

## Two-photon laser dynamics

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Degenerate as well as nondegenerate three-level two-photon laser (TPL) models are derived. In the limit of equal cavity losses for both fields, it is shown that the nondegenerate model reduces to the degenerate one. We also demonstrate the isomorphism existing between our degenerate TPL model and that of a dressed-state TPL. All these models contain ac-Stark and population-induced shifts at difference from effective Hamiltonian models. The influence of the parameters that control these shifts on the nonlinear dynamics of a TPL is investigated. In particular, the stability of the periodic orbits that arise at the Hopf bifurcation of the system and the extension of the self-pulsing domains of the system are studied, revealing the role played by the detuning and ac-Stark parameters.

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### I. INTRODUCTION

Although a definitive experimental implementation of a two-photon laser (TPL) has not been possible until very recently [1], TPL's have been the subject of continued theoretical attention since the early days of the laser era [2]. The theoretical interest of the TPL lies in the intrinsic nonlinear nature of the two-photon interaction. This fact makes this system a potential source for nonclassical light and thus the major part of the literature has been devoted to the quantum description of such a laser [3].

Contrarily there is not much work on semiclassical modeling of TPL's and, in particular, there still lacks a complete understanding of its stability and dynamical properties. In a classical paper [4] Narducci *et al.* established a semiclassical two-photon amplifier model. By assuming quasi-two-photon resonance between the field and two atomic levels (not dipolarly connected) and large detuning between the field and other transitions involving intermediate atomic levels, the variables related with the latter were removed. Nevertheless the resulting equations still retained important information concerning the intermediate levels through (i) the ac-Stark shifts, (ii) a frequency shift due to the population inversion between the two lasing levels, and (iii) a static frequency shift that depends on the intermediate levels population. If these three shifts are neglected one can speak of an *effective Hamiltonian* model since the resulting equations are the same that one obtains by considering the TPL as a two-level laser with a pure nonlinear interaction with the field. If, on the contrary, one preserves the shifts one speaks of an *exact* model since it is obtained via a *microscopic* (or *com-*

*plete*) Hamiltonian. The different TPL behavior predicted by effective and microscopic Hamiltonians has been the subject of interest from the quantum viewpoint [3(a)] but not from the semiclassical one until our recent paper [5].

The first paper devoted to the TPL dynamics seems to be that of Ovadia and Sargent [6] in which the stability of an effective Hamiltonian TPL model [7] was analyzed. They describe two important features of the TPL: (i) the nondestabilization of the trivial solution with the consequent necessity of triggering, and (ii) the existence of a Hopf bifurcation that destabilizes the steady lasing solution. This instability occurs for pump parameter values *below* that corresponding to the bifurcation as can be deduced from that paper. This is a difference from the majority of laser systems [8]. In a subsequent communication Ovadia, Sargent, and Hendow [9] did a preliminary investigation of the influence of the ac-Stark shifts on the TPL dynamics, but their results were not clearly quantified and they did not take into account the rest of the frequency shifts.

Later, Ning and Haken systematically studied the dynamic behavior of the effective Hamiltonian TPL in a series of papers. They extended the previous results of Ovadia and Sargent, pointing out the direction of the Hopf bifurcation in Ref. [10]. They also considered the influence of cavity detuning [11], studied the stability of the Hopf orbits [12], and did a preliminary study of the phase dynamics [13]. Recently, Concannon and Gauthier [14] have proposed a simplified effective model for a class-B two-photon laser with injected signal that contains some of the more relevant characteristics of the TPL.

A different kind of TPL model, namely, the dressed-state TPL, was proposed by Zakrzewski, Lewenstein, and Mossberg [15], being that model appropriate for describing the experimental situation of Gauthier *et al.* [1]. In a subsequent paper, Zakrzewski and Lewenstein [16] extended their previous model to the bad cavity limit, taking into account the

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competition between one-photon and two-photon processes. They also paid special attention to the dynamical instabilities, and showed that self-pulsing and chaos can be observed in experiments on dressed-state lasers. As will be shown, our TPL model is isomorphic to the dressed-state TPL model of these authors. Apart from those works it is also to be mentioned the study of the chaotic behavior of a two-photon micromaser model by Davidovich *et al.* [17].

Recently [5] we have proposed a microscopic Hamiltonian semiclassical TPL model and studied its emission conditions and stability properties, paying special attention to the deviations with respect to the predictions of the effective Hamiltonian models of Ovidia and Sargent and of Ning and Haken. In the present paper we largely extend our previous analysis in several directions. In Sec. II we obtain the TPL model both in the degenerate and nondegenerate cases, discussing the possible connections between them, and show that our degenerate TPL model is isomorphic to the two-photon dressed-state laser model of Zakrzewski, Lewenstein, and Mossberg [15]. We analyze the stability of the Hopf orbits in Sec. III, where also the main results of [5] concerning the stability of the steady state are summarized. In Sec. IV the global dynamic behavior of the TPL is numerically investigated. Finally the main conclusions are summarized in Sec. V.

## II. TWO-PHOTON LASER MODELS

In the first two subsections below we obtain the Maxwell-Bloch equations governing our TPL model, both in the nondegenerate case, in which pairs of photons with different frequency are created (Sec. II A), and in the degenerate case, in which pairs of photons with the same frequency are created (Sec. II B). In Sec. II C we establish the relations between both models. Finally, we demonstrate in Sec. II D the isomorphism existing between our degenerate model and that of Zakrzewski, Lewenstein, and Mossberg for a dressed-state TPL [15].

### A. Nondegenerate TPL model

The starting point is the nondegenerate cascade laser model of Ref. [18]. We consider a ring cavity filled with a three-level active medium [levels 2 (upper), 0 (intermediate), and 1 (lower), with energies  $W_j$  and transition frequencies  $\omega_{jk} = (W_j - W_k)/\hbar$  ( $j, k = 2, 0, 1$ )] that interacts with two unidirectional plane-wave, single-mode electric fields of the form  $\mathbf{E}_j(z, t) = \mathbf{e}_j E_j(t) \cos[k_j z - \omega_{cj} t - \phi_j(t)]$ , with  $k_j$  the wave number,  $\omega_{cj}$  the empty cavity mode frequency closest to the transition  $0-j$ ,  $\phi_j(t)$  a phase [ $(\omega_{cj} + \dot{\phi}_j)$  is the frequency of the field  $E_j$ ], and  $z$  the propagation direction ( $j = 1, 2$ ) (Fig. 1).

Assuming that the field  $\mathbf{E}_2$  ( $\mathbf{E}_1$ ) interacts only with the 2-0 (0-1) dipole allowed atomic transition, and in the usual rotating-wave and slowly-varying-envelope approximations, the semiclassical [19] Maxwell-Bloch equations of the laser system are [18]

$$\dot{\rho}_{22} = \gamma_{\parallel}(n_2 - \rho_{22}) - 2\alpha_2 \text{Im}(\rho_{02}), \quad (1a)$$

$$\dot{\rho}_{00} = \gamma_{\parallel}(n_0 - \rho_{00}) + 2\alpha_2 \text{Im}(\rho_{02}) - 2\alpha_1 \text{Im}(\rho_{10}), \quad (1b)$$

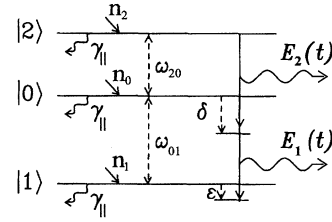


FIG. 1. The three-level cascade scheme considered in the two-photon laser model. A large value of the one-photon detunings  $\delta$  is assumed. See text for details.

