Competition between one- and two-photon lasing in two cavity modes

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We examine two-photon lasing in competition with the one-photon lasing into a second mode near resonance with a transition to an intermediate state. The model assumes pumping to the upper state of the two-photon transition. After a derivation of the relevant master equation for the atomic system, we consider the Maxwell-Bloch equation on the basis of which we study the steady state and the time-dependent dynamical behavior of the system. We show that two-photon lasing can be sustained in coexistence with one-photon lasing after the latter is saturated. The results are illustrated with an application to a specific atomic system (potassium) as an amplifying medium. [S1050-2947(99)00607-1]

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I. INTRODUCTION

Although the quest for the two-photon laser began soon after the single-photon laser became operational, to this day such a device does not exist in the optical range of wavelength, and in a form directly analogous to the single-photon one. By ''directly analogous,'' we mean a system in which the upper state of the lasing two-photon transition is pumped externally, with a cavity mode tuned on or around resonance with one-half of the energy separation between the upper and lower state. This has not prevented a number of interesting developments over the years such as (a) the clever exploration of the properties of dressed states by Zakrzewski, Lewenstein, and Mossberg [1] and Gauthier *et al.* [2] to achieve two-photon gain and even continuous operation, and (b) the thorough work of Brune and co-workers [3] on the two-photon micromaser. At the same time much theoretical work has been performed, both semiclassical [4–6] and quantum [7–9], on many aspects of the prospective device, which does indeed continue to reveal more layers of interesting features.

One of the main obstacles in the realization of the standard form of the two-photon laser is a technological one, namely, the difficulty in constructing a cavity in which the modes are sufficiently separated in frequency for the two-photon gain to prevail over the single-photon gain in an adjacent frequency corresponding to a dipole transition from the upper state to an intermediate one of opposite parity. In other words, the quality factor and finesse of presently available cavities in the optical range are not sufficiently favorable. Recall that a two-photon transition at optical wavelengths between two atomic states, with rare exceptions, will involve between them states of parity opposite to that of the active pair which serve as virtual intermediate states. In principle, it is desirable for one such intermediate state to be sufficiently near one-photon resonance in order to enhance the two-photon transition. This is in fact the case in the two-photon micromaser. If, however, a cavity mode happens to be sufficiently near resonance with the one-photon transition between the upper (pumped) and intermediate levels, the gain into that transition will take over. In the microwave range, it is possible to have a cavity such that no cavity mode is sufficiently near that transition. But at optical frequencies, the situation is at best problematic. We are not in a position to judge whether this can be viewed as a technological difficulty which can be overcome soon. In pondering this issue, one should not forget the rapid progress on microcavities in the optical range which has already made possible the realization of the microlaser [10]. Thus it may well be meaningful to keep in mind the possibility of a two-photon microlaser, where the issues discussed in this paper would be of immediate relevance.

Let us assume then that such a coincidence is inevitable, and that a nearby one-photon transition will feed from the pumped upper-state population. Because of the very strength of that transition, relative to the two-photon one, the one-photon oscillation should saturate, in which case oscillation into the two-photon mode should grow. Under what conditions would two-photon lasing be established in coexistence with single-photon lasing? This is the question we have formulated and explored in this paper.

We do indeed show that it is possible to have oscillation in both modes, which implies that under such conditions, the two-photon laser will be accompanied by a satellite one-photon laser. In some sense this is equivalent to a two-mode laser, which under the proper conditions will have a steady state [11]. But unlike the two-mode one-photon laser, the frequencies of the two modes are in this case very different from each other. As we will see in the following sections, the main issue is whether the system can be pumped to a degree sufficient to sustain both oscillations. This, of course, does depend on the parameters of the cavity and the amplifying medium. In an effort to present a quantitative assessment as well, we have performed calculations for a particular set of parameters, some of which are realistic, others perhaps too optimistic. However, many of our results are scalable and can be useful under a different scenario.

Our formalism is based on a traditional laser scheme

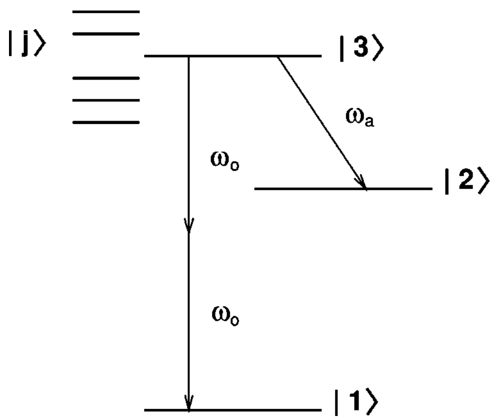

FIG. 1. Schematic representation of the atomic system.

where the pumped exited atomic state is connected to a lower state by a two-photon transition and, in addition, to an intermediate state by a one-photon transition (Fig. 1). In this scheme, there are several spontaneous decay processes, which must also be taken into account since these effects, unlike those in the microwave regime, are significant in the case of optical transitions.

The paper is organized as follows. In Sec. II we derive the master equation for the time evolution of the atomic density matrix; the equations governing the time evolution of the field are derived in Sec. III. The steady state under some reasonable approximations is discussed in Sec. IV. In Sec. V we present the time-dependent behavior of our model applied to a cavity filled with potassium vapor, with the conclusions summarized in Sec. VI.

