Inversionless gain versus efficient gain: The autoionizing states configuration

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The absorption and stimulated emission of coherent electromagnetic radiation tuned close to an autoionizing state is investigated with the aim of clarifying some of the aspects behind the proposals of inversionless gain involving such atomic configurations. This is carried out within the framework of individual atomic evolution rather than by the use of density matrix equations, thus providing better insight into the fundamental processes of absorption and emission. In particular, we find that stimulated emission does not overcome absorption in spite of quantum interference of the Fano type being present. It is also shown that gain, in this configuration, although it can take place without population inversion between the involved levels, requires in any case a strong departure from thermal equilibrium conditions. Thereby a strong pumping mechanism, or equivalently a strong excitation power, is needed to reach the lasing threshold. This raises some questions on the relevance of the population inversion indicator of efficient gain implicitly assumed in similar configurations and in many related works. [S1050-2947(98)05707-2]

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I. INTRODUCTION

A few years ago, atomic configurations making use of autoionizing states were shown to provide gain without population inversion [1,2]. Indeed, it was found that, for a laser field tuned close to the Fano minimum, the stimulated emission probability for an atom initially in the upper autoionizing state would overcome absorption for an atom initially in the lower state. It was surmised that such apparently singular behavior had its premises in the peculiar shape of the absorption spectra of the autoionizing states that, as is well known, can even yield no absorption when the proper set of conditions is satisfied [3]. The reading of these results in terms of the basic concepts underlying the exchange of energy between atomic systems and coherent radiation would imply the possibility of defying a well-known and important limitation always present in the current design of laser systems, thus opening prospects of lasing action in regions of the optical spectra hard to reach on the basis of the standard working principles. It is therefore of no surprise that there has been ensuing intense theoretical activity aiming to test similar ideas also in more standard configurations working only with bound atomic systems [4].

However, within a short period from the initial proposal, it was recognized that the transient response of the atomic polarization, neglected in the initial investigation, played a very important role [5] such as to modify in a considerable way the initial picture of emission and absorption. In spite of this, at least two followup papers [6,7] enforced the original claim in a way that would have appeared contradictory. In fact, while in [6] an ideal laser without inversion configuration, i.e., a configuration where any small amount of population in the upper state yields gain independently of the lower level population, was shown to be possible, in [7], which reported a numerical analysis in the pulsed regime, there were no signs of such an ideal response even though the pulse duration in a number of cases treated was much larger than the autoionizing decay time, thus allowing a rather direct comparison between the two calculations. Since then, to our knowledge, no further relevant works have been carried out on this configuration, and most of the attention turned to systems with discrete state configurations. Nonetheless, it is our opinion that it is possible to improve the understanding of these mechanisms in the autoionizing states' configurations by adopting a different and intuitively more accessible point of view than that used so far in previous investigations. We refer here to the concepts of quantum trajectories and individual atomic evolution that have received a great impulse in connection to the observation of single atoms in electromagnetic traps [8,9] and to recent developments in atomic manipulation by laser light [10].

It is well known that the optical Bloch equations are essentially averages over individual atomic evolutions and this aspect, with special regard to the physical reality of these trajectories, has been widely emphasized in the current literature. We focus our attention here on the very clear understanding of the fundamental processes of gain and absorption of photons by atomic systems that such a picture provides [11]. Indeed, some important symmetry relations relating to multiphoton processes that open a possible way toward a generalization of the principle of inversion [12] appear rather natural in this framework. These results, established by virtue of the very simple symmetry properties owned by the Hamiltonian evolution between wave function collapses caused by the dissipation processes, offer a different and nontraditional perspective to the phenomenon of inversionless gain [13]. Moreover, we believe that they can profitably be adapted also to the case of the autoionizing states' configuration. The advantage of such an approach has also been partially recognized in [5], but it has been more completely realized in [11]. The analysis reported in this work is based on a very simple parallelism that one can rather naturally establish between the autoionizing states' configurations and a bound atomic system with a single ground level interacting via a coherent field with a multitude of upper states. Considering the very simple structure of this last system, one would be somewhat reluctant to accept that, as would be implicitly implied by the possibility of gain without population inver-

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sion, its gain efficiency could outreach that normally obtained when operating with standard configurations. Indeed, pursuing such a point of view and adopting a very simple model of dissipation we find that in a medium initially prepared with equal population in the ground and in the upper autoionizing state, and thereafter coupled to a pulsed field, no overall gain is possible. Thus the question of whether inversionless gain and efficient gain are truly closely related to one another requires further accurate investigation, as we plan to do in this work.

We develop our point of view with an introductory section concerning the procedures at the basis of the calculation of absorption and stimulated emission. Although these are already well known and it could seem pointless to insist on this aspect, we believe that the point of view of individual atomic evolution and the preeminent use of the energy in place of atomic polarization to calculate absorption and emission, as is naturally implied by the quantum trajectory point of view, will shed additional light on the concurrent role of coherent evolution and dissipation. An interesting result derived here is represented by the sum rule concerning the energy exchanged with the field as a function of the initial atomic state. In the second section we apply these considerations to the particular case of autoionizing states by taking into account two configurations that have been previously investigated in the framework of inversionless gain proposals. The interpretation of these results will be different from that usually proposed and based on the mechanism of quantum interference. In particular, we will find that, quite surprisingly, quantum interference does not have the desired effect of providing a reduction of the absorption over the stimulated emission. A further section in which the meaning of the population inversion condition is analyzed will help to better clarify the conditions under which many previous results were obtained and will also serve to inspire a critical attitude toward the use that could be made of this quantity as an indicator of efficient gain. In other words, we will investigate the question of whether or not the no-population inversion condition can provide any advantage in terms of the usual parameters related to the efficiency of the lasing process.