$$\dot{\rho}_{11} = \gamma_{\parallel}(n_1 - \rho_{11}) + 2\alpha_1 \text{Im}(\rho_{10}), \quad (1c)$$

$$\dot{\rho}_{02} = -[\gamma_{\perp} + i(\dot{\phi}_2 + \delta_2)]\rho_{02} + i\alpha_2 d_2 + i\alpha_1 \rho_{12}, \quad (1d)$$

$$\dot{\rho}_{10} = -[\gamma_{\perp} + i(\dot{\phi}_1 + \delta_1)]\rho_{10} + i\alpha_1 d_1 - i\alpha_2 \rho_{12}, \quad (1e)$$

$$\dot{\rho}_{12} = -[\gamma_{\perp} + i(\dot{\phi}_1 + \dot{\phi}_2 + \epsilon_{12})]\rho_{12} - i\alpha_2 \rho_{10} + i\alpha_1 \rho_{02}, \quad (1f)$$

$$\dot{\alpha}_{2(1)} = -\kappa_{2(1)}\alpha_{2(1)} + g_{2(1)} \text{Im}(\rho_{02(10)}), \quad (1g)$$

$$\dot{\phi}_{2(1)} = -g_{2(1)} \text{Re}(\rho_{02(10)})/\alpha_{2(1)}, \quad (1h)$$

where  $\rho_{ii}$  represents the population of level  $i$ ,  $n_i$  being its value in the absence of fields [ $d_2 \equiv (\rho_{22} - \rho_{00})$  and  $d_1 \equiv (\rho_{00} - \rho_{11})$  are population inversions], and  $\rho_{ij}$  is the slowly varying complex amplitude of the coherence associated to the transition  $i-j$  ( $i, j = 1, 0, 2$ ) being  $\rho_{12}$  the responsible for the two-photon processes. Here we have assumed that all the populations (coherences) relax with the same rate  $\gamma_{\parallel}(\gamma_{\perp})$ .  $2\alpha_j = (\boldsymbol{\mu}_{ij} \cdot \mathbf{e}_j / 2\hbar) E_j$  is the Rabi frequency for the field  $\mathbf{E}_j$ ,  $\boldsymbol{\mu}_{0j}$  being the electric dipole element between states  $|0\rangle$  and  $|j\rangle$  that is taken to be real ( $\boldsymbol{\mu}_{12} = \boldsymbol{\mu}_{jj} = \mathbf{0}$ ).  $\kappa_j$  and  $g_j$  represent the cavity losses and the gain parameter ( $g_j = \omega_{cj} \boldsymbol{\mu}_{0j}^2 N / 2\epsilon_0 \hbar$ , with  $N$  the density of active molecules) for the field  $\mathbf{E}_j$ . The atom-cavity detunings  $\delta_j$  are defined as  $\delta_2 = \omega_{c2} - \omega_{20}$  and  $\delta_1 = \omega_{c1} - \omega_{01}$ , and  $\epsilon_{12} = \delta_1 + \delta_2 = \omega_{c2} + \omega_{c1} - \omega_{21}$  represents the two-photon cavity detuning.

This model is valid in both resonant and nonresonant situations. When resonance or small detuning is assumed we speak of a cascade laser. The emission and dynamical properties of the resonant cascade laser have been studied in Refs. [18] and [20]. The influence of moderate detuning has also been preliminarily studied in Ref. [21]. In order to obtain a pure TPL, in which the laser works without one-photon processes' contributions, some conditions must be fulfilled. Roughly speaking, these conditions imply that (i) the detuning of the fields with respect to the one-photon transitions be much larger than the relaxation rates and the two-photon cavity detuning (in order to ensure that the cavity is nearly tuned to the two-photon transition at the time that it is highly detuned from the one-photon transitions); (ii) the deviations of the field frequencies from the empty cavity frequencies be negligible in order to ensure far off-resonant one-photon processes; and (iii) the ac-Stark shifts do not modify the above far off-resonance conditions. See Refs. [22–25] for a discussion of these points.

If the above conditions are verified we can apply the adiabatic following model of Takatsuji [22] and Grischkowsky, Loy, and Liao [23]. As discussed in [24,25] it consists of the adiabatic elimination of the dipoles and of the intermediate level population. This is explicitly done in the Appendix. Then, from Eqs. (A10) and introducing the adimensional parameters

$$\begin{aligned} \chi &= \frac{g_1/\kappa_1}{g_2/\kappa_2}, \quad g = \frac{\sqrt{g_1 g_2}}{2\gamma_\perp |\delta|}, \quad n_{21} = (n_2 - n_1), \\ r &= \frac{n_{21}}{|\delta|} \left( \frac{g_1 g_2}{\kappa_1 \kappa_2} \right)^{1/2}, \quad \varepsilon = \text{sgn}(\delta) \varepsilon_{12} / \gamma_\perp, \\ \sigma_j &= \kappa_j / \gamma_\perp, \quad b = \gamma_\parallel / \gamma_\perp \end{aligned} \quad (2)$$

( $\chi$  is the relative efficiency of the two atomic transitions for one-photon amplification [20],  $g$  is the effective two-photon gain parameter,  $r$  is the pump parameter, and  $\varepsilon$ ,  $\sigma_j$ , and  $b$  are the two-photon cavity detuning, cavity losses of mode  $j$ , and population relaxation rate, normalized to the coherences relaxation rate), and adimensional variables

$$\begin{aligned} E_j &= \left( \frac{2}{\gamma_\perp |\delta| \sqrt{\chi}} \right)^{1/2} \alpha_j, \quad D = \frac{1}{2|\delta|} \left( \frac{g_1 g_2}{\kappa_1 \kappa_2} \right)^{1/2} (\rho_{22} - \rho_{11}), \\ \tau &= \gamma_\perp t, \\ Q &= \begin{cases} \frac{1}{2|\delta|} \left( \frac{g_1 g_2}{\kappa_1 \kappa_2} \right)^{1/2} \rho_{12} & \text{if } \delta > 0 \\ \frac{1}{2|\delta|} \left( \frac{g_1 g_2}{\kappa_1 \kappa_2} \right)^{1/2} \rho_{21} & \text{if } \delta < 0 \end{cases} \end{aligned} \quad (3)$$

(where  $\delta = \delta_2 \approx -\delta_1$ , since  $|\varepsilon_{12}| \ll |\delta|$  is assumed), the non-degenerate TPL equations read

$$\dot{D} = b(r - D) - E_1 E_2 \text{Im}(Q), \quad (4a)$$

$$\dot{Q} = -[1 + i(\Delta + \Omega)]Q + iE_1 E_2 D, \quad (4b)$$

$$\dot{E}_1 = \sigma_1 [E_2 \text{Im}(Q) - E_1], \quad (4c)$$

$$\dot{E}_2 = \sigma_2 [E_1 \text{Im}(Q) - E_2], \quad (4d)$$

where the derivatives are with respect to the dimensionless time  $\tau$ . Other symbols are

$$\Delta = \varepsilon - \Theta, \quad (5a)$$

$$\Theta = g(1 - 3n_0) \frac{\sigma_1 + \chi \sigma_2}{\sqrt{\chi \sigma_1 \sigma_2}}, \quad (5b)$$

$$\Omega = -\frac{\sigma_2 E_1^2 + \sigma_1 E_2^2}{E_1 E_2} \text{Re}(Q) + \frac{E_2^2 - \chi E_1^2}{2\sqrt{\chi}} - \frac{\sigma_2 - \chi \sigma_1}{\sqrt{\chi}} D. \quad (5c)$$

From Eq. (4b), and following the analysis of [5],  $\Delta$  can be interpreted as an effective atom-cavity detuning, which depends both on the actual two-photon cavity detuning and the intermediate level population through  $\Theta$ . Let us remark that

$n_0$  is usually set to zero in the literature [25] when applying the adiabatic following model. Contrarily effective TPL models neglect  $\Theta$  (implicitly assuming  $n_0 = \frac{1}{3}$  [11]). In Ref. [5] we showed that a non-null value of  $\Theta$  can deeply modify the laser behavior. Along similar lines,  $\Omega$  can be considered as an effective frequency pulling that contains an ac-Stark (second term) shift and a population difference (third term) shift.

