when the Hermitian coupling of the modes by the Faraday effect equals the non-Hermitian coupling introduced by the reservoir. We find this behavior only as long as the quasimode frequency is faster than the time scales of the isotropic part of the reservoir. Otherwise the isotropic part of the reservoir dominates the stationary state and the excess noise

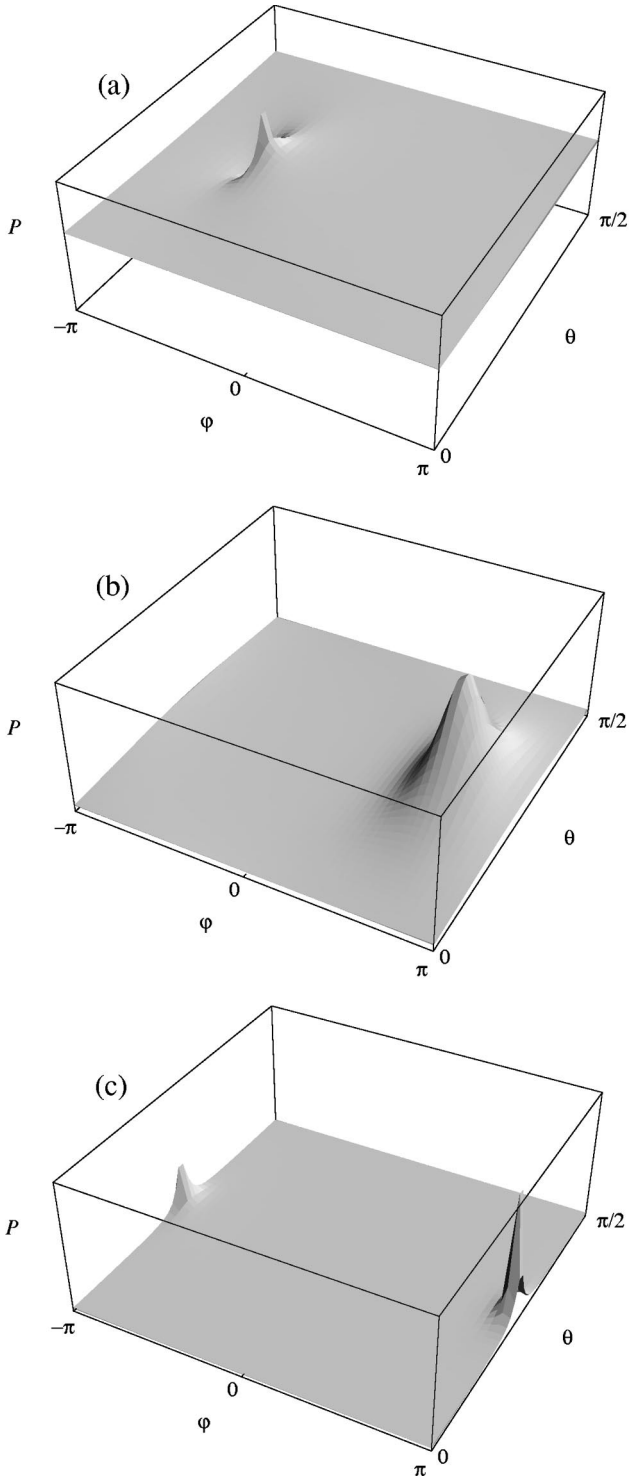


FIG. 4. Stationary P functions. In (a) we find the stationary state given by $\theta = \pi/2$ and $\varphi \approx -0.48\pi$ for the parameters $A/2 = 2.0001$, $C_h/2 = 2$, $C_v/2 = 0$, and $\Omega = 1$. The excess noise factor is then around $K \approx 1000$. In (b) and (c) $A/2 = 5$ and 36 , respectively. For increasing A , the stationary state moves to $\varphi = \pi$ with no excess noise.

becomes limited. Since at the point of maximal excess noise the quasimode frequency tends to zero, the noise never diverges.

Another interesting point is that the spectral line shape

itself does not show an excess width. The excess-noise factor appears only in the Schawlow-Townes linewidth formula where the relation of the noise to the intensity is expressed. In the usual lasers, an increase of the intensity is hence related to a decrease of the linewidth. In the case of excess noise, the intensity may increase but the linewidth remains constant. Therefore the excess-noise factor appears in the Schawlow-Townes formula.