II. CALCULATION OF ABSORPTION AND STIMULATED EMISSION

A. General expressions

Let us consider a quantum system whose Hamiltonian H is time dependent. The rate of energy out from this system can be written as

$$w=-\mathrm{Tr}\left(\rho\frac{\partial H}{\partial t}\right), \tag{1}$$

an expression that has its counterpart in the classical Hamiltonian mechanics [14] and leads to the usual expression in terms of the oscillating atomic polarization in quadrature with the driving field.

A very simple relation can be derived from the one above if we consider an isolated atomic system, i.e., a system not subjected to dissipation. This can be the case of interaction with an electromagnetic pulse faster than any atomic relaxation, broad-band pulse, or, in the case of dissipation processes fulfilling the so-called impact approximation, in the period between two subsequent interactions with the dissipation source where the evolution can be considered to be purely Hamiltonian. Thus, by representing with ρ the state of the system, we can write

$$w=-\mathrm{Tr}\left(\rho\frac{\partial H}{\partial t}\right)=-\frac{d}{dt}\mathrm{Tr}(\rho H), \tag{2}$$

as is easily proved by using the Schrödinger equation of motion for ρ.

B. Some useful relations and sum rules

1. Pulsed regime

The physical meaning of the above expression is rather evident. As a matter of fact, it can be immediately appreciated in the pulsed regime where the interaction V vanishes both at the initial and final time of the interaction process. In this case, the total energy into the field can be written, by integration of Eq. (2), as

$$\epsilon=-\Delta H_0=\mathrm{Tr}(\rho_{\mathrm{in}}H_0)-\mathrm{Tr}(\rho_{\mathrm{fin}}H_0). \tag{3}$$

In the special case of a two level system, this can be further transformed in order to display a symmetry between absorption and emission, which is reminiscent of Einstein's B coefficients. To this end, let us indicate by ϵ_g the energy delivered into the field when the atom is initially in the ground state $|g\rangle$ and by ϵ_e that corresponding to the atom initially in the excited state $|e\rangle$: thus the simple relation

$$\epsilon_g+\epsilon_e=0 \tag{4}$$

holds. This can be proved by noticing that such a sum corresponds to the energy exchanged with the electromagnetic wave packet when the atom is initially in a density matrix proportional to the identity, i.e., with equal probability in the upper and lower state. This density matrix is left unchanged by the Hamiltonian evolution, therefore on the basis of Eq. (3) the total energy into the field, represented by the sum of ϵ_g and ϵ_e, is zero. It is to be noted that, for the validity of Eq. (4), it is not necessary that V vanishes at the initial and final time, being understood that ϵ_g and ϵ_e are accordingly interpreted. This rests on the fact that H is a constant of motion when the initial state is the identity matrix.

An atomic system consisting of a lower level $|g\rangle$ coupled with an electromagnetic field to a multitude of upper levels $|e_i\rangle$, as shown in Fig. 1(a), satisfies a similar relation. The total Hamiltonian can be written as

$$H=H_0+V=H_0+\frac{\hbar\Omega}{2}(|\Phi_c\rangle\langle g|e^{-i\omega t}+|g\rangle\langle\Phi_c|e^{i\omega t}), \tag{5}$$

where $|\Phi_c\rangle=\Sigma_i c_i|e_i\rangle$ is the normalized coupled state. A laser pulse with constant polarization can be represented here by a slowly varying Ω parameter. In the case of a pulsed electromagnetic interaction the same kind of argument used above allows us to establish the sum rule

$$\epsilon_g+\sum_i \epsilon_i=0, \tag{6}$$

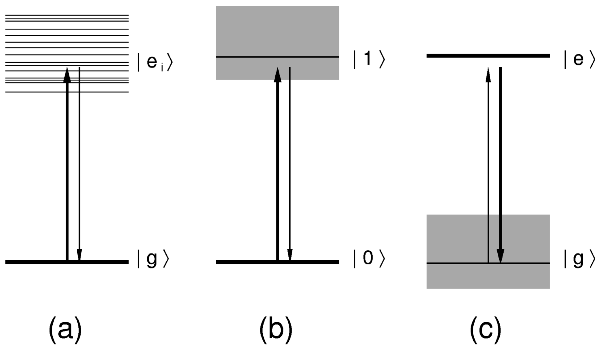

FIG. 1. Atomic configuration of one coherent field coupled with atomic systems discussed in this article. (a) Discrete set of upper states. (b) Continuum set of upper states with an autoionizing level; upon Fano diagonalization this is equivalent to a structured continuum. (c) Autoionizing state as lower level of an atomic configuration. The level thickness is in relation to level population and thickness of arrows indicates the strength of the relative stimulated emission and absorption.