## B. Degenerate TPL model

In this case we assume the frequencies of the two fields to be equal and we deal with the only field  $\mathbf{E}(z, t) = \mathbf{e}E(t) \cos[kz - \omega_c t - \phi(t)]$  that interacts with both the dipole-allowed atomic transitions. The Bloch equations (1a)–(1f) can be adapted to this case by substituting  $\phi_1$  and  $\phi_2$  by  $\phi$ ,  $\omega_{c1}$  and  $\omega_{c2}$  by  $\omega_c$  ( $\varepsilon_{12} = 2\omega_c - \omega_{21}$ ,  $\delta = \omega_c - \omega_{20}$ ), being now  $2\alpha_k = (\boldsymbol{\mu}_{0k} \cdot \mathbf{e} / \hbar) E$ . The field equations (1g) and (1h), however, must be substituted. The new equations are obtained by taking into account that both the coherences  $\rho_{02}$  and  $\rho_{10}$  are sources for the only field  $E$ . One easily gets

$$\dot{\alpha} = -\kappa \alpha + g_{12} [\sqrt[4]{\chi} \text{Im}(\rho_{10}) + (\sqrt[4]{\chi})^{-1} \text{Im}(\rho_{02})], \quad (6a)$$

$$\dot{\phi} = g_{12} [\sqrt[4]{\chi} \text{Re}(\rho_{10}) + (\sqrt[4]{\chi})^{-1} \text{Re}(\rho_{02})] / \alpha, \quad (6b)$$

with  $\alpha^2 = \alpha_1 \alpha_2$ , i.e.,

$$\alpha = \sqrt{(\boldsymbol{\mu}_{02} \cdot \mathbf{e})(\boldsymbol{\mu}_{10} \cdot \mathbf{e})} E / 2\hbar \quad (7a)$$

and

$$g_{12} = \omega_c |\boldsymbol{\mu}_{02}| |\boldsymbol{\mu}_{10}| N / 2\varepsilon_0 \hbar. \quad (7b)$$

In this case the relative efficiency for two-photon amplification  $\chi$  takes the simpler form  $\chi = \mu_{01}^2 / \mu_{20}^2$ .

In order to obtain the degenerate TPL model we must consider again the conditions that ensure large one-photon detuning enunciated in the preceding subsection for the non-degenerate case (see Appendix). Then, from Eqs. (A14), defining the adimensional parameters

$$g = \frac{g_{12}}{\gamma_\perp |\delta|}, \quad r = \frac{g_{12} n_{21}}{\kappa |\delta|}, \quad \sigma = \frac{\kappa}{\gamma_\perp}, \quad (8)$$

and by introducing the adimensional variables

$$I = \frac{2\alpha^2}{\gamma_\perp |\delta|}, \quad D = \frac{g_{12}}{\kappa |\delta|} (\rho_{22} - \rho_{11}),$$

$$Q = \begin{cases} \frac{2g_{12}}{\kappa |\delta|} \rho_{12} & \text{if } \delta > 0 \\ \frac{2g_{12}}{\kappa |\delta|} \rho_{21} & \text{if } \delta < 0, \end{cases} \quad (9)$$

the degenerate TPL model equations read [5]

$$\dot{D} = b(r - D) - \text{Im}(Q)I, \quad (10a)$$

$$\dot{Q} = -[1 + i(\Delta + \Omega)]Q + iDI, \quad (10b)$$

$$\dot{I} = 2\sigma I[\text{Im}(Q) - 1], \quad (10c)$$

$$\Omega = -2\sigma \text{Re}(Q) + \eta(I - 2\sigma D), \quad (10d)$$

where the derivatives are with respect to  $\tau$ . Parameters  $\varepsilon$  and  $b$  keep the same definitions as in the nondegenerate case [Eqs. (2b) and (5a)] and

$$\Delta = \varepsilon - \Theta = \varepsilon - g(1 - 3n_0) \frac{1 + \chi}{\sqrt{\chi}}. \quad (11a)$$

The ac-Stark parameter  $\eta$  has been defined as

$$\eta = \frac{1 - \chi}{2\sqrt{\chi}}. \quad (11b)$$

Notice that the definitions (8) and (9) are equivalent to those corresponding to the nondegenerate case [definitions (2) and (3)] except for a factor 2 in the gain parameter  $g$  (we keep the same symbols for both models to not complicate the notation unnecessarily) and in the definitions of  $D$ ,  $Q$ , and  $r$ . It is also to be pointed out that in the degenerate case [Eqs. (10)] only the field intensity is relevant, whereas in the nondegenerate case [Eqs. (4)] the field amplitudes  $E_j$  cannot be substituted by their intensities.

The connection between  $\chi$  and the ac-Stark shifts appearing in Eq. (10d) is now evident: when  $\chi = 1$  [ $\eta = 0$ , Eq. (11b)] the ac-Stark ( $\eta I$ ) and the population ( $-2\eta\sigma D$ ) shifts become null. Contrarily, in the nondegenerate case,  $\chi = 1$  does not eliminate the ac-Stark shifts [Eq. (5c)] unless  $\sigma_1 = \sigma_2$  (see next subsection). On the other hand the static frequency shift  $\Theta$  becomes null for  $n_0 = \frac{1}{3}$  in both the degenerate and nondegenerate cases [Eqs. (5b) and (11a)].

When  $\eta = 0$  (i.e.,  $\chi = 1$ ) and  $\Theta = 0$  (i.e.,  $n_0 = \frac{1}{3}$ ) Eqs. (10) are isomorphic to the Ning and Haken effective Hamiltonian TPL model [10]. Thus, the influence of the intermediate level manifests through the ac-Stark frequency shift  $\eta I$ , the population inversion frequency shift  $-2\sigma\eta D$ , and the static frequency shift  $\Theta$ , their strength being a function of the parameters  $\eta$  (or  $\chi$ , which is a more intuitive physical parameter) and the intermediate level population  $n_0$ .

### C. The degenerate TPL model as a limiting case of the nondegenerate model

The degenerate TPL model constitutes a limiting case of the nondegenerate one. In effect, if we take equal losses for both fields in the nondegenerate model of Eqs. (4), it is trivial to show that  $E_1^2(t) = E_2^2(t) \equiv I(t)$  asymptotically. Then we can rewrite Eqs. (4) and (5) in terms of the only intensity  $I(t)$ . What we obtain are Eqs. (10), i.e., the degenerate model. Thus, *the laser behavior predicted from a nondegenerate TPL model with equal cavity losses for both fields coincides with that predicted from a degenerate TPL model.*

Nevertheless there is a difference in a factor 2 in the gain parameter that makes the pump parameter different in both cases:  $r_{\text{deg}} = 2r_{\text{nondeg}}$  [Eqs. (2) and (8)]. In particular this implies that the minimum population inversion  $n_{21}$  required for laser action in the nondegenerate case must be twice that in the degenerate case. This fact was first commented by Scza-

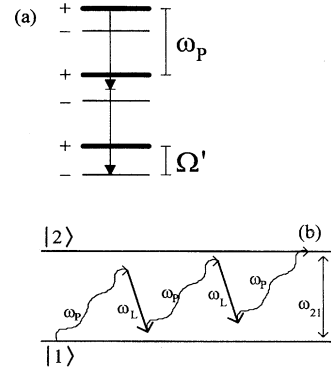


FIG. 2. (a) Dressed-state two-photon laser scheme. + and - represent the dressed states (the thickness of the line is indicative of the level population).  $\omega_p$  and  $\Omega'$  are the driving field and generalized Rabi frequencies, respectively. (b) The equivalent multiphoton-Raman semiclassical scheme, where  $\omega_L$  is the lasing frequency.

niecki when analyzing the steady solution of the TPL [26], and was interpreted by Wang and Haken [27]. What occurs is that the validity conditions of both models do not coincide: in the degenerate case the field interacts with both atomic transitions, whereas in the nondegenerate case each field interacts with only one transition. This explains, roughly, the factor 2 in the gain parameter.

In any case, this is not relevant when studying the dynamic behavior of the TPL. Moreover, in the following we will restrict to the degenerate model of Eqs. (10) for the sake of simplicity.

### D. Isomorphism between the degenerate and the dressed-state TPL models

Up to now the only experimental verification of a TPL has been carried out in dressed-atom doublets [1]. This laser consists of an atomic beam of two-level atoms which are strongly driven by a dressing or pump field. If the pump field is detuned from the atomic transition then the atom plus field quantum states form a ladder of doublets such that the populations of the two states of each doublet are different in general. If a high- $Q$  cavity is suitably tuned one can obtain one-photon [28,29] or two-photon [1] lasing between some of the states [Fig. 2(a)]. From the semiclassical viewpoint this TPL system can be understood as a multiphoton-Raman laser in which the atom passes from the lower to the upper lasing level through the absorption of three photons of the pump (dressing) field and the emission of two photons of the lasing field [Fig. 2(b)]. An appropriate model for describing such a system is that of Zakrzewski, Lewenstein, and Mossberg [15] and in this subsection we will show that our degenerate TPL model [Eqs. (10)] is isomorphic to their dressed-state TPL model. This constitutes an extension to the two-photon case of the isomorphism existing between one-photon dressed-state lasers and normal one-photon (two-level) lasers [30].