II. MASTER EQUATION

We consider the interaction of a quantized amplifying medium with a time-dependent electromagnetic field in the semiclassical formalism [12] describing the electric field classically through Maxwell's equations and the medium in terms of the density matrix. The system under consideration is depicted in Fig. 1, where the unperturbed atomic levels $|1\rangle$, $|2\rangle$, $|3\rangle$, and $|j\rangle$ have energies $\hbar\omega_1$, $\hbar\omega_2$, $\hbar\omega_3$, and $\hbar\omega_j$, respectively. The atom interacts with the electric field $\boldsymbol{E}(z,t)$, which is assumed to contain two components E_o and E_a having the same polarization $\hat{\boldsymbol{e}}$ end well separated frequencies ω_o and ω_a:

$$\boldsymbol{E}(z,t)=\tfrac{1}{2}\hat{\boldsymbol{e}}[E_o e^{-i\omega_o t}+E_a e^{-i\omega_a t}+\text{c.c.}], \tag{1}$$

$E_i=\mathcal{E}_i(t)U_i(z)$, $i=o,a$, where $\mathcal{E}_i(t)$ is the slowly varying (during an optical cycle) amplitude of the electric field, and the mode function $U_i(z)$, which is assumed to depend only on the longitudinal coordinate z, is either $\exp(ik_iz)$ for a running-wave cavity or $\sin(k_iz)$ for a standing-wave cavity, with k_i the wave number; ω_a is tuned around the $3\rightarrow2$ transition, and $2\omega_o$ is near the $3\rightarrow1$ two-photon resonance. The total Hamiltonian corresponding to this system can be written as

$$H=H_0+V, \tag{2}$$

where

$$H_0=\hbar\omega_1|1\rangle\langle1|+\hbar\omega_2|2\rangle\langle2|+\hbar\omega_3|3\rangle\langle3|+\sum_j \hbar\omega_j|j\rangle\langle j| \tag{3}$$

is the Hamiltonian of the free atom, and

$$V=-e\boldsymbol{r}\boldsymbol{E} \tag{4}$$

is the interaction with the electromagnetic field. In the dipole approximation, the electric dipole moment operator $e\boldsymbol{r}$ has nonvanishing matrix elements $\mu_{nk}=\langle n|e\boldsymbol{r}|k\rangle$, $n,k=1,2,3,j$, only between levels having opposite parities. In our model, levels $|1\rangle$ and $|3\rangle$ have the same parity, and levels $|2\rangle$ and $|j\rangle$ have the opposite parity. The levels $|j\rangle$ are those which are sufficiently far from resonance with either frequency ω_o or ω_a to serve as virtual intermediate states in the effective two-photon matrix element coupling levels $|1\rangle$ and $|3\rangle$, and are eliminated adiabatically from the density-matrix equations, as discussed below.

The time evolution of the system's density matrix ρ obeys the master equation

$$\frac{d}{dt}\rho=-\frac{i}{\hbar}[H,\rho]. \tag{5}$$

Applying the transformation $\rho\rightarrow e^{-i(H_o/\hbar)t}\tilde{\rho}e^{i(H_o/\hbar)t}$, we obtain the master equation in the interaction picture. The transformed master equation for $\tilde{\rho}$ reads

$$\frac{d}{dt}\tilde{\rho}=-\frac{i}{\hbar}[\tilde{V},\tilde{\rho}], \tag{6}$$

with $\tilde{V}=e^{i(H_o/\hbar)t}Ve^{-i(H_o/\hbar)t}$. We now make a rotating-wave approximation (RWA) in the following way: we keep in $\tilde{V}$ only terms oscillating with frequencies $\omega_{32}-\omega_a$, $\omega_{3j}\pm\omega_o$ and $\omega_{j1}\pm\omega_o$, where $\omega_{nk}=\omega_n-\omega_k$ is the energy difference between levels n and k. We do not make a RWA for field E_o, since ω_o differs substantially from $\pm\omega_{3j}$ and $\pm\omega_{j1}$. Since the detuning of $2\omega_a$ from the two-photon $3\rightarrow1$ resonance is assumed to be very large, the field E_a cannot contribute to this transition, but it does contribute to the dynamic Stark shift of the levels $|1\rangle$ and $|3\rangle$. From Eq. (6), we have

$$\frac{d}{dt}\tilde{\rho}_{j1}=-\gamma_{j1}\tilde{\rho}_{j1}+i\frac{\mu_{j1}}{2\hbar}(E_oe^{-i\omega_ot}+\text{c.c.})e^{i\omega_{j1}t}(\tilde{\rho}_{11}-\tilde{\rho}_{jj})+i\frac{\mu_{j3}}{2\hbar}(E_oe^{-i\omega_ot}+\text{c.c.})e^{-i\omega_{3j}t}\tilde{\rho}_{31}, \tag{7a}$$

$$\frac{d}{dt}\tilde{\rho}_{j2}=-\gamma_{j2}\tilde{\rho}_{j2}+i\frac{\mu_{j1}}{2\hbar}(E_oe^{-i\omega_ot}+\text{c.c.})e^{i\omega_{j1}t}\tilde{\rho}_{12}+i\frac{\mu_{j3}}{2\hbar}(E_oe^{-i\omega_ot}+\text{c.c.})e^{-i\omega_{3j}t}\tilde{\rho}_{32}-i\frac{\mu_{32}}{2\hbar}E_ae^{i\Delta_at}\tilde{\rho}_{j3}, \tag{7b}$$

$$\frac{d}{dt}\tilde{\rho}_{j3}=-\gamma_{j3}\tilde{\rho}_{j3}+i\frac{\mu_{j3}}{2\hbar}(E_o e^{-i\omega_o t}+\text{c.c.})e^{-i\omega_{3j}t}(\tilde{\rho}_{33}-\tilde{\rho}_{jj})+i\frac{\mu_{j1}}{2\hbar}(E_o e^{-i\omega_o t}+\text{c.c.})e^{i\omega_{j1}t}\tilde{\rho}_{13}-i\frac{\mu_{23}}{2\hbar}E_a^* e^{-i\Delta_a t}\tilde{\rho}_{j2}, \quad (7c)$$