where the terms ϵ_g and ϵ_i are the energies delivered into the field when the atom is initially in the state $|g\rangle$ and $|e_i\rangle$, respectively. The very simple form of this relation suggests some interesting considerations. Under some very broad conditions we can assume ϵ_i is positive for all i, i.e., $\epsilon_i \geqslant 0$; and as a consequence of Eq. (6), also $\epsilon_g \leqslant 0$. This would certainly happen within the limit of small fields where, if the atom is initially in a state $|e_i\rangle$, the dominant part of the wave function change, taken as a basis for the calculation of the energy in Eq. (3), will be found mostly in the lower state. In the case where the small field intensity condition is removed, one should account also for multiphoton processes. In fact, it could in principle happen that a two-photon process starting in the upper state returns part of the final atomic wave function also in some excited states higher in energy than the initial one. However, the energy associated with this last transition can be considered small with respect to the one coming from the one-photon process that, besides being stronger in amplitude, involves also the usually large energy corresponding to the atomic transition. Thus we can safely assume the conditions stated above regarding the sign of ϵ_i and ϵ_g, a condition that implies $|\epsilon_g| \geqslant |\epsilon_i|$ for all i. Thus stimulated emission from any of the upper states is always smaller than absorption from the ground state. This result, by virtue of the similarity mentioned above, can rather easily be extended also to the autoionizing state configuration.

2. Stationary regime

It is useful to discuss also the case of an atom driven by a steady field. By integrating Eq. (2), and noticing that when Ω is constant $H + \hbar\omega P_g$ is a constant of motion, we can write

$$\begin{aligned}\epsilon &= -\langle\psi|H|\psi\rangle + \langle\psi(0)|H(0)|\psi(0)\rangle \\ &= \hbar\omega[\langle\psi|P_g|\psi\rangle - \langle\psi(0)|P_g|\psi(0)\rangle],\end{aligned} \tag{7}$$

where P_g and P_e are the projection operator on the ground state and on the manifold of upper states, respectively. This final expression becomes rather meaningful if read in probabilistic terms. To better understand it, let us assume that the atom is initially in the ground state, then

$$\epsilon = -\hbar\omega\langle\psi|P_e|\psi\rangle, \tag{8}$$

meaning that a photon of energy $\hbar\omega$ is absorbed with the same probability $\langle\psi|P_e|\psi\rangle$ as that of finding the atom in the upper state. On the contrary, if the atom is initially in one of the upper states the expression for the energy becomes

$$\epsilon = \hbar\omega\langle\psi|P_g|\psi\rangle, \tag{9}$$

meaning that a photon is emitted with the same probability as that of finding the atom in the lower state. This can also be understood in a different way. By adopting the picture used in [11], and based on the dressed atom representation, the state $|e_i, N\rangle$, upper atomic states with a field state containing N photons, is coupled to $|g, N+1\rangle$, lower atomic state with a field state containing $N+1$ photons, by means of the matrix element

$$\langle g, N+1|H_{a+f}|e_i, N\rangle = -c_i\frac{\Omega}{2}. \tag{10}$$

A slow dynamic evolution ruled by the Schrödinger equation takes place within these two states for all values of N close to the one corresponding to the intensity of the field. The quantum-mechanical average number of photons in the evolved wave function

$$|\psi(N)\rangle = a|g, N+1\rangle + \sum_i b_i|e_i, N\rangle \tag{11}$$

is given by

$$\langle N\rangle = \mathcal{P}_g(N+1) + \mathcal{P}_e N = N + \mathcal{P}_g, \tag{12}$$

where $\mathcal{P}_g = |a|^2$ is the probability of having $N+1$ photons in the electromagnetic field and $\mathcal{P}_e = \Sigma_i |b_i|^2$ is the probability for N photons, while the change in the average number of photons from an initial to a final wave function results in

$$\langle\Delta N\rangle = \mathcal{P}_g^{(f)} - \mathcal{P}_g^{(i)}, \tag{13}$$

which obviously differs from Eq. (7) for ϵ only by a multiplicative factor representing the photon energy.

C. Modeling of dissipation

A dissipation process is needed in order to produce an irreversible transfer of energy between the atomic system and the field. This, usually produced by a collisional or a radiative process, perturbs the atomic coherent evolution and downgrades, in an irreversible way, the atom+field wave function to a density matrix. It also turns out that, in most cases, the basic mechanism of this action can be a very simple and intuitive one. In the collisional case it is found that the impact approximation, based on the fact that the perturbation produced by the collisional partner takes place in a very short time, has a wide range of validity. From the formal point of view, a very similar picture is found to hold also in the case of spontaneous emission dissipation due to the fact that the photon emission takes place instantaneously and at the same time forces the atom in the corresponding ground state [9]. Thus, in general, we can picture the atomic evolution as a sequence of coherent evolution periods interrupted by instantaneous perturbations whose result is to transform the actual wave function into a density matrix. In the context of the quantum trajectories point of view only one of the alternatives, properly represented with a given probability in the final density matrix resulting from the instantaneous interaction with the dissipation source, is chosen by the evolution. In this way only wave-function evolutions need to be carried out, with some advantage from the computational point of view in atomic systems with many involved atomic states [10]. This is, in a certain sense, similar to what occurs in dynamical systems subjected to stochastic forces where one decides to solve the Langevin equation instead of the Fokker-Planck equation. Thus, if the dissipation mechanism is known, the time statistics of the jumps as well its final state are also supposed to be known. Thus the energy delivered in the field can be calculated by applying Eq. (7) to each coherent period and summing up the contributions over a sufficient period of time or applying Eq. (13) if the dressed atom picture is adopted [15].