The equations describing the dressed-state TPL model read [15]

$$\dot{S} = -(\gamma_1 + i\Omega')S - i\Lambda_1 S_3 a^2 - 2i\Lambda_2 S(|a|^2 + \frac{1}{2}), \quad (12a)$$

$$\dot{S}_3 = -\gamma_2(S_3 - \bar{S}_3) - 2i\Lambda_1[(a^*)^2 S - S^* a^2], \quad (12b)$$

$$\dot{a} = -(\Gamma + i\Delta_2)a - 2i\Lambda_1 S a^* - i\Lambda_2 S_3 a, \quad (12c)$$

where  $S$ ,  $S_3$ , and  $a$  are proportional to the medium polarization, medium inversion, and field amplitude, and  $\gamma_1$ ,  $\gamma_2$ , and  $\Gamma$  are their respective damping constants.  $\bar{S}_3$  is the pump parameter,  $\Omega' = (\Omega^2 + \Delta_1^2)^{1/2}$  with  $\Omega$  the Rabi frequency of the pump (dressing) field,  $\Delta_1 = (\omega_{21} - \omega_p)$  and  $\Delta_2 = (\omega_c - \omega_p)$ ,  $\omega_p$  being the frequency of the dressing field and  $\Delta_2$  that of the cavity mode.  $\Lambda_1$  and  $\Lambda_2$  are defined as

$$\Lambda_1 = \frac{g^2}{4\Omega'} \sin 2\alpha (1 + \cos 2\alpha), \quad (13a)$$

$$\Lambda_2 = \frac{g^2}{8} \left[ \frac{(1 + \cos 2\alpha)^2}{\Omega'} + \frac{(1 - \cos 2\alpha)^2}{4\Delta_2 + \Omega'} + \frac{2 \sin^2 2\alpha (\Omega' - 2\Delta_2)}{\Omega' (4\Delta_2 - \Omega')} \right], \quad (13b)$$

with  $g$  the gain parameter and  $\alpha$  the ‘‘rotation angle’’ defined through  $\Omega = \Omega' \sin 2\alpha$  and  $\Delta_1 = \Omega' \cos 2\alpha$ . It must be verified that  $2\Delta_2 \approx \Omega'$ .

Let us note that  $\gamma_1$ ,  $\gamma_2$ , and  $\bar{S}_3$  depend on the pump strength through the rotation angle

$$\bar{S}_3 = \frac{-2N \cos 2\alpha}{1 + \cos^2 2\alpha}, \quad \gamma_1 = \frac{1}{2} \gamma (2 + \sin^2 2\alpha), \quad (14)$$

$$\gamma_2 = \gamma (1 + \cos^2 2\alpha),$$

$\gamma$  being the relaxation rate of the bare atom occupation probabilities.

With the change of variables

$$I = \frac{2\Lambda_1}{\gamma_1} [a e^{i(\Delta_2 t + \phi)}]^2, \quad D = \frac{\Lambda_1}{\gamma_1 \Gamma} S_3, \quad (15)$$

$$Q = \frac{2\Lambda_1}{\gamma_1 \Gamma} [S^* e^{-2i(\Delta_2 t + \phi)}]^2,$$

and of parameters

$$r = \frac{\Lambda_1}{\gamma_1 \Gamma} \bar{S}_3, \quad \eta = -\frac{\Lambda_2}{\Lambda_1}, \quad \Delta = \frac{2\Delta_2 - \Omega'}{\gamma_1}, \quad (16)$$

$$\sigma = \frac{\Gamma}{\gamma_1}, \quad b = \frac{\gamma_2}{\gamma_1},$$

and assuming  $|a^2| \gg \frac{1}{2}$  ( $|a|^2$  is the photon number expected value), one recovers Eq. (10) of the degenerate TPL model.

Since  $\Delta$  does not contain now the  $\Theta$  contribution [compare Eqs. (16) and (11a)],  $n_0 = \frac{1}{3}$  here, and this laser is less sensitive to pumping through increasing gain (e.g., increasing the density of atoms) than a three-level two-photon laser (see later in Sec. III A).

Although the results that we present in the following are applicable to the dressed-state TPL, the relations (16) between both systems' parameters prevent a straightforward translation of the results since we assume that the parameters

keep constant while the pump is varied, and this cannot be applied to the dressed-state TPL.

### III. STABILITY PROPERTIES

In this section we address the problem of the stability properties of the degenerate TPL model. In Sec. III A we resume the linear stability since it was treated in detail [5,6,10,11]. In Sec. III B we study the stability of the Hopf orbits paying particular attention to the influence of  $\chi$  and  $\Delta$ .

#### A. Steady states and linear stability

It is straightforward to show that the trivial solution of the TPL is always stable. Thus it is necessary to trigger the laser for reaching the nontrivial steady solution, as has been demonstrated experimentally [1]. The lasing solution of Eqs. (10) is given by (we only give its intensity  $\bar{I}$ )

$$c_1 \bar{I}^2 + c_2 \bar{I} + c_3 = 0, \quad (17a)$$

with

$$c_1 = 1 + \frac{1}{b} \left( \frac{2\sigma + b}{2\sigma + 1} \right)^2 \eta^2, \quad (17b)$$

$$c_2 = - \left[ b + 4\sigma - \frac{2\sigma + b}{(2\sigma + 1)^2} \eta^2 \right] r + 2 \frac{2\sigma + b}{(2\sigma + 1)^2} \eta \Delta, \quad (17c)$$

$$c_3 = b \left[ 1 + \left( \frac{\Delta - 2\sigma \eta r}{2\sigma + 1} \right)^2 \right]. \quad (17d)$$

This stationary intensity has two branches: in one of them the intensity grows with increasing pump, and in the other branch the intensity has an opposite behavior. Both branches appear at a pump value  $r_0$  that is given by

$$d_1 r_0^2 + d_2 r_0 + d_3 = 0, \quad (18a)$$

$$d_1 = b^2 [(1 + 2\sigma)^2 + 8\sigma \eta^2], \quad (18b)$$

$$d_2 = 4b \eta \Delta (2\sigma - b), \quad (18c)$$

$$d_3 = -4b [(1 + 2\sigma)^2 + \Delta^2] - 4\eta^2 (b + 2\sigma)^2 \quad (18d)$$

(some of these coefficients were incorrectly given in Ref. [5]).

The most relevant new feature in the steady solution with respect to the effective Hamiltonian models not considering the intermediate level is its dependence on the static shift  $\Theta$ . As  $\Theta$  depends on the gain parameter  $g$ , if the pump mechanism involves a variation in the density of active molecules (for example by changing the gas pressure) then the shift  $\Theta$  becomes pump dependent, and so does the detuning  $\Delta$ . This can occur but in the case  $n_0 = \frac{1}{3}$  for which  $\Theta = 0$ . This fact strongly influences the emission threshold and even can prevent laser action [5]. In the following we assume that the pump mechanism involves variations only in  $n_{21}$ , i.e., in the inversion in the absence of fields.

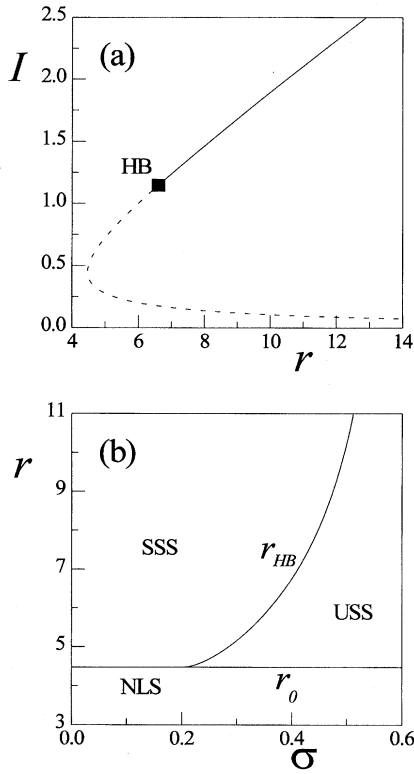


FIG. 3. (a) Steady intensity vs pump for  $\sigma=4$  and  $b=0.2$ . The continuous (dashed) curve denotes stable (unstable) solution. (b)  $r_{HB}$  vs  $\sigma$  for  $b=0.2$ . Both figures correspond to the resonant ( $\varepsilon=0$ ) effective limit ( $\chi=1$ ,  $\Theta=0$ ) TPL model. SSS, stable steady state; USS, unstable steady state; NLS, no lasing solution.