where $\Delta_a=\omega_{32}-\omega_a$ is the detuning of the E_a field from the $3\rightarrow 2$ transition resonance, and γ_{nk} is the relaxation constant of the respective matrix element of $\tilde{\rho}$, which we include here phenomenologically. If, as assumed, the frequencies ω_o and ω_a are far from the $j\rightarrow 1$ and $j\rightarrow 3$ resonances, we can neglect the population transfer to the levels $|j\rangle$ and substitute $\tilde{\rho}_{jj}=0$ everywhere. Then we integrate Eqs. (7) making the slowly varying envelope approximation for the field amplitudes E_o and E_a and the density matrix elements $\tilde{\rho}_{nk}$ (adiabatic elimination). Using γ_{nk} to drop the lower limit of integration (it is valid for times $t\gg 1/\gamma_{nk}$) and neglecting γ_{nk} in the nonresonant denominators, we obtain

$$\tilde{\rho}_{j1}=i\frac{\mu_{j1}}{2\hbar}\left(\frac{E_o^* e^{i\omega_o t}}{\omega_{j1}+\omega_o}+\frac{E_o e^{-i\omega_o t}}{\omega_{j1}-\omega_o}\right)e^{i\omega_{j1}t}\tilde{\rho}_{11}-i\frac{\mu_{j3}}{2\hbar}\left(\frac{E_o^* e^{i\omega_o t}}{\omega_{3j}-\omega_o}+\frac{E_o e^{-i\omega_o t}}{\omega_{3j}+\omega_o}\right)e^{-i\omega_{3j}t}\tilde{\rho}_{31}, \quad (8a)$$

$$\tilde{\rho}_{j2}=i\frac{\mu_{j1}}{2\hbar}\left(\frac{E_o^* e^{i\omega_o t}}{\omega_{j1}+\omega_o}+\frac{E_o e^{-i\omega_o t}}{\omega_{j1}-\omega_o}\right)e^{i\omega_{j1}t}\tilde{\rho}_{12}-i\frac{\mu_{j3}}{2\hbar}\left(\frac{E_o^* e^{i\omega_o t}}{\omega_{3j}-\omega_o}+\frac{E_o e^{-i\omega_o t}}{\omega_{3j}+\omega_o}\right)e^{-i\omega_{3j}t}\tilde{\rho}_{32}-i\frac{\mu_{32}}{2\hbar}\frac{E_a e^{-i\omega_a t}}{\omega_{32}-\omega_a}e^{i\omega_{32}t}\tilde{\rho}_{j3}, \quad (8b)$$

$$\tilde{\rho}_{j3}=-i\frac{\mu_{j3}}{2\hbar}\left(\frac{E_o^* e^{i\omega_o t}}{\omega_{3j}-\omega_o}+\frac{E_o e^{-i\omega_o t}}{\omega_{3j}+\omega_o}\right)e^{-i\omega_{3j}t}\tilde{\rho}_{33}+i\frac{\mu_{j1}}{2\hbar}\left(\frac{E_o^* e^{i\omega_o t}}{\omega_{j1}+\omega_o}+\frac{E_o e^{-i\omega_o t}}{\omega_{j1}-\omega_o}\right)e^{i\omega_{j1}t}\tilde{\rho}_{13}+i\frac{\mu_{23}}{2\hbar}\frac{E_a^* e^{i\omega_a t}}{\omega_{32}-\omega_a}e^{-i\omega_{32}t}\tilde{\rho}_{j2}. \quad (8c)$$

We substitute these expressions into the remaining density-matrix equations found from Eq. (6). Since the conditions are such that $\omega_{3j}+\omega_{j1}-2\omega_o\equiv\Delta_o\ll|\omega_o-\omega_{j1,3j}|$, we have $\omega_{j1}-\omega_o\simeq\omega_o-\omega_{3j}$. Now making the two-$\omega_o$-photon RWA we obtain the final set of equations, which govern the time evolution of the density matrix elements of the effective three-level system, where the levels $|j\rangle$ have been eliminated adiabatically:

$$\frac{d}{dt}\tilde{\rho}_{11}=-R\tilde{\rho}_{11}+\gamma_2\tilde{\rho}_{22}+\frac{i}{2\hbar}(E_o^{*2}e^{-i\Delta_o t}\mu_o^{(2)}\tilde{\rho}_{31}-\text{c.c.}), \quad (9a)$$

$$\frac{d}{dt}\tilde{\rho}_{22}=-\gamma_2\tilde{\rho}_{22}+\gamma_3\tilde{\rho}_{33}+\frac{i}{2\hbar}(E_a^* e^{-i\Delta_a t}\mu_{23}\tilde{\rho}_{32}-\text{c.c.}), \quad (9b)$$

$$\frac{d}{dt}\tilde{\rho}_{33}=R\tilde{\rho}_{11}-\gamma_3\tilde{\rho}_{33}-\frac{i}{2\hbar}(E_o^{*2}e^{-i\Delta_o t}\mu_o^{(2)}\tilde{\rho}_{31}-\text{c.c.})-\frac{i}{2\hbar}(E_a^* e^{-i\Delta_a t}\mu_{23}\tilde{\rho}_{32}-\text{c.c.}), \quad (9c)$$