The advantage of the quantum trajectory method is that it allows a better insight into the transfer of energy produced in these processes. In fact, in the simple case of collisional dissipation, the exchange of energy with the reservoir takes place only in a very short time period during which the field is not affected. On the contrary, during the coherent evolution the exchange of energy takes place only between the field and the atom. This simplifies the analysis of these phenomena in such a way that the rate point of view and the energy associated with each correspondent transition acquire a preeminent role, as opposed to the off-diagonal density matrix elements used in more popular approaches to this problem. Both points of view lead, of course, to the same result, but we believe that the first one appeals to more fundamental concepts.

1. Collisional dissipation

When a colliding partner interacts with our atom and goes away from it, different components of the final density matrix correspond, in the final wave function of both atoms, to different internal states of the colliding partner. By averaging over the partner states one obtains the atomic density matrix immediately after the collision. In general, we may assume that collisions take place with a rate given by γ and that, as a result of them all coherences are broken and we can associate a given probability with any stationary state of our atom. Since, in the time between one collision and the next one, evolution occurs with the Hamiltonian of Eq. (5), the equivalent density matrix evolves according to

$$\dot{\rho}=\imath[\rho,H]-\gamma\rho+\sum_i \phi_i|i\rangle\langle i|, \tag{14}$$

where the ϕ_i's are the rates into the level $|i\rangle$. These contain all the information about the redistribution of atomic population taking place in the collision and are usually functions of the level's populations [12]. Although a model in which all the coherences have the same decay constant could appear a rather simplified one, it will be, indeed, sufficient for our investigations.

One can now easily derive a sume rule similar to that of Eq. (6) also in the stationary situation by adopting the quantum trajectory point of view corresponding to Eq. (14). We indicate with ϵ_i and ϵ_g the energies delivered into the field during a coherent period starting from the upper states $|e_i\rangle$ and the ground state $|g\rangle$, respectively. With the same kinds of arguments used above, but this time applying Eq. (7) to a coherent period, that lasts γ^{-1} on average, we can derive a sum rule

$$\epsilon_0+\sum_i \epsilon_i=0. \tag{15}$$

We can further notice that these results do not rest on any hypothesis of $|e_i\rangle$ being energy eigenstates. On the contrary, any set of states, as long as they form a complete set, will work. It is also very simple to realize that all the energies ϵ_i from the upper states are greater than or equal to zero: from Eq. (7) we read that for any initial state in the manifold of the upper states the quantity $\langle\psi|P_g|\psi\rangle$ is equal to zero, whereas at the generic time t it is different from zero because $|\psi\rangle$ may have acquired a component into the lower state $|g\rangle$; this implies that absorption from the ground state is always larger than stimulated emission from any of the upper states. Again, in a fashion similar to the case of the pulsed regime seen above, this conclusion can be tailored to the autoionizing states' configuration. We will show in the next section how this point can be better understood within the framework of the procedures usually employed to deal with the dynamical behavior of autoionizing states amplitudes.

2. Radiative dissipation

Radiative dissipation has been the starting point for the development and understanding of quantum trajectories and jumps since these concepts were initially proposed to analyze the statistics of photon emission [9]. Here we describe some of its features, even if we will not use them in our ensuing calculations. In the context of such dissipation one assumes that the spontaneous emission modes are coupled with a broadband detector in order to reveal the emission of a spontaneous emission photon. If we start with a given atomic state after a small time interval Δt two alternatives, properly represented in the final density matrix, are possible: (i) the photon may have been detected with probability $\Gamma\Delta t$ times the probability in the upper level and the atom found in the lower state; (ii) the photon is not detected and the corresponding final atomic wave function is obtained by the result of the evolution of the whole system, atom+radiative modes, corresponding to zero photons [9]. Since the probability must be conserved, this latter state will differ $\mathcal{O}(\Delta t)$ from that at the initial time of this interval; this makes possible the definition of an evolution operator to be included in the Hamiltonian. In fact, according to the Wigner-Weisskopf approximation, we obtain the effective evolution Hamiltonian governing the wave-function evolution between jumps simply by adding the complex factor $-(i/2)\Gamma$ to the energy of the upper level. It has also been shown that the coherent evolution with this effective Hamiltonian corresponds to a picture of a classical dipole emitting a classical field as in a model put forward some time ago [16] and recently reinterpreted in the context of the quantum jumps picture of the dissipation processes [17]. Thus, in the case of spontaneous emission dissipation, the dissipation source acts continuously also on the coherent evolution and accounts for the classical radiation reaction on the atom and for the corresponding loss of energy. On the contrary, the jump is a purely quantum mechanical effect that does not have a counterpart in the classical equations.