With respect to the stability of the lasing solutions, the one decreasing with increasing pump is always unstable and the other one can undergo a Hopf bifurcation (HB). The most relevant feature of this HB is that, at a difference from the majority of lasers [8], the laser is stable for pump values larger than (and unstable for values smaller than) that corresponding to the HB,  $r_{HB}$ . An explicit expression of  $r_{HB}$  can only be given for the effective limit of the model (i.e., for  $n_0=\frac{1}{3}$  and  $\chi=1$ ) and it was first obtained by Ovadia and Sargent [6]. In the perfectly tuned case ( $\Delta=0$ ) it reads

$$r_{HB} = \frac{2\sigma(1-b)}{b\sqrt{(1+b-2\sigma)(2\sigma-b-b^2)}}, \quad (19)$$

whenever the *bad cavity* condition  $\sigma > \sigma_{BCL} = b$  is verified (Fig. 3). Notice that  $\sigma_{BCL}$  is much smaller than that corresponding to the one-photon two-level laser (Lorenz-Haken model [31]). On the other hand the steady solution is always unstable for  $\sigma > \sigma_a \equiv (1+b)/2$  since  $r_{HB}$  has an asymptote for this value of  $\sigma$ . In the particular case  $b=1$ , the steady solution is stable for  $\sigma < 1$  and unstable for  $\sigma > 1$  irrespective of the pump value (the Hopf bifurcation disappears). It is to be marked that instabilities are still possible in the case of adiabatic elimination of the medium polarization  $Q$  (class- $B$  laser) [14]. In this case the asymptote  $\sigma_a = \frac{1}{2}$  and cannot be

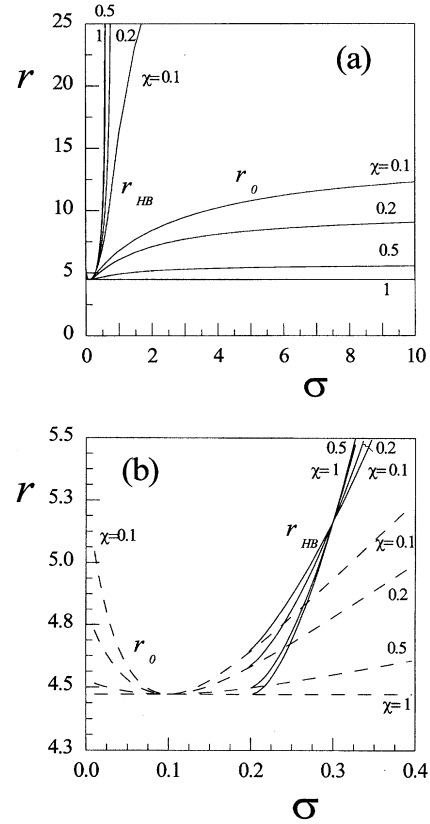


FIG. 4. Dependence of  $r_0$  and  $r_{HB}$  with  $\sigma$  for  $b=0.2$  and several values of  $\chi$ . In (b) the intersection zone is magnified.

reached since  $\sigma, b \ll 1$  is assumed in the class- $B$  model. This fact was not clearly identified in Ref. [14].

The dependence of  $r_{HB}$  with the ac-Stark parameter and the detuning was studied in detail in Ref. [5]. There we showed that  $\chi$  influences smoothly the position of  $r_{HB}$  (as well as that of its asymptote  $\sigma_a$ ) when  $0.15 \leq \chi \leq 1/0.15$  and that  $\sigma_{BCL}$  is almost not affected. For smaller values of  $\chi$  a new feature appears since the asymptote  $\sigma_a$  disappears (Fig. 4). With respect to the influence of  $\Delta$  on  $r_{HB}$  (previously treated by Ning [11] with the effective Hamiltonian model) it was shown that the instability domain tends to increase with increasing  $\Delta$  and it can be proven that the asymptote  $\sigma_a$  remains unaltered. For  $\chi \neq 1$  there is an asymmetry of both  $r_0$  and  $r_{HB}$  with respect to positive and negative  $\Delta$  (Fig. 5).

### B. Stability of the Hopf orbits

Now we address the problem of determining the stability of the Hopf orbits. For that we make use of a reelaboration of the general method of Iooss and Joseph [32] that we carried out in Ref. [20]. It consists basically in expanding the Hopf orbits close to the bifurcation (roughly speaking in terms of their amplitude) and imposing a solvability condition (Fredholm alternative). This last condition together with the Floquet theory of the stability of periodic orbits provides a criterium for the stability of the orbits. In Ref. [12] Ning and Haken addressed the same problem in the effective limit of the perfectly tuned model by applying the normal form and

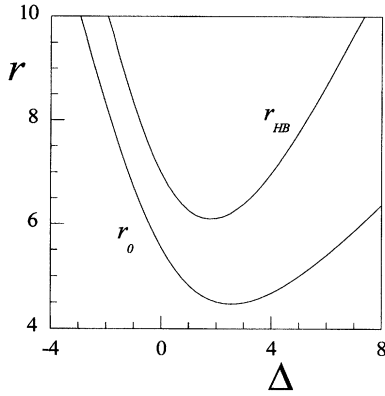


FIG. 5. Dependence of  $r_{HB}$  with  $\Delta$  for  $b=0.2$ ,  $\sigma=0.5$ , and  $\chi=0.1$ .

slaving principle. Our calculations extend their results, reproducing them in the appropriate limit ( $\chi=1$ ,  $n_0=\frac{1}{3}$ , and  $\varepsilon=0$ ). In particular they showed that when the bifurcation is subcritical (i.e., that the orbits are unstable in the vicinity of the bifurcation) the laser turns off, i.e., when the orbit is unstable at the bifurcation the whole branch of the Hopf orbits is always unstable. So the knowledge of the subcritical or supercritical character of HB provides a large amount of information in this laser model.

By defining  $\mathbf{u}=\mathbf{U}-\mathbf{U}_0$  with  $\mathbf{U}=(I, \text{Re}Q, \text{Im}Q, D)$  and  $\mathbf{U}_0$  the stationary value of  $\mathbf{U}$ , and choosing an arbitrary control parameter  $\mu$ , Eqs. (10) can be written as

$$\frac{d}{dt} \mathbf{u} = \underline{L}(\mu) \mathbf{u} + \frac{1}{2} \mathbf{N}(\mu; \mathbf{u}, \mathbf{u}), \quad (20)$$

where  $\underline{L}$  is the linear matrix (of elements  $L_{ij}$ ) and  $\mathbf{N}$  is the nonlinear vector (with components  $N_i$ ), which are given by

$$\underline{L}_{ij}(\mu) = \left[ \frac{\partial}{\partial u_j} \left( \frac{du_i}{d\tau} \right) \right]_{\mathbf{u}=0}, \quad (21a)$$

$$N_i(\mu; \mathbf{a}, \mathbf{b}) = \sum_{k,l=1}^4 a_k b_l \left[ \frac{\partial^2}{\partial u_k \partial u_l} \left( \frac{du_i}{d\tau} \right) \right]_{\mathbf{u}=0}, \quad (21b)$$

$\mathbf{a}$  and  $\mathbf{b}$  being arbitrary four-dimensional vectors (we do not give the explicit expressions of  $\underline{L}$  and  $\mathbf{N}$  for the sake of brevity). In general, the expansion (20) contains higher-order nonlinearities that should be taken into account. Nevertheless Eqs. (10) only contain second-order nonlinearities and thus (20) is exact in our case.