$$\frac{d}{dt}\tilde{\rho}_{21}=-\gamma_{21}\tilde{\rho}_{21}-\frac{i}{2\hbar}\sum_{i=o,a}s_1^i|E_i|^2\tilde{\rho}_{21}+\frac{i}{2\hbar}E_a^* e^{-i\Delta_a t}\mu_{23}\tilde{\rho}_{31}+\frac{i}{2\hbar}E_o^2 e^{i\Delta_o t}\mu_o^{(2)}\tilde{\rho}_{23}, \quad (9d)$$

$$\frac{d}{dt}\tilde{\rho}_{31}=-\gamma_{31}\tilde{\rho}_{31}+\frac{i}{2\hbar}\sum_{i=o,a}(s_3^i-s_1^i)|E_i|^2\tilde{\rho}_{31}+\frac{i}{2\hbar}E_a e^{i\Delta_a t}\mu_{32}\tilde{\rho}_{21}+\frac{i}{2\hbar}E_o^2 e^{i\Delta_o t}\mu_o^{(2)}(\tilde{\rho}_{11}-\tilde{\rho}_{33}), \quad (9e)$$

$$\frac{d}{dt}\tilde{\rho}_{32}=-\gamma_{32}\tilde{\rho}_{32}+\frac{i}{2\hbar}\sum_{i=o,a}s_3^i|E_i|^2\tilde{\rho}_{32}+\frac{i}{2\hbar}E_a e^{i\Delta_a t}\mu_{32}(\tilde{\rho}_{22}-\tilde{\rho}_{33})+\frac{i}{2\hbar}E_o^2 e^{i\Delta_o t}\mu_o^{(2)}\tilde{\rho}_{12}. \quad (9f)$$

We have adopted the notation γ_{nk} for the relaxation rate of the nondiagonal matrix elements $\tilde{\rho}_{nk}$ and γ_n for the relaxation of the populations (diagonal matrix elements) $\tilde{\rho}_{nn}$. The quantity R denotes an effective incoherent pumping rate from $|1\rangle$ to $|3\rangle$ through which inversion of the two-photon active medium can be controlled.

$$\mu_o^{(2)}=\frac{1}{2\hbar}\sum_j\frac{\mu_{3j}\mu_{j1}}{\omega_{j1}-\omega_o} \quad (10)$$

is the two-photon effective dipole moment of the $3\rightarrow 1$ transition, while

$$s_3^i=\frac{1}{\hbar}\sum_j\frac{|\mu_{3j}|^2\omega_{3j}}{\omega_i^2-\omega_{3j}^2}, \quad (11)$$

$$s_1^i=\frac{1}{\hbar}\sum_j\frac{|\mu_{1j}|^2\omega_{j1}}{\omega_{j1}^2-\omega_i^2} \quad (12)$$

are the Stark shift coefficients (polarizabilities) of levels $|3\rangle$ and $|1\rangle$, respectively, due to the field E_i, where in this context E_i is either E_o or E_a. Note that in Eqs. (9d)–(9f) we have taken into account the Stark shift induced by the field E_a as well, since, as mentioned above, for sufficiently large amplitude E_a, this can reach values of significance for the problem under consideration. Moreover, the ac Stark shift can have interesting effects on the quantum statistical properties of the generated field, namely, the squeezing of light, which has been shown elsewhere [9] using a purely quantum-mechanical approach. It is thus worthwhile to keep these terms in the formalism.

III. MEDIUM POLARIZATION

Consider now the polarization of the medium of atomic density N. The general expression which can be used here is

$$P(\boldsymbol{r},t)=N\,\mathrm{Tr}(e\boldsymbol{r}\rho)=N\sum_n\sum_k \mu_{nk}\tilde{\rho}_{kn}e^{-i\omega_{nk}t}, \tag{13}$$

$$n,k=1,2,3,j.$$

On the other hand, the electric field (1) induces the polarization

$$P(\boldsymbol{r},t)=\tfrac{1}{2}[P_oe^{-i\omega_ot}+P_ae^{-i\omega_at}+\text{c.c.}], \tag{14}$$

where $P_i=\mathcal{P}_i(t)U_i(z)$, $i=o,a$, vary slowly in an optical period. We substitute Eqs. (8a) and (8c) into Eq. (13), and equate the resulting expression with Eq. (14). After identifying and grouping together terms oscillating with the same frequency, we obtain

$$P_o=2NE_o(s_1^o\tilde{\rho}_{11}+s_3^o\tilde{\rho}_{33})+4N\mu_o^{(2)}E_o^*\tilde{\rho}_{31}e^{-i\Delta_ot}, \tag{15}$$

$$P_a=2NE_a(s_1^a\tilde{\rho}_{11}+s_3^a\tilde{\rho}_{33})+2N\mu_{23}\tilde{\rho}_{32}e^{-i\Delta_at}. \tag{16}$$

The spatially slowly varying (in an optical wavelength) polarization components $\mathcal{P}_i$ are found by projecting P_i onto the mode function $U_i(z)$:

$$\mathcal{P}_i=\frac{1}{\mathcal{N}_i}\int_0^L dz\,U_i^*(z)P_i\,. \tag{17}$$

Here $\mathcal{N}_i=\int_0^L dz|U_i(z)|^2$ is the mode normalization factor, and L the cavity length.

The evolution of each mode of the electric field is determined by Maxwell's equation

$$\frac{d}{dt}\mathcal{E}_i=-K_i\mathcal{E}_i+i\frac{\omega_i}{2\epsilon_0}\mathcal{P}_i\,, \tag{18}$$

where $K_i=\omega_i/(2Q_i)$ is the relaxation (loss) rate of the field of the ith mode, and Q_i the corresponding quality factor of the cavity. Both, the electric field $\mathcal{E}_i$ and polarization $\mathcal{P}_i$ contain the phase factor $\exp(-i\phi_i)$. This equation, together with polarizations (15)–(17) and the density-matrix equation of motion (9), form a closed system of equations providing a complete description of our system.