III. ABSORPTION AND STIMULATED EMISSION WITH AUTOIONIZING STATES

A. Atomic configuration with one autoionizing state

We will assume in the following a dissipation mechanism that interrupts the coherent evolution at a rate γ as that considered in the previous section. For, as crude as it may be, one can assume this as modeling for the recombination mechanism or for the finite time of interaction between atoms and field. The configuration with one autoionizing state is shown in Fig. 1(b); it is essentially the same configuration treated in [2]. Fano diagonalization techniques allow us to deal only with the amplitude of the autoionizing state. This is obtained by including the effect of the continuum into an effective Hamiltonian that under certain simplifying assumptions, reads [3]

$$H_{\mathrm{eff}}=\hbar\begin{pmatrix} 0 & -\Omega/2 \\ -\Omega/2 & -\Delta\omega \end{pmatrix}-\frac{i\hbar}{2}|R\rangle\langle R|, \tag{16}$$

where $|R\rangle=\sqrt{\Gamma}|1\rangle+\sqrt{w}|\Omega||0\rangle$ is the state effectively coupled to the continuum. Here we have indicated the photoionization rate to the continuum with $w|\Omega|^2$ to make explicit its dependence on the laser field intensity. As a consequence of this choice the standard Fano parameter q is given here by $q=1/\sqrt{\Gamma w}$. We can thus compute absorption and emission by solving the associated Schrödinger equation. This can be done numerically by expanding the wave function in terms of the eigenvectors of the above Hamiltonian. However, since this Hamiltonian is not Hermitian, some care must be taken in the application of this procedure and in this respect the completeness and orthogonality properties of these eigenvectors discussed in [11] are useful. It is interesting to notice here in passing that the condition of Fano interference corresponds to the slowest possible decay for the ground state $|0\rangle$. For small Rabi intensities this condition can be understood without the need of diagonalizing the whole Hamiltonian H_{eff}. Let $|\overline{0}\rangle=|0\rangle-(\Omega/2\Delta\omega)|1\rangle$ be the eigenstate of the Hermitian part of this Hamiltonian that in the limit of small Ω goes into $|0\rangle$; this will not leak into the continuum and will also be an eigenstate of H_{eff} with an eigenvalue having a zero imaginary part only if it is orthogonal to the coupled state $|R\rangle$. This orthogonality requirement leads to $\Delta\omega=\frac{1}{2}\sqrt{\Gamma/w}$, which is the well-known condition for the Fano minimum.

We now calculate the response of this configuration during the coherent evolution period starting in the ground state and in the upper autoionizing state, respectively. If we indicate by a_{ik} the amplitude in the state $|k\rangle$ of a wave function with initial condition in $|i\rangle$, to calculate the absorption we will set $a_{00}(0)=1$ and $a_{01}(0)=0$ as initial condition. At any time we can thus evaluate the energy stored in the atom at the expense of the field by using Eq. (7); this reads

$$\epsilon_a(t)=\hbar\omega[1-|a_{00}(t)|^2]. \tag{17}$$

Interruptions of the wave function's coherent evolution occurring with a probability per unit time γ leads to the average absorbed energy as

$$W_a=\int_0^\infty \epsilon_a(t)e^{-\gamma t}\gamma\,dt. \tag{18}$$

Similarly, the emission from the autoionizing state, as by definition, is calculated by using the initial conditions $a_{10}(0)=0$ and $a_{11}(0)=1$. The decrease of the atomic energy and correspondingly the amount delivered into the field will be given according to Eq. (7) by

$$\epsilon_e(t)=\hbar\omega|a_{10}(t)|^2, \tag{19}$$

while its average value is

$$W_e=\int_0^\infty \epsilon_e(t)e^{-\gamma t}\gamma\,dt. \tag{20}$$

We have computed these quantities, W_a and W_e, as a function of the laser detuning from the autoionizing state energy. For some values of the parameters of the autoionizing state configuration, our results are shown in Fig. 2. As is possible to see from this figure, the emission never overcomes absorption, but is always well below it. Quite unexpectedly, the mechanism of quantum interference that would quench the absorption, but not the stimulated emission [1,4,18], does not seem to work here.

B. Atomic configuration with two autoionizing states

We repeat the same calculations as above for the case of two autoionizing upper levels, the same system that has been treated in Refs. [1,5]. Under the same simplifying assumptions the effective Hamiltonian reads

$$H_{\mathrm{eff}}=\hbar\begin{pmatrix} 0 & -\dfrac{\Omega_1}{2} & -\dfrac{\Omega_2}{2} \\ -\dfrac{\Omega_1}{2} & -\Delta\omega_1 & 0 \\ -\dfrac{\Omega_2}{2} & 0 & -\Delta\omega_2 \end{pmatrix}-\frac{i\hbar}{2}|R\rangle\langle R|, \tag{21}$$

where in this case $|R\rangle=\sqrt{\Gamma_1}|1\rangle+\sqrt{\Gamma_2}|2\rangle+\sqrt{w}|\Omega||0\rangle$, with $\Delta\omega_1$ and $\Delta\omega_2$ being the detunings of the laser fields in the transitions leading to states $|1\rangle$ and $|2\rangle$, respectively. If Δ_{12} is the spacing between these two levels we have $\Delta\omega_2+\Delta_{12}=\Delta\omega_1$. The relation between Ω, Ω_1, and Ω_2 is not our concern here since we will assume a negligible photoionization rate, i.e., $w=0$. This configuration displays interference in the absorption that can be understood qualitatively as associated to the two different paths, represented by the two autoionizing states $|1\rangle$ and $|2\rangle$, leading to the same continuum. In the limit of small fields and nonzero detunings the