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APPENDIX: NONLINEAR AMPLIFIER

In this section we derive the master equation for a nonlinear multimode amplifier. Here, we do not consider damping which will be added later by a separate reservoir—and hence by a separate Liouville operator in the master equation.

The free evolution of the field is given by the Hamiltonian

$$\hat{H}_{0F} = \hbar \sum_n \omega_n \hat{a}_n^\dagger \hat{a}_n. \quad (\text{A1})$$

Here, \hat{a}_n^\dagger and \hat{a}_n are the creation and annihilation operators of the various modes with frequencies ω_n and orthonormal mode functions $u_n(x)$, respectively. We use an atomic model for the reservoir as has been used in the single-mode case [14]. We assume that the light only interacts with the two levels $|e\rangle$ and $|g\rangle$ of the atoms with their corresponding Hamiltonian H_{0A} where we neglect the center-of-mass motion. The interaction is then described by the interaction Hamiltonian

$$\hat{H}_I = d \sum_n \varepsilon_n u_n(x) (\hat{a}_n^\dagger \hat{\sigma}^- + \hat{a}_n \hat{\sigma}^+), \quad (\text{A2})$$

where $\hat{\sigma}^\pm$ are the atomic transition operators and the vector d is the dipole moment of the transition.

The master equation for the total system is then given by

$$\frac{d}{dt} \hat{\rho}_{F+A}(t) = -\frac{i}{\hbar} [\hat{H}_{0F} + \hat{H}_{0A} + \hat{H}_I, \hat{\rho}_{F+A}(t)] + \mathcal{L}_P \hat{\rho}_{F+A}(t). \quad (\text{A3})$$

The Liouvillian \mathcal{L}_P causes a decay from the atomic levels $|e\rangle$ and $|g\rangle$ to some auxiliary levels with a decay rate γ . From there the population is pumped to the level $|e\rangle$ and the center-of-mass state $\hat{\rho}_{c.m.}$. We use the center-of-mass freedom of the atom only to introduce a possible position dependence $P(x) = \langle x | \hat{\rho}_{c.m.} | x \rangle$ of the reservoir. For simplicity, we neglect here diffusion, recoil, or other effects due to the motion of the atom.

The time evolution of the field state,

$$\begin{aligned} \frac{d}{dt}\hat{\rho}_F(t) = & -\frac{i}{\hbar}[\hat{H}_{0F}, \hat{\rho}_F(t)] - \frac{i}{\hbar}\text{Tr}_A\{\hat{H}_I\hat{\rho}_{F+A}(t) \\ & - \hat{\rho}_{F+A}(t)\hat{H}_I\}, \end{aligned} \quad (\text{A4})$$

is obtained from Eq. (A3) by tracing out the atomic degrees of freedom. Describing only the internal degree of freedom of the atom in the subspace of the levels $|e\rangle$ and $|g\rangle$, we can write the total master equation (A3) as

$$\begin{aligned} \frac{d}{dt}\hat{\rho}_{F+A}(t) = & -\frac{i}{\hbar}[\hat{H}_{0F} + \hat{H}_{0A} + \hat{H}_I, \hat{\rho}_{F+A}(t)] - \gamma\hat{\rho}_{F+A}(t) \\ & + r_0|e\rangle\langle e|\hat{\rho}_{c.m.}\hat{\rho}_F(t). \end{aligned} \quad (\text{A5})$$

We obtain the term $r_0|e\rangle\langle e|\hat{\rho}_{c.m.}\hat{\rho}_F(t)$ by an adiabatic elimination of the auxiliary levels where r_0 is the repumping rate to the state $|e\rangle$. Note that the normalization $\text{Tr}_{A+F}\hat{\rho}_{F+A}(t)$ is not conserved on this subspace and that $\gamma > r_0$.