In Ref. [20] we showed that the Hopf bifurcation is supercritical (subcritical) and consequently the Hopf orbits are stable (unstable) in the vicinity of HB whenever the quantity

$$\Sigma = \text{Re}\{\mathbf{l} \cdot [\mathbf{N}(\mu_{HB}; \mathbf{r}, \mathbf{u}_{20}) + \mathbf{N}(\mu_{HB}; \mathbf{r}^*, \mathbf{u}_{22})]\} \quad (22)$$

is negative (positive). In Eq. (22),  $\mathbf{r}$  and  $\mathbf{l}$  are the right and left eigenvectors of  $\underline{L}$  at HB corresponding to the pure imaginary eigenvalue  $i\omega_0$ , and  $\mathbf{u}_{2j}$  ( $j=0,2$ ) are Fourier components of the second-order terms of the Taylor expansion of the Hopf orbits and are obtained from

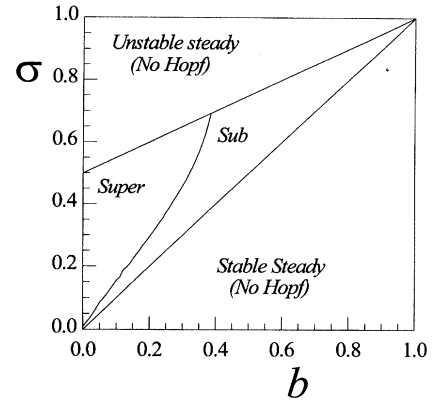


FIG. 6. Domains of stability of the Hopf orbits for  $\chi=1$  and  $\Delta=0$  in the cavity losses  $\sigma$  vs population decay rate  $b$  plane. The value of  $r_{HB}$  is given by Eq. (19).

$$[\underline{L}(\mu_{HB}) - 2i\omega_0\delta] \mathbf{u}_{22} = -\mathbf{N}(\mu_{HB}; \mathbf{r}, \mathbf{r}), \quad (23a)$$

$$\underline{L}(\mu_{HB}) \mathbf{u}_{20} = -2\mathbf{N}(\mu_{HB}; \mathbf{r}, \mathbf{r}^*), \quad (23b)$$

with  $\mu_{HB}$  the value of the control parameter  $\mu$  at the Hopf bifurcation,  $\delta$  the  $n \times n$  identity matrix, and  $n$  the dimension of the problem ( $n=4$  in our case). We address the reader to the Appendix of Ref. [20] and to the book of Iooss and Joseph [32] for details.

An explicit expression for  $\Sigma$  cannot be obtained in general. Hence we will make a numerical study of it. In Fig. 6 the different domains of stability of the Hopf orbits are displayed in the plane  $(b, \sigma)$  for the perfectly tuned effective limit of Eqs. (10), i.e.,  $\chi=1$  and  $\Delta=0$ . This is the case investigated by Ning and Haken [12] and we have verified that our results are exactly coincident. Three main domains can be clearly appreciated in the figure: a domain of stable steady state on the lower right-hand side (that corresponds to  $\sigma < \sigma_{BCL}$ ), a domain in which the steady solution is always unstable on the upper left-hand side (that corresponds to  $\sigma < \sigma_a$ ), and the domain of dynamic behavior between the other two. This last domain is subdivided into two zones corresponding to stable (unstable) Hopf orbits in the left (right) side. Thus, for  $b > 0.4$  the Hopf orbits are always unstable and the laser turns off when the Hopf bifurcation is trespassed (with decreasing pump  $r$ ), and for  $b < 0.4$  the orbits are stable if  $\sigma$  is larger than a certain value and unstable below it. Let us finally point out that when HB is degenerated (i.e., when it passes from subcritical to supercritical) the Hopf orbits do not exist as already noted in Ref. [12].

In Fig. 7 we show analogous representations for two representative values of the detuning  $\Delta$  keeping  $\chi=1$ . A continuous and smooth deformation of the border between the subcritical and supercritical domains can be appreciated and it tends to increase the domain of stable orbits. It can also be appreciated that instabilities can be observed for cavity losses below the *bad cavity condition* as  $\Delta$  is increased, as already stated by Ning [11]. Notice also how the asymptote  $\sigma_a$  cannot be trespassed, as commented above.

In Fig. 8 we go back to resonance conditions ( $\Delta=0$ ) and we show the dependence of the extension of the domains for

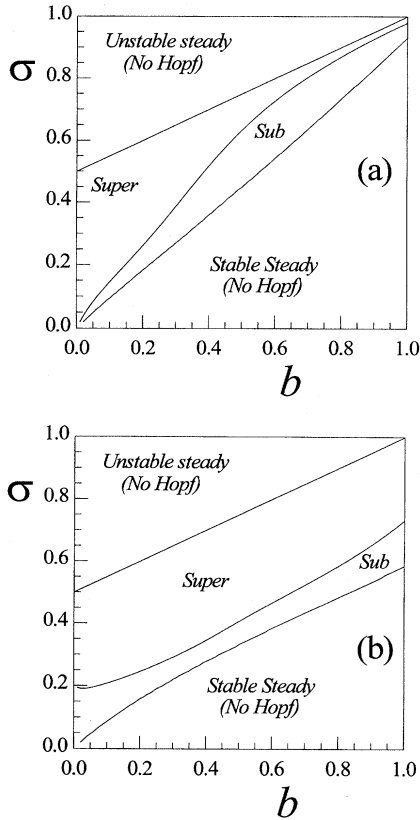


FIG. 7. Same as Fig. 6 for  $\chi=1$  and two values of  $\Delta$ : (a)  $\Delta=1$  and (b)  $\Delta=4$ .

three values of the ac-Stark parameter  $\chi$ . As in the case of  $\Delta$  a deformation of the domains of supercritical and subcritical Hopf bifurcations is observed. The most relevant feature is that for  $\chi$  different enough from 1, there is no value of  $b$  for which the bifurcation is always subcritical. There is always a value of  $\sigma$  for which it becomes supercritical, at difference from the perfectly tuned effective case (Fig. 6). For very low values of  $\chi$  [Fig. 8(c)] there is a “splitting” of the supercritical domain for low values of  $b$  into two domains separated by a subcritical zone. In this case instabilities for cavity losses larger than the asymptote  $\sigma_a$  can be found, as already commented.

#### IV. DYNAMIC BEHAVIOR

The dynamical instabilities of the TPL have been investigated in the past by Ning and Haken in the effective model [12] and by Davidovich *et al.* in a two-photon micromaser model [17]. In both cases it was found that the periodic orbits follow a classical Feigenbaum route to chaos through period doubling with decreasing pump.

We study here quantitatively the extension in the parameter space of the different domains of periodic and chaotic behavior and the influence of  $\chi$  and  $\Delta$  on them. We have chosen a constant value of  $b=0.2$  which is not singular and represents qualitatively other cases as we have tested.

Figure 9(a) shows the domain of self-pulsing behavior on the plane  $\langle \sigma, r \rangle$  in the resonant effective Hamiltonian TPL

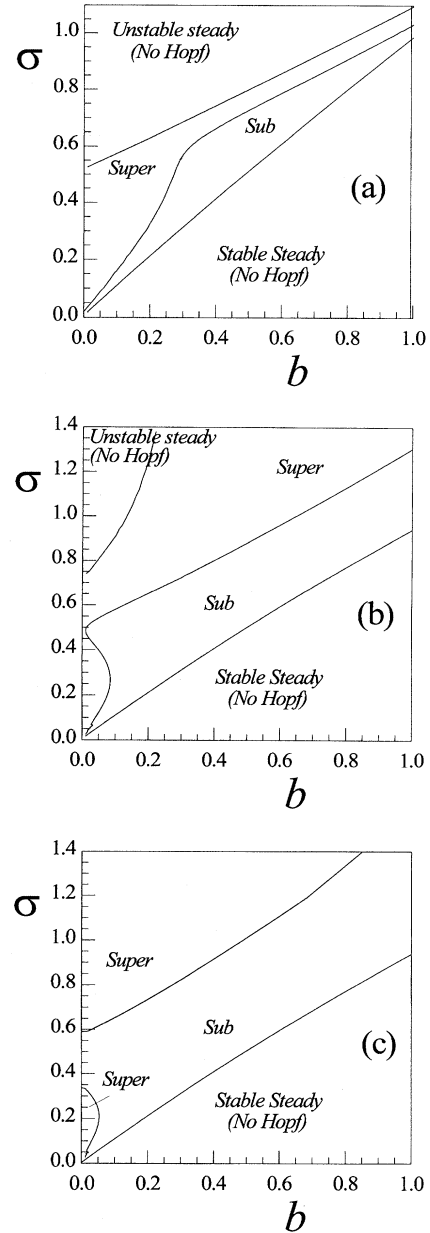


FIG. 8. Same as Fig. 6 for  $\Delta=0$  and several values of  $\chi$ : (a)  $\chi=0.5$ ; (b)  $\chi=0.15$ , and (c)  $\chi=0.1$ .