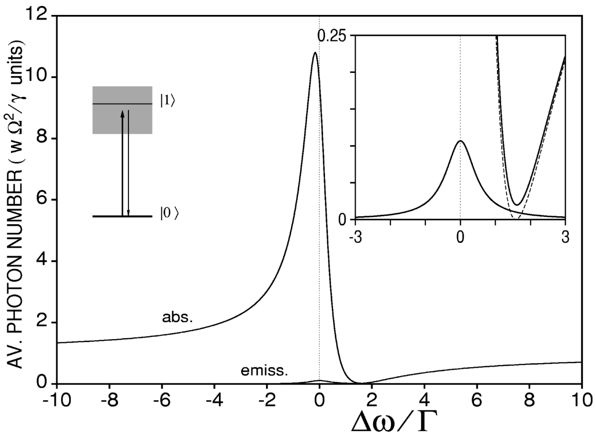

FIG. 2. Absorption W_a and stimulated emission W_e for the case of one autoionizing level. These are measured as the average number of photons absorbed or emitted during a coherent evolution starting from the states $|0\rangle$ and $|1\rangle$, respectively, and are normalized to $w|\Omega|^2/\gamma$, i.e., to the photoionization average photon number. Although W_a looks to be practically indistinguishable from the Fano profile $(\epsilon+q)^2/(1+\epsilon^2)$ with $q=\sqrt{10}$ and $\epsilon=-2\Delta\omega/\Gamma$, a closeup view in the upper right corner, with the dotted line corresponding to the original Fano profile, evidences that the minimum characteristic of the Fano interference is not zero and that stimulated emission is weaker than absorption. The detuning $\Delta\omega$ is measured in units of Γ. Parameter values are $\gamma=0.01$, $\Gamma=1$, $w=0.1$, and $\Omega=10^{-3}$.

vector $|\overline{0}\rangle=|0\rangle-(\Omega_1/2\Delta\omega_1)|1\rangle-(\Omega_2/2\Delta\omega_2)|2\rangle$ obtained by prediagonalizing the bound state Hamiltonian has a zero decay rate if it is orthogonal to the coupled state $|R\rangle$, a requirement that gives $\Omega_1\sqrt{\Gamma_1}/\Delta\omega_1=-\Omega_2\sqrt{\Gamma_2}/\Delta\omega_2$, a condition already known from previous works [6]. Absorption W_a, and stimulated emission W_{e1} from the state $|1\rangle$ are shown in Fig. 3 for the same set of parameters as in Ref. [5].

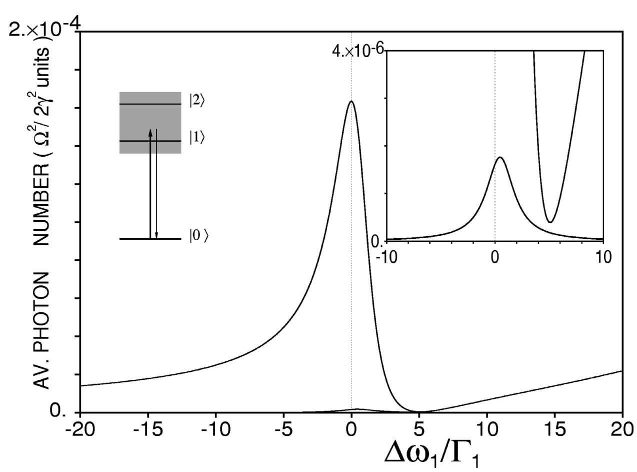

FIG. 3. Absorption W_a and stimulated emission W_{e1} for an atom initially in upper level $|1\rangle$ for a configuration with two autoionizing levels. In the upper right corner there is a magnified view showing details of the absorption and emission near the interference. These quantities are normalized to $(\Omega_1^2+\Omega_2^2)/2\gamma^2=\Omega/2\gamma^2$, i.e., to the average photon number for an equivalent two-level system without losses into the continuum. The detuning $\Delta\omega_1$ is given in units of Γ_1. The parameters are the same of those in [5]: $w=0$, $\Gamma_1=1$, $\Gamma_2=10$, $\Omega_1=1/\sqrt{10}$, $\Omega_2=1$, $\Delta_{12}=55$, and $\gamma=0.01$.

For this same set of parameters W_{e2} was found to be negligible for the range of $\Delta\omega_2$ values plotted in Fig. 3. As in the previous case absorption does not overcome emission and, here too, the argument according to which quantum interference quenches absorption while stimulated emission remains unaffected does not apply.