Assuming that the dynamics associated with the atomic levels is much faster than the dynamics of the field, we want to eliminate adiabatically the degrees of freedom of the atomic levels. To do so, we transform Eq. (A5) into the interaction picture which removes the extremely fast free time evolution. Denoting the interaction picture by a tilde, we obtain

$$\begin{aligned} \frac{d}{dt}\tilde{\rho}_{F+A}(t) = & -\frac{i}{\hbar}[\tilde{H}_I, \tilde{\rho}_{F+A}(t)] - \gamma\tilde{\rho}_{F+A}(t) \\ & + r_0|e\rangle\langle e|\tilde{\rho}_{c.m.}\tilde{\rho}_F(t) \approx 0. \end{aligned} \quad (\text{A6})$$

This approximation means that the decay rate γ is much faster than all various detunings $\omega_n - \omega_A$ between the field modes and the transition $|e\rangle - |g\rangle$ with frequency ω_A . Consequently, we are neglecting all frequency dependences of the reservoir. Transforming back to the laboratory frame and defining the operator

$$\hat{O} = \frac{\gamma}{r_0}\hat{\rho}_{F+A}, \quad (\text{A7})$$

we get

$$\hat{O}(t) = |e\rangle\langle e|\left[\hat{\rho}_{c.m.}\hat{\rho}_F(t) - \frac{i}{\hbar\gamma}[\hat{H}_I, \hat{O}(t)]\right]. \quad (\text{A8})$$

The explicit solution of Eq. (A8) is

$$\begin{aligned} \hat{O}(t) = & \sum_{n=0}^{\infty} \left(\frac{i}{\hbar\gamma}\right)^n \sum_{m=0}^n \binom{n}{m} \\ & \times (-1)^m H_I^m |e\rangle\langle e|\hat{\rho}_{c.m.}\hat{\rho}_F(t) H_I^{n-m}. \end{aligned} \quad (\text{A9})$$

Equation (A4) does not change when we restrict ourselves to the subspace of the levels $|e\rangle$ and $|g\rangle$. Using Eqs. (A7) and (A8) we rewrite Eq. (A4) as

$$\begin{aligned} \frac{d}{dt}\hat{\rho}_F(t) = & -\frac{i}{\hbar}[\hat{H}_{0F}, \hat{\rho}_F(t)] + r_0[\text{Tr}_A\{\hat{O}(t)\} - \hat{\rho}_F(t)], \end{aligned} \quad (\text{A10})$$

which is a closed master equation for $\hat{\rho}_F(t)$ since, because of Eq. (A8), $\hat{O}(t)$ can be expressed linearly in terms of $\hat{\rho}_F(t)$.

We now simplify the master equation. We can express the trace of \hat{O} as $\text{Tr}_A\{\hat{O}\} = \int dx \langle x|(\langle e|\hat{O}|e\rangle + \langle g|\hat{O}|g\rangle)|x\rangle$. Note that from now on we drop the notation of the explicit time dependence of the operators \hat{O} and $\hat{\rho}_F$. We define the position-dependent pumping rate $r(x) = r_0\langle x|\rho_{c.m.}|x\rangle$ and the mode operator,

$$\hat{B}(x) = \frac{1}{\hbar} \sum_k \varepsilon_k(u_k(x)d)\hat{a}_k \quad (\text{A11})$$

which has the dimension of a Rabi frequency, yielding

$$\begin{aligned} r_0\langle x|\langle e|\hat{O}|e\rangle|x\rangle = & r(x) \sum_{n=0}^{\infty} (-\gamma^{-2})^n \sum_{m=0}^n \binom{2n}{2m} \\ & \times [\hat{B}(x)\hat{B}^\dagger(x)]^m \hat{\rho}_F[\hat{B}(x)\hat{B}^\dagger(x)]^{n-m} \end{aligned} \quad (\text{A12})$$

and

$$\begin{aligned} r_0\langle x|\langle g|\hat{O}|g\rangle|x\rangle = & -r(x) \sum_{n=1}^{\infty} (-\gamma^{-2})^n \sum_{m=0}^{n-1} \binom{2n}{2m+1} \hat{B}^\dagger(x) \\ & \times [\hat{B}(x)\hat{B}^\dagger(x)]^m \hat{\rho}_F[\hat{B}(x)\hat{B}^\dagger(x)]^{n-m-1} \hat{B}(x). \end{aligned} \quad (\text{A13})$$