IV. STEADY STATE

In order to gain some physical understanding of the process and discuss some aspects of the threshold conditions, we first analyze the steady-state behavior of the system. From here on we restrict ourselves to the easier case of a running-wave cavity, since it is more tractable and in any case contains the basic physics.

In Eq. (18) one can identify the first term on the right-hand side as the cavity losses per unit time, while the second term is responsible for amplification in the system. The general procedure which we use here is as follows: After applying some straightforward unitary transformations, we solve the density-matrix equations (9) for the steady state and then use this solution for the derivation of the polarizations entering the Maxwell's equation (18). Here we neglect the Stark shifts, keeping in mind that, in principle, they can be incorporated into the detuning of the corresponding mode from the atomic resonance. For simplicity we neglect the contribution of $\tilde{\rho}_{21}$ too, assuming that γ_{21} is much larger than all other terms in Eq. (9d). We further set $\Delta_o=0$ and take $\gamma_3=0$, which is quite realistic for many atomic systems, and our aim in this section is a qualitative rather than quantitative evaluation. With these approximations Eq. (18) provides an expression for the ratio of the gain ($\mathcal{G}$) [12] in the two lasing frequencies (o and a) provided that the system operates above threshold:

$$\frac{\mathcal{G}_o}{\mathcal{G}_a}=4\frac{\omega_o\mu_o^{(2)2}}{\omega_a\mu_{32}^2}E_o^2\frac{R(\Delta_a'^2+\gamma_{32}^2)\gamma_2+2\Omega_a^2(R-\gamma_2)\gamma_{32}}{(2\Omega_o^{(2)2}+R\gamma_{31})\gamma_2\gamma_{32}}, \tag{19}$$

where $\Omega_a=\mu_{32}|\mathcal{E}_a|/2\hbar$ and $\Omega_o^{(2)}=\mu_o^{(2)}|\mathcal{E}_o|^2/2\hbar$ are the one- and two-photon Rabi frequencies induced by the fields $\mathcal{E}_a$ and $\mathcal{E}_o$, respectively. The detuning Δ_a' is shifted now from Δ_a by the amount of the steady-state value of $\dot{\phi}_a(t\to\infty)\equiv\dot{\phi}_a^{\rm ss}$: $\Delta_a'=\omega_{32}-\omega_a-\dot{\phi}_a^{\rm ss}$.

Equation (19) has a simple physical interpretation: First of all, we note that in the case $\mathcal{E}_o=0$ the ratio of $\mathcal{G}$'s vanishes and, consequently, the inverse ratio tends to infinity even if $\mathcal{E}_a=0$ as well. This stems from the well known fact that, unlike one-photon lasing, the two-photon laser operation needs to be triggered by an initial field [4], since the two-photon spontaneous emission is practically zero. Consider further the last fraction of Eq. (19). Here we see that $\mathcal{G}_o$ increases with the detuning Δ_a of the competing process. In addition, $\mathcal{G}$ in each mode is inversely proportional to its own saturation. Thus, if we continue pumping, depending on the Rabi frequencies and other parameters of system, in the course of time, one of the modes (in this case $\mathcal{E}_a$) will be amplified more than the other until it reaches saturation. Further pumping, if it is sufficiently strong to be still above threshold, will cause the amplification of the second field. It is worth noting also that the ratio of $\mathcal{G}$'s is proportional to the ratio of the squares of the dipole moments of the two transitions. It should be stressed that steady-state conditions and a relation between the gains can be obtained without the simplifying assumptions preceding Eq. (19). The resulting expression is, however, too complicated to be of inspectional value.

We turn now to the derivation of the threshold conditions for laser operation of the system. In the same approximation as above, from Eq. (18), after making the Taylor expansion of $\mathcal{P}_i$ for small parameter $|\mathcal{E}_i|$, respectively, and omitting terms responsible for self-saturation, the following two conditions are obtained.

(i) For the ω_o mode (providing that $|\mathcal{E}_o|$ is small enough so that $\mathcal{G}_o$ is far below the saturation),

$$R=\frac{2\Omega_a'^2(\Omega_o^{(2)}\eta Q_o+\gamma_{31})\gamma_2\gamma_{32}}{2\Omega_a'^2(\Omega_o^{(2)}\eta Q_o-2\gamma_{31})\gamma_{32}+(\Omega_o^{(2)}\eta Q_o-\gamma_{31})\gamma_2}, \tag{20}$$

where $\eta=4\mu_o^{(2)}N/\epsilon_0$ and $\Omega_a'=\Omega_a/(\Delta_a'^2+\gamma_{32}^2)^{1/2}$. This equation provides the threshold value of the pumping for lasing into the $\mathcal{E}_o$ mode, and implies that the threshold is raised as the intensity I_a (inherent in Ω_a^2) of the ω_a mode is increased, since the probability of stimulated emission into ω_a increases with that intensity. On the other hand, further increase of I_a leads to the saturation of this process (the term proportional to Ω_a^2 in the denominator) which leads to a saturation of the value of R. This demonstrates that the simultaneous oscillation in a nearby single-photon transition will not necessarily prevent oscillation into the two-photon mode. Its effect will be to raise the threshold value of the pumping.