C. Absorption versus stimulated emission

The main disagreement mentioned above seems to come from the fact that the Fano minimum is not zero as, on the contrary, is found by means of the Fermi "golden rule" in the original work of Fano. However, we should point out that this last way of calculating absorption assumes, as usually occurs in the classical spectroscopy of photoionization, that the autoionizing times, represented here by Γ, are much faster than any other dissipation mechanism, as, for example, recombination or dephasing mechanisms that could be represented here by γ. As a matter of fact, within this last approximation we recover the usual Fano profile, proportional to the square of the transition operator matrix element [3], with the only small but significant difference being that the Fano minimum is not exactly zero. This fact is not new in the spectroscopy of the autoionizing states even though in the experimental setups this difference may pass undetected, since it is indistinguishable from instrumental noise [19]. Lambropoulos and Zoller [20] have shown how the autoionization profiles are modified by the finite time of interaction and how the ideal Fano profile is recovered only in the limiting case of the interaction time, a quantity to be put in relation with γ^{-1} in our model, becoming infinity. However, our calculations go a little further by showing that the Fano minimum in absorption never drops below the stimulated emission that one can obtain by pumping atoms in the autoionizing state. This, in our opinion, is the consequence of a strong unavoidable correlation between absorption and stimulated emission that is to be traced back to their common physical origin, i.e., the atomic dissipation mechanisms. In particular, in our case a pumping mechanism in the autoionizing state at rate γ, while increasing the stimulated emission rate, also has the effect of breaking the phase correlation between the atom and the field. This is equivalent to reducing the interaction time and to increasing the minimum of the absorption profile. A similar behavior was also found by the author in configurations working with bound states where a reduction of absorption is always accompanied by a correspondent reduction of emission, by virtue of certain symmetry relations discussed in [11,12].

D. Can stimulated emission ever overcome absorption?

The impossibility of obtaining stimulated emission greater that absorption with systems using autoionizing states is a simple and general property of the evolutions represented by the Hamiltonians like that of Eq. (16) and Eq. (21). These give rise to a wave-function evolution whose norm decreases in time because of the irreversible leaking into the continuum. In fact, the norm obeys

$$\frac{d}{dt}\langle\psi|\psi\rangle=\left\langle\psi\middle|\left(-\frac{i}{\hbar}H_{\mathrm{eff}}+\frac{i}{\hbar}H_{\mathrm{eff}}^{\dagger}\right)\middle|\psi\right\rangle=-|\langle\psi|R\rangle|^2\leqslant 0, \tag{22}$$

which can immediately be translated into

$$\langle \psi | \psi \rangle \leqslant 1. \tag{23}$$

Let us indicate with $|\psi_i\rangle$ the wave function at time t whose initial condition is the state $|i\rangle$. We can write

$$\sum_k |a_{ik}|^2 = \sum_k \langle \psi_i | k \rangle \langle k | \psi_i \rangle = \langle \psi_i | \psi_i \rangle \leqslant 1 \quad \forall i. \tag{24}$$

Now, by virtue of the symmetry of the coefficients a_{ik} proved in [11], which holds when the effective Hamiltonian is symmetrical, such as the ones we are dealing with, we can write

$$\sum_k |a_{ki}|^2 \leqslant 1 \quad \forall i. \tag{25}$$

This last equation is at the basis of the relationship between absorption and stimulated emission mentioned above. For the case of one autoionizing state this reads $|a_{00}|^2 + |a_{10}|^2 \leqslant 1$, which is equivalent to $\epsilon_a \geqslant \epsilon_e$ and consequently to $W_a \geqslant W_e$. We notice here that at Fano interference the loss term is the smallest possible and the ratio between stimulated emission and absorption, although being the closest to unity, never overcomes such a value. The same can be said with the case of two autoionizing levels: we have $|a_{00}|^2 + |a_{10}|^2 + |a_{20}|^2 \leqslant 1$, which is equivalent to $W_a \geqslant W_{e1} + W_{e2}$. Thus we have proved that these inequalities hold in general and not only for the specific parameters that we have chosen in the previous section.

The physical reason behind this result should be related to the additional energy dissipation channel represented by the ionization of an atom in the upper state, a channel that does not exist for the lower state. In other words, to an atom initially in the upper state two possibilities are given: to create a photon and transform its energy into radiant energy, or to become ionized. In this last case the atomic energy is lost to the amplification process. On the contrary, an atom in the ground state can make transitions only by absorbing a photon. It is therefore clear that, in order to obtain gain, the pumping rate to the upper state must overcome the rate in the lower state. It is quite surprising that the argument of quantum interference used in the investigations of inversionless lasing mechanisms plays no role here. Unfortunately, as is well known, the chances of increasing the number of these channels increases with the upper level energy, thus rendering amplification in the high-frequency region more and more difficult in any kind of atomic system, no matter whether discrete or continuum.