limit ( $\chi=1$  and  $\Delta=0$ ). This domain is delimited by the curves  $r_{HB}$  and  $r_{off}$ . A surprising fact is the small width of the domain. It could be expected that the dynamic behavior should increase in complexity as  $r$  decreases from  $r_{HB}$ , but the strongly stable zero-intensity solution attracts the system's trajectory in the phase space and laser emission turns off at  $r_{off}$ . Thus, most of dynamic behavior in Fig. 9(a) corresponds to single-orbit periodic emission (period-1 attractor, or  $P_1$ ). Complex behavior (periodlike or chaoticlike) found below  $r_{off}$  is metastable. Then, self-pulsing instabilities tend to switch off the laser. On the other hand the inexistence of dynamic behavior when the HB is subcritical (i.e., for  $\sigma < 0.32ca$  in this case, see Fig. 6) is to be pointed out, as we stated above in general.



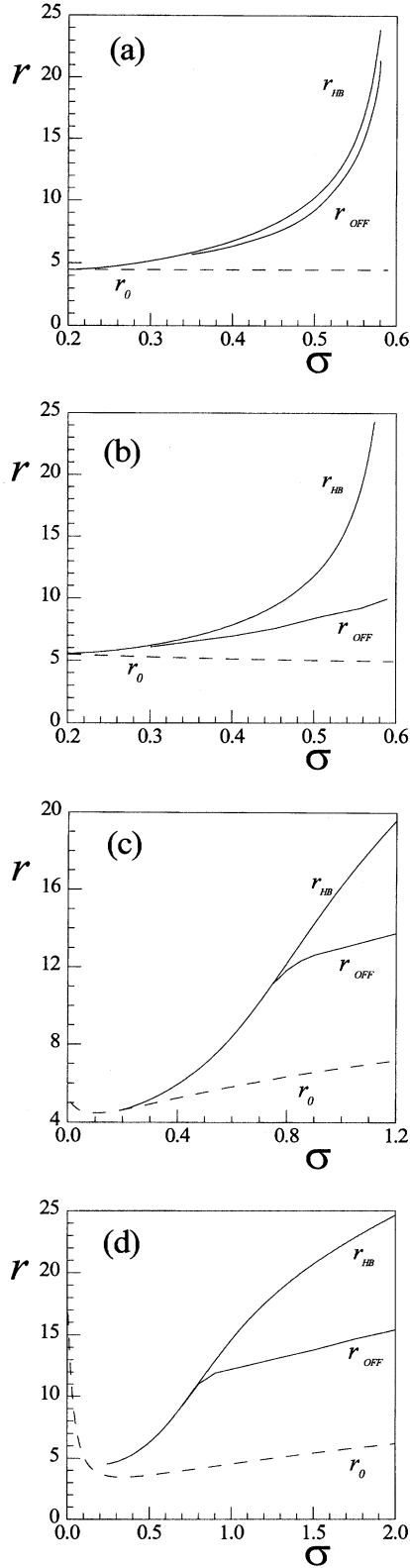


FIG. 9. Dynamic domain in the plane  $r$  vs  $\sigma$  for  $b=0.2$  and several values of  $\chi$  and  $\Delta$ : (a)  $\Delta=0, \chi=1$ ; (b)  $\Delta=1, \chi=1$ ; (c)  $\Delta=0, \chi=0.1, \chi=0.1$ , and (d)  $\Delta=1, \chi=0.1$ .

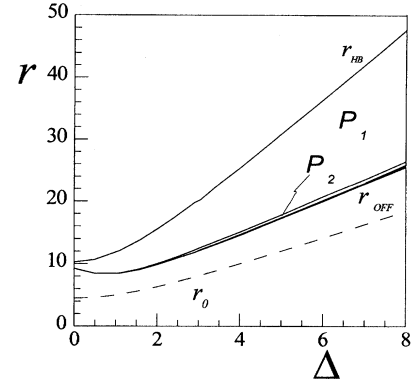


FIG. 10. Dynamic domain in the plane  $r$  vs  $\Delta$  for  $b=0.2, \sigma=0.5$ , and  $\chi=1$ .

The influence of the detuning  $\Delta$  and ac-Stark parameter  $\chi$  (or  $\eta$ ) is shown in the rest of Fig. 9:  $\Delta=1$  and  $\chi=1$  (b),  $\Delta=0$  and  $\chi=0.1$  (c), and  $\Delta=1$  and  $\chi=0.1$  (d). Clearly, making  $\Delta \neq 0$  or  $\chi \neq 1$  increases the domain of self-pulsing behavior: the  $r_{OFF}$  curve shifts down approaching the first laser threshold  $r_0$ . The only differences between varying  $\Delta$  or  $\chi$  are on the behavior of  $r_{HB}$  and on the fact that the value of  $\sigma$  for which HB passes from subcritical to supercritical is different in both cases [see Figs. 7(a) and 8(d)].

If a fixed value of  $\sigma$  and  $b$  is chosen ( $\sigma=0.5, b=0.2$ ) the extension of the self-pulsing domains as a function of  $\chi$  or  $\Delta$  can be easily analyzed. For this particular value of  $\sigma$ ,  $r_{HB}$  is always supercritical for  $\chi=1$  irrespective of the value of  $\Delta$  (Fig. 7) and the effect of varying  $\Delta$  is to increase the extension of the self-pulsing domains, as can be clearly appreciated in Fig. 10 [the behavior for negative  $\Delta$  is the same due to the symmetry properties of Eqs. (10) for  $\eta=0$ ]. Even the period-doubling zones can be partially appreciated if  $\Delta$  is large enough but the chaotic domains continue being very narrow in  $r$ . This case exemplifies well what normally happens when  $\Delta$  is increased: zones that are supercritical for  $\Delta=0$  continue being supercritical while most of the zones that are subcritical in resonance become supercritical for large  $\Delta$  (see Fig. 7).

Contrary to the case of  $\Delta$ , as  $\chi$  is varied from 1 the subcritical or supercritical character of  $r_{HB}$  normally changes (Fig. 8). Thus, for a fixed value of  $\Delta$  and  $\sigma$ , variations of  $\chi$  from 1 usually tend to lead the system from no self-pulsing to a self-pulsing domain (and vice versa), the larger the domain in  $r$  the larger  $|\eta|$ . In this sense, Fig. 11 illustrates how the dynamic domain diminishes as  $\chi$  becomes different from 1 (and even disappears), for  $\sigma=0.5$  and  $\Delta=1$ . If the value  $\Delta=0$  had been chosen the figure would be symmetric with respect to  $\chi=1$  due to the symmetry properties of Eqs. (10) [5]. Contrarily in the case of Fig. 11 the system does not behave symmetrically for  $\chi > 1$  and  $\chi < 1$ .

## V. CONCLUSIONS

In this paper we have derived a two-photon laser (TPL) model from a cascade laser model [18,20,21] by applying the adiabatic following approximation [22–24]. Our model generalizes previous effective Hamiltonian models [6,10] since

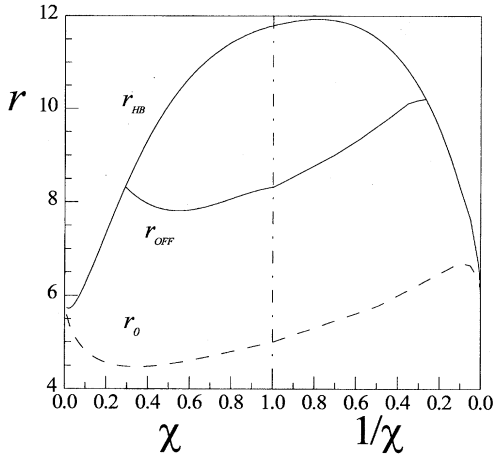


FIG. 11. Dynamic domain in the planes  $r$  vs  $\chi$  and  $r$  vs  $1/\chi$  for  $b=0.2$ ,  $\sigma=0.5$ , and  $\Delta=1$ .

it contains information about the intermediate level through its population  $n_0$  and the ac-Stark parameter  $\eta$ . We have concentrated on the influence of these two parameters on the TPL model stability and dynamical properties.