(ii) For the ω_a mode (providing that $|\mathcal{E}_a|$ is small enough so that $\mathcal{G}_a$ is far below the saturation), we have

$$R=\frac{2\Omega_o^{(2)2}}{\gamma_{31}}\left(\frac{1}{\gamma_{32}\zeta Q_a-1}-1\right), \tag{21}$$

where $\zeta=\mu_{32}^2N/2\hbar\epsilon_0(\Delta_a'^2+\gamma_{32}^2)$. The interpretation of this condition is more subtle. When the term inside the parentheses is positive, the threshold [as in (i) above] is proportional to the square of the two-photon Rabi frequency or, in other words, to the square of the intensity I_o of the $\mathcal{E}_o$ field, since for a two-photon transition $\Omega_o^{(2)}\propto|\mathcal{E}_o|^2\propto I_o$. In the opposite case, when the term inside the parenthesis is negative, the pumping rate threshold also becomes negative and condition (i) is, certainly, not satisfied. This means that absorption of the $\mathcal{E}_o$ field is taking place, due to which some population is transferring to level $|3\rangle$ with the result that enhancement of the field $\mathcal{E}_a$ becomes possible. It should be mentioned that conditions (i) and (ii) are completely independent of each other, and that each of them determines the lasing threshold of the corresponding mode as a function of the cavity decay rate, Rabi frequency of the competing process, and the other parameters of the system.

V. DYNAMICS

The quantitative analysis of the dynamics of the system does not lend itself to simple analytical approximations, but requires a rigorous solution of the time-dependent equations. It is the purpose of this section to present and discuss results of the numerical solution of the equations derived in Secs. II and III. The theory has been applied to a cavity assumed to be filled with potassium vapor of density $N=10^{13}$ cm^{-3}. Not that we literally propose the construction of a laser with that active medium but only in order to have a context of parameters corresponding to a real atom for numerical illustration of the ideas. Levels $|1\rangle$, $|2\rangle$, and $|3\rangle$ of our model are assumed to correspond to the states $4P_{3/2}$, $3D_{5/2}$, and $6P_{3/2}$ of potassium, respectively. The two-photon transition frequency is $\omega_{31}/2=2\pi\,2.39\times10^{14}$ rad/s, and the one-photon transition resonance is $\omega_{32}=2\pi\,2.24\times10^{14}$ rad/s [13]. There is also the state $3D_{3/2}$ lying above the level $|2\rangle$ by the amount $\Delta=2.3$ cm^{-1}, which must be taken into account when choosing Δ_a, because $\Delta_a\ll\Delta$ should be satisfied to have the RWA valid; we take $\Delta_a=5\times10^8$ rad/s everywhere below. The upper level $|3\rangle$ spontaneously decays to level $|2\rangle$ with the decay rate $\gamma_3=3.7\times10^5$ s^{-1}; the decay of $|3\rangle$ to the state $3D_{3/2}$ is almost ten times smaller, and we neglect it in further calculations. Level $|2\rangle$ decays further to level $|1\rangle$ with the rate $\gamma_2=2.6\times10^7$ s^{-1}. The main reason for choosing these states is that the photons ω_o connecting levels $|1\rangle$ and $|3\rangle$ are sufficiently near resonant with the intermediate $5S_{1/2}$ state (with detuning $\omega_{j1}-\omega_{31}/2\simeq2\pi\,3.77\times10^{10}$ rad/s), which enhances the two-photon coupling significantly.

The above combination of choices provides us with a model in which an intermediate level ($5S_{1/2}$) serves the purpose of enhancing the two-photon transition, while another intermediate level ($3D_{5/2}$) participates in the single-photon lasing. One could just as easily construct a model using only one intermediate level for the dual purpose. This is in fact what must be done in the context of the two-photon micromaser, which we will treat in a forthcoming paper. Having, however, made the decision to adopt the above combination of levels in potassium as one model, we have the freedom to separate the roles of the two intermediate states. This, of course, assumes some flexibility with the finesse of the cavity, which in reality may require lasing in both intermediate states as well as in $3D_{3/2}$. All these further complications could be incorporated into our treatment, but at this stage the added formal complexity would probably detract from rather than enhance an understanding of the basic effect under consideration.

A calculation of the dipole matrix elements and Stark shift parameters yields the following numbers [14], measured in meter-kilogram-second units (MKS): $\mu_{32}/2\hbar=2.67\times10^4$, $\mu_o^{(2)}/2\hbar=1.74\times10^{-2}$, $s_1^o/2\hbar=1.02\times10^{-1}$, $s_3^o/2\hbar=2.95\times10^{-3}$, $s_1^a/2\hbar=2.91\times10^{-4}$, and $s_3^a/2\hbar=1.71\times10^{-4}$. In order to satisfy the threshold conditions, the pumping rate has been chosen relatively large ($R=5\times10^8$ s^{-1}). Although our choice of potassium has been made only as an illustration, in the interest of consistency we must follow it a bit further. R is large compared to the spontaneous decay rate of $4P_{3/2}$ to $4S_{1/2}$, which is equal 3.8×10^7 s^{-1}. Let us then assume that an additional depopulating light source causes $4P_{3/2}$ to make a transition to $4S_{1/2}$ at a rate a bit more than ten times faster. Then some pumping mechanism R can take the system from $4S_{1/2}$ directly or indirectly to the upper lasing state. This is a rather unconventional pumping concept, but given our idealized model all we can require is that it not be unphysical. To be more definitive about the pumping mechanism, one would need to know the medium and the cavity, which is beyond our expertise. For the convenience of illustration, the cavity width for both modes has been assumed to be extremely small: $K_a=10^3$ s^{-1} and $K_o=2\times10^2$ s^{-1}, because otherwise the intensities of the fields do not reach significant values before the system saturates. These, together with R, are the optimistic parameters alluded to earlier. If, however, the cavity width $K_{a,o}$ and the density of atoms N are changed by a factor β, we obtain the same behavior of the system by simply rescaling by the same factor β the time scale of the process, as easily seen from Eq. (18). The only limitation on this reasoning is the necessity of keeping the process within the adiabatic regime, i.e., the rate of change of both field amplitudes must be small enough for