Using the operator

$$\begin{aligned} r_0\hat{\mu}(x) = & \frac{1}{2}r(x) \sum_{n=1}^{\infty} (-\gamma^{-2})^{n-1} \sum_{m=0}^{n-1} \binom{2n}{2m+1} \\ & \times [\hat{B}(x)\hat{B}^\dagger(x)]^m \hat{\rho}_F[\hat{B}(x)\hat{B}^\dagger(x)]^{n-m-1}, \end{aligned} \quad (\text{A14})$$

we can write

$$\begin{aligned} \langle x|\langle e|\hat{O}|e\rangle|x\rangle = & \hat{\mu}(x) + \gamma^{-2}[\hat{B}(x)\hat{B}^\dagger(x)\hat{\mu}(x) \\ & + \hat{\mu}(x)\hat{B}(x)\hat{B}^\dagger(x)] \end{aligned} \quad (\text{A15})$$

and

$$\langle x|\langle g|\hat{O}|g\rangle|x\rangle = 2\gamma^{-2}\hat{B}^\dagger(x)\hat{\mu}(x)\hat{B}(x). \quad (\text{A16})$$

Hence, we find the time evolution given by the equation

$$\begin{aligned}
\hat{\rho}_F = & \hat{\mu}(x) + 2\gamma^{-2}[\hat{B}(x)\hat{B}^\dagger(x)\hat{\mu}(x) + \hat{\mu}(x)\hat{B}(x)\hat{B}^\dagger(x)] \\
& + \gamma^{-4}[\hat{B}(x)\hat{B}^\dagger(x)\hat{B}(x)\hat{B}^\dagger(x)\hat{\mu}(x) \\
& - 2\hat{B}(x)\hat{B}^\dagger(x)\hat{\mu}(x)\hat{B}(x)\hat{B}^\dagger(x) \\
& + \hat{\mu}(x)\hat{B}(x)\hat{B}^\dagger(x)\hat{B}(x)\hat{B}^\dagger(x)] \quad (A17)
\end{aligned}$$

together with the master equation

$$\begin{aligned}
\frac{d}{dt}\hat{\rho}_F = & -\frac{i}{\hbar}[\hat{H}_{0F}, \hat{\rho}_F] + \int dx r(x)\{(\hat{\mu}(x) - \hat{\rho}_F) \\
& + \gamma^{-2}[\hat{B}(x)\hat{B}^\dagger(x)\hat{\mu}(x) + \hat{\mu}(x)\hat{B}(x)\hat{B}^\dagger(x) \\
& + 2\hat{B}^\dagger(x)\hat{\mu}(x)\hat{B}(x)]\}. \quad (A18)
\end{aligned}$$

In the limit of a linear approximation, this master equation reproduces the previously derived in Ref. [6]. In the case of one mode and a distribution of the atomic reservoir concentrated at one single point, the master equation agrees with the one introduced by Sargent, Scully and Lamb [2,14].

Approximating the implicit master equation (A18) up to the order of γ^{-4} , we find the explicit master equation

$$\begin{aligned}
\frac{d}{dt}\hat{\rho}_F = & -\frac{i}{\hbar}[\hat{H}_{0F}, \hat{\rho}_F] + \frac{1}{2} \sum_{n,m} L_{m,n} \{2\hat{a}_n^\dagger \hat{\rho}_F \hat{a}_m - \hat{a}_m \hat{a}_n^\dagger \hat{\rho}_F \\
& - \hat{\rho}_F \hat{a}_m \hat{a}_n^\dagger\} + \sum_{n,m,k,l} \mathcal{L}_{n,m,k,l} \{\hat{\rho}_F \hat{a}_n \hat{a}_m^\dagger \hat{a}_k \hat{a}_l^\dagger \\
& - 4\hat{a}_l^\dagger \hat{\rho}_F \hat{a}_n \hat{a}_m^\dagger \hat{a}_k + 6\hat{a}_k \hat{a}_l^\dagger \hat{\rho}_F \hat{a}_n \hat{a}_m^\dagger - 4\hat{a}_m^\dagger \hat{a}_k \hat{a}_l^\dagger \hat{\rho}_F \hat{a}_n \\
& + \hat{a}_n \hat{a}_m^\dagger \hat{a}_k \hat{a}_l^\dagger \hat{\rho}_F\}, \quad (A19)
\end{aligned}$$

where the matrix L describes the linear part of the amplification as defined in Eq. (5) and the tensor

$$\begin{aligned}
\mathcal{L}_{n,m,k,l} = & (\hbar\gamma)^{-4} \varepsilon_n \varepsilon_m \varepsilon_k \varepsilon_l \frac{1}{V} \int d^3x r(x) [u_n(x)d][u_m(x)d] \\
& \times [u_k(x)d][u_l(x)d] \quad (A20)
\end{aligned}$$

describes the nonlinear effects in the lowest order. Such an approximation is often called the fourth-order approximation in the literature [2].

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