We have shown that the presence of  $n_0$  and  $\eta$  is essential for demonstrating the isomorphism that exists between our TPL model and the dressed-state TPL model of Zakrzewski, Lewenstein, and Mossberg [15], which applies to the experimental situation of Gauthier *et al.* [1]. Another isomorphism existing between the degenerate (one frequency) TPL and the nondegenerate (two frequencies) TPL with equal losses for both fields has also been demonstrated.

From the point of view of the stability properties of the degenerate model we have discussed the influence of  $\eta$  and  $\Delta$  on the Hopf bifurcation, HB, that can affect the lasing solution. This HB has unusual properties in the sense that (i) the laser is stable for pump values  $r > r_{HB}$  and unstable below  $r_{HB}$ , (ii) the bad cavity condition  $\sigma > \sigma_{BCL}$  can be verified even when adiabatic elimination of the medium polarization is made, and (iii) there exists a maximum value of the cavity losses  $\sigma_a$  above which the lasing solution is always unstable. We have seen that the influence of  $\eta$  on HB is mainly quantitative although qualitative new features (such as the disappearance of the limit  $\sigma_a$ ) may appear for  $\eta$  large enough. Special attention has been paid to the stability of the Hopf orbits. We have shown how the ac-Stark parameter  $\eta$  and especially the detuning  $\Delta$  tend to extend the domain of existence of stable orbits (supercritical HB) as a function of cavity losses and population relaxation rates.

Finally the extension in pump parameter  $r$  of the domain of self-pulsing behavior as a function of  $\sigma$ ,  $\chi$ , and  $\Delta$  has been studied. The quite surprising result is that chaos is hard to be observed (it exists only in very small regions of the parameter space) and that the domain of self-pulsing behavior with decreasing  $r$  is much smaller than the region between  $r_{HB}$  and the lasing solution threshold,  $r_0$ . The trivial solution seems to have a wider basin of attraction than the dynamic solutions. We have shown that  $\eta$  and especially  $\Delta$  tend to increase notably the extension of the domain of self-pulsing behavior.

## ACKNOWLEDGMENTS

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## APPENDIX

In this Appendix we apply the AFM [22–25] to the cascade laser models of Secs. II A and II B. Let us begin with the nondegenerate cascade laser model of Eqs. (1). Formal integration of Eq. (1d),

$$\dot{\rho}_{02} = -[\gamma_{\perp} + i(\dot{\phi}_2 + \delta_2)]\rho_{02} + h_{02}, \quad (\text{A1})$$

with

$$h_{02} = i\alpha_2 d_2 + i\alpha_1 \rho_{12} \quad (\text{A2})$$

gives

$$\rho_{02}(t) = \int_0^t dt' h_{02}(t') \exp[(\gamma_{\perp} + i\delta_2)(t' - t) + i\phi_2]. \quad (\text{A3})$$

Repeated integration by parts of Eq. (A3) leads to

$$\begin{aligned} \rho_{02}(t) = & \frac{h_{02}(t) - h_{02}(0) \exp[-(\gamma_{\perp} + i\delta_2)t - i\phi_2]}{\gamma_{\perp} + i(\delta_2 + \dot{\phi}_2)} \\ & + \frac{\dot{h}_{02}(t) - \dot{h}_{02}(0) \exp[-(\gamma_{\perp} + i\delta_2)t - i\phi_2]}{[\gamma_{\perp} + i(\delta_2 + \dot{\phi}_2)]^2} + \dots \end{aligned} \quad (\text{A4})$$

Now we impose large one-photon detuning, i.e.,

$$|\delta_2| \gg \gamma_{\perp}, |\dot{\phi}_2|. \quad (\text{A5})$$

Then, in the long-time limit

$$\rho_{02}(t) = \frac{h_{02}(t)}{i\delta_2} + \frac{\dot{h}_{02}(t)}{(i\delta_2)^2} + \dots \quad (\text{A6})$$

For  $\delta_2$  sufficiently large [22–24]

$$\dot{h}_{02}(t) \ll \delta_2 h_{02}(t) \quad (\text{A7})$$

and consequently

$$\rho_{02}(t) = \frac{h_{02}(t)}{i\delta_2} = \frac{1}{\delta_2} (\alpha_2 d_2 + \alpha_1 \rho_{12}), \quad (\text{A8})$$

which is the same result that one obtains by adiabatically eliminating  $\rho_{02}$ .

Proceeding with  $\rho_{10}$  in the same way one obtains

$$\rho_{10}(t) = \frac{h_{10}(t)}{i\delta_1} = \frac{1}{\delta_1} (\alpha_1 d_1 - \alpha_2 \rho_{12}). \quad (\text{A9})$$

Now, substitution of Eqs. (A8) and (A9) into Eqs. (2) leads to

$$\dot{d}_{21} = \gamma_{\parallel}(n_{21} - d_{21}) - \frac{4}{\delta} \alpha_1 \alpha_2 \text{Im}(\rho_{12}), \quad (\text{A10a})$$

$$\dot{\rho}_{12} = - \left[ \gamma_{\perp} + i \left( \varepsilon_{12} + \dot{\phi}_1 + \dot{\phi}_2 + \frac{\alpha_2^2 - \alpha_1^2}{\delta} \right) \right] \rho_{12} + i \alpha_1 \alpha_2 \frac{d_{21}}{\delta}, \quad (\text{A10b})$$

$$\dot{\alpha}_2 = -\kappa_2 \alpha_2 + \frac{g_2}{\delta} \alpha_1 \text{Im}(\rho_{12}), \quad (\text{A10c})$$

$$\dot{\alpha}_1 = -\kappa_1 \alpha_1 + \frac{g_1}{\delta} \alpha_2 \text{Im}(\rho_{12}), \quad (\text{A10d})$$

$$\dot{\phi}_2 = -\frac{g_2}{2\delta} \left[ (1 - 3n_0) + d_{21} + 2 \frac{\alpha_1}{\alpha_2} \text{Re}(\rho_{12}) \right], \quad (\text{A10e})$$

$$\dot{\phi}_1 = -\frac{g_1}{2\delta} \left[ (1 - 3n_0) - d_{21} + 2 \frac{\alpha_2}{\alpha_1} \text{Re}(\rho_{12}) \right]. \quad (\text{A10f})$$

where we have made  $\delta_2 = \delta$  and  $\delta_1 = (\varepsilon_{12} - \delta)$  and we have

assumed that  $\rho_{00} = n_0$ . Equations (A10) constitute our non-degenerate cascade laser model.

In the degenerate case the procedure is exactly the same and after adiabatic elimination of the dipoles the degenerate TPL equations read

$$\dot{d}_{21} = \gamma_{\parallel}(n_{21} - d_{21}) - \frac{4}{\delta} \alpha^2 \text{Im}(\rho_{12}), \quad (\text{A11a})$$

$$\dot{\rho}_{12} = - \left[ \gamma_{\perp} + i \left( \varepsilon_{12} + 2\dot{\phi} + \frac{1 - \chi}{\sqrt{\chi}} \frac{\alpha^2}{\delta} \right) \right] \rho_{12} + i \alpha^2 \frac{d_{21}}{\delta}, \quad (\text{A11b})$$

$$\dot{\alpha} = -\kappa \alpha + 2 \frac{g_{12}}{\delta} \alpha \text{Im}(\rho_{12}), \quad (\text{A11c})$$

$$\dot{\phi} = -\frac{g_{12}}{2\delta} \left[ (1 - 3n_0) \frac{(1 + \chi)}{\sqrt{\chi}} + \frac{(1 - \chi)}{\sqrt{\chi}} d_{21} + 4 \text{Re}(\rho_{12}) \right]. \quad (\text{A11d})$$

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