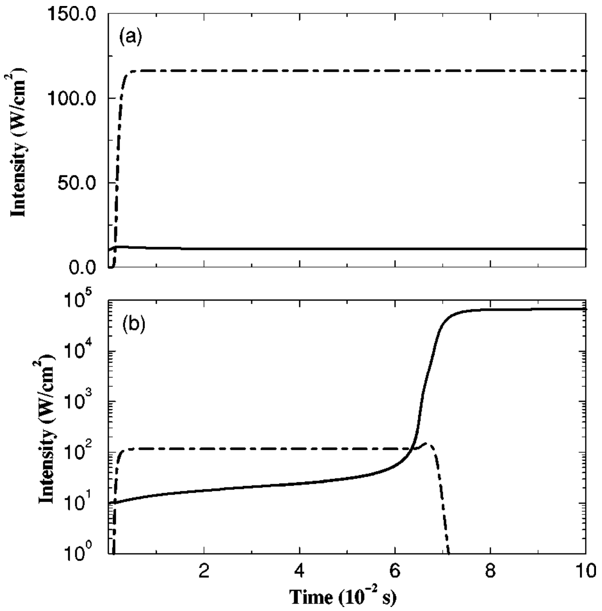

FIG. 2. Intensities I_a (dot-dashed curves) and I_o (solid curves) of the two cavity fields as functions of time, for two different values of detuning Δ_o: (a) $\Delta_o=0$, and (b) $\Delta_o=\frac{1}{2}(\Delta_a-\sqrt{\Delta_a^2+4\Omega_a^{(s)2}})$. All other parameters are given in the text (Sec. V).

the atoms to be close to the state of equilibrium at any point in time.

In Fig. 2, we show the time evolution of the intensities I_a and I_o of the competing fields for two different values of the detuning Δ_o. We trigger the lasing on the ω_o mode by an external field; the operation on the ω_a mode in principle triggers itself by spontaneous emission. In this treatment, however, which is based on a semiclassical formalism, it also must be triggered externally. For the case $\Delta_o=0$ [Fig. 2(a)], the strength of the $\mathcal{E}_a$ field grows quickly at first, until it reaches saturation and then remains constant throughout, while I_o, after a small gain at the very beginning of the process, decays back to the triggering field's value, which we assume always to be on. The reason for such a behavior of I_o is the ac Stark splitting of the levels $|2\rangle$ and $|3\rangle$, which increases proportionally to the intensity I_a moving the transition $3\rightarrow1$ away from resonance.

A brief analysis in terms of dressed states [15] may be useful at this point. Recall that the pair of dressed states $|\Psi_\pm\rangle$ is determined through the eigenvalue problem

$$[\Delta_a|3\rangle\langle3|-\Omega_a(|3\rangle\langle2|+|2\rangle\langle3|)]|\Psi_\pm\rangle=\lambda_\pm|\Psi_\pm\rangle, \tag{22}$$

where $\lambda_\pm=\frac{1}{2}(\Delta_a\pm\sqrt{\Delta_a^2+4\Omega_a^2})$. These dressed states $|\Psi_\pm\rangle$ can then be expressed in terms of the bare atomic states $|2\rangle$ and $|3\rangle$ as

$$|\Psi_-\rangle=\cos\theta|2\rangle-\sin\theta|3\rangle, \tag{23a}$$

$$|\Psi_+\rangle=\sin\theta|2\rangle+\cos\theta|3\rangle, \tag{23b}$$

where the mixing angle θ is defined by

$$\tan\theta=\left(\frac{\sqrt{\Delta_a^2+4\Omega_a^2}-\Delta_a}{\sqrt{\Delta_a^2+4\Omega_a^2}+\Delta_a}\right)^{1/2}. \tag{24}$$

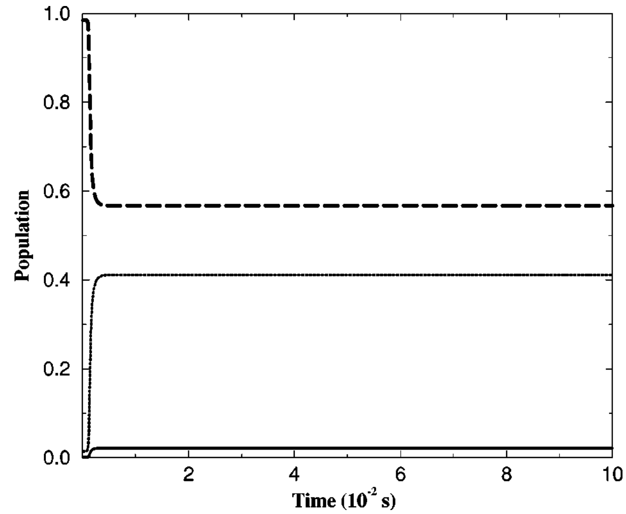

FIG. 3. Population of the levels $|1\rangle$ (solid line), $|2\rangle$ (dotted line), and $|3\rangle$ (dashed line) as a function of time for the case of $\Delta_o=0$.

Now setting $\Delta_o=\frac{1}{2}(\Delta_a-\sqrt{\Delta_a^2+4\Omega_a^{(s)2}})$ [Fig. 2(b)], where $\Omega_a^{(s)}$ is the saturated value of the Rabi frequency Ω_a, we tune the $\mathcal{E}_o$ field into the two-photon resonance with the upper one of the pair of dressed states $|\Psi_\pm\rangle$. In this case the intensity I_o grows very slowly at first, since the two-photon transition is nonresonant at the beginning of the process; however, as soon as I_a reaches saturation, the gain of I_o becomes substantial. Then further increase of the strength of the $\mathcal{E}_o$ field induces, in its turn, the Stark splitting of the levels $|1\rangle$ and $|3\rangle$, which is now proportional to the square of the two-photon effective Rabi frequency or the fourth power of the field strength. When this splitting becomes equal to the detuning Δ_a of the $\mathcal{E}_a$ field, the latter begins to grow again, but then, with a further increase of the splitting, decreases fast since it drifts far from resonance. The behavior of the populations of levels $|1\rangle$, $|2\rangle$, and $|3\rangle$ for the two cases discussed above is shown in Figs. 3 and 4. The population difference between levels $|3\rangle$ and $|2\rangle$, after reaching the

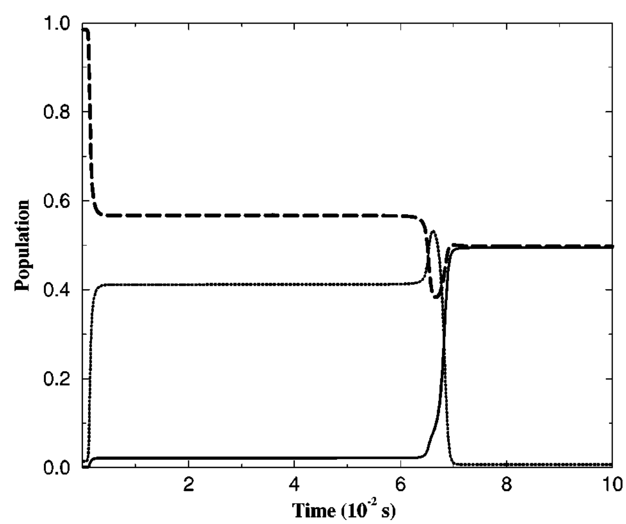

FIG. 4. Population of the levels $|1\rangle$ (solid line), $|2\rangle$ (dotted line), and $|3\rangle$ (dashed line) as a function of time for the case of $\Delta_o=\frac{1}{2}(\Delta_a-\sqrt{\Delta_a^2+4\Omega_a^{(s)2}})$.

saturated value, always remains constant in the case of $\Delta_o = 0$. On the other hand, when Δ_o is adjusted to resonance with $|\Psi_+\rangle$, once the Stark shift of level $|3\rangle$ induced by $\mathcal{E}_o$ field compensates for the detuning Δ_a, the population difference becomes negative, but the intensity I_a grows at this short interval of time; this effect can be referred to lasing without inversion (LWE) [16]. This LWI results from quantum interference which suppresses the absorption from level $|2\rangle$ and leads to amplification on the transition $|3\rangle \rightarrow |2\rangle$, even in the absence of a population inversion. Further increase of I_o causes the equalizing of the populations of levels $|3\rangle$ and $|1\rangle$, and consequently the saturation, for the specific values of parameters we have used, is reached when $I_o \sim 6 \times 10^4$ W/cm^2.

VI. CONCLUSIONS

In this paper we have derived the general master equation for a system consisting of the three-level Λ-type upper-level-pumped atoms with one dipole-forbidden transition, placed in a two-mode optical cavity, where the first mode is tuned to the two-photon transition of the atom and the second mode is near the one-photon resonance. The treatment has been carried out in the framework of the semiclassical laser theory, which describes the electric field by the Maxwell equations and the medium by the density-matrix formalism. The steady-state behavior of the system has been analyzed under some reasonable approximations, and the threshold conditions have been discussed. For a quantitative assessment, time-dependent solutions for a cavity assumed to be filled with potassium vapor have been presented, illustrating the point that the existence of the possible one-photon competing process does not necessarily destroy the lasing on the two-photon transition of the atom. One aspect that has emerged from this analysis, which sets it apart from the analogous two-mode problem in the single-photon laser, is the significance of the Stark shift in the dynamics. Although the role of the Stark shift in the dynamics, as well as the photon statistics of the two-photon laser, has been noted and discussed [5,8,9], its role in the present context is different and perhaps more influential on the behavior of the system. Another aspect that has emerged from our treatment is the connection with the concept of lasing without inversion. Of course, the basic scheme of the two-photon laser studied in this paper assumes inversion of populations through pumping, but the connection with lasing without inversion enters through the coupling of the modes. We are not in the position to ascertain at this moment whether present or foreseeable technological possibilities, in terms of cavity quality factor and finesse, place experimental realization near or outside the parameter space we have explored. In any case, our model is scalable, and hopefully technical refinement will be continuing.

Given the idealized model we have employed in our simulations, some not negligible issues are necessarily left unanswered. Not knowing the specific form of the cavity, we cannot say anything about its length, which does not allow us to say anything about pulsations on the one-photon lasing transitions in the case of excessive gain path length. Clearly, such pulsations would disrupt the two-photon lasing. If and when schemes such as the one modeled herein are explored experimentally, those in charge will be much more qualified than we are to assess the relevant issues.

The problem we have formulated and solved in this paper has an interesting counterpart in the microwave regime, where one can in fact tailor the cavity at will, in combination with the choice of the principal quantum number of the pumped Rydberg state, so as to have or not have a second mode in near resonance with a single-photon transition. The experimental realization of such a scenario should be relatively easy with present day technology, and in a followup paper we expect to report on predictions relevant to that case. Finally, it may be technologically feasible at this time to contemplate the exploration of these effects in the context of the microlaser, which combines the single- or few-photon aspect with a more convenient mode structure in the optical range.

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