Above we have enforced a rather orthodox point of view. This same point of view leads to the consideration that a loss in the lower level will invert the role of absorption and emission established above. Let us imagine a hypothetical configuration in which the autoionizing state is the lower state, that is, in Fig. 1(b) we exchange upper and lower levels as shown in Fig. 1(c). Now the previous situation is reversed. An atom in the lower state has the possibility of being ionized or making a transition to the upper state, while for an atom in the upper state there is only the possibility of emitting a photon and making a transition to the lower ground state. It is now clear that, in order to have gain, the rate in the upper state must be less than that in the ground state. This behavior is reminiscent of the well-known fact that a fast depletion rate of the lower state will make the amplification process easier. We show that our picture set out above is consistent with these conclusions. If we indicate with $|e,N\rangle$ the upper dressed level and with $|g_i, N+1\rangle$ the set of lower dressed levels, we find, by using the same arguments of Sec. II, that the average change in the photon number is given by

$$\langle \Delta N \rangle = \mathcal{P}_e^{(i)} - \mathcal{P}_e^{(f)}, \tag{26}$$

where $\mathcal{P}_e = |\langle e,N|\psi\rangle|^2$ is the probability for the atom being in the upper state and where $\mathcal{P}_e^{(i)}$ stands for its initial value. Thus stimulated emission, being proportional to $1-|a_{ee}|^2$, exceeds absorption, proportional $|a_{ge}|^2$, by virtue of Eq. (25). However, if seen in terms of populations, zero gain will require more population in the upper level as compared to that in the lower autoionizing state that, because of its fast decaying rate to the continuum, will be quickly depleted. Nonetheless, this would be a more efficient configuration than the one considered in this paper. Indeed, the above described mechanism has some similarities with that operating in dimer excimer lasers where the lower lasing state is unstable.

IV. POPULATION INVERSION VERSUS EFFICIENT GAIN

A. Population of the autoionizing state

Here we come to the main point of the present work; i.e., we discuss in some detail the relation between efficient gain and population inversion. To do so we review some already known results concerning the autoionizing state configuration [1,2]. Although it has been shown that no population inversion between the autoionizing state and the ground state is needed to achieve gain, our point is that this must not be read as a softening of the usual physical requirements associated with it. On the contrary we believe that there is a certain amount of ambiguity in assuming population inversion as indicators of efficient gain.

Let us assume that the dissipation mechanism produces a redistribution of the atomic population between the ground state and the upper autoionizing state with probability π_0 and π_1, respectively, thus the rates in the upper and lower level are $\phi_0 = \gamma\pi_0$ and $\phi_1 = \gamma\pi_1$. Although fully efficient pumping into the autoionizing state is rather hypothetical, and in general also other states of the upper manifold will be pumped, this modeling does serve well to illustrate our ideas. The average gain can be written as

$$W = \phi_1 W_e - \phi_0 W_a = \gamma(\pi_1 W_e - \pi_0 W_a), \tag{27}$$

since the average duration of a coherent period is γ^{-1}. This immediately shows that in order to have gain the rate ϕ_1 of atoms in the upper level must be greater than the rate ϕ_0 in the ground state. The population can also be calculated as the average over the coherent evolution period, we can write

$$\rho_{00} = \gamma\pi_0 \mathcal{P}_{00} + \gamma\pi_1 \mathcal{P}_{10},$$

$$\rho_{11} = \gamma\pi_0 \mathcal{P}_{01} + \gamma\pi_1 \mathcal{P}_{11}, \tag{28}$$

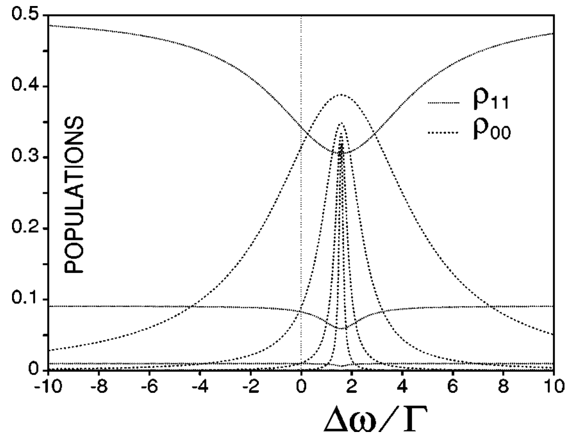

FIG. 4. Population of the ground state ρ_{00} and of the autoionizing state ρ_{11} corresponding to zero gain, for the case of one autoionizing upper level for different values of the parameter $\gamma = 0.001$, 0.01, 0.1, 1. In this figure both ρ_{00} and ρ_{11} monotonically increase with γ. Other parameters are $\Gamma=1$, $w=0.1$, and $\Omega = 10^{-3}$, i.e., the same as those in Fig. 2.

$$\mathcal{P}_{ij} = \int_0^\infty |a_{ij}(t)|^2 e^{-\gamma t} dt. \tag{29}$$

We now choose π_0 and π_1 values in such a way as to satisfy the zero gain or threshold condition in Eq. (27) above and we show in Fig. 4 the ground-state populations ρ_{00} and the upper state population ρ_{11} for different values of γ while the other parameters are the same as those in Fig. 2. In order to read this figure we notice that, with respect to γ, ρ_{00} and ρ_{11} are monotonically increasing. As we can see, near the Fano interference, there is no population inversion and this corresponds to the results found in [2,7,6]. In particular for small γ the upper autoionizing state population becomes vanishingly small. In Fig. 4 the population ρ_{11} corresponding to $\gamma = 0.001$ is practically indistinguishable from the zero level. This, however, only partially describes the situation, since, as we can see from this same figure, the total population of the group of the upper levels, given by $1-\rho_{00}$, is always larger than the ground-state population. Especially in the case corresponding to small values of γ the autoionizing state is found almost empty and the continuum always contains most of the population. This has a simple explanation that is due to the very fast decay to the continuum of the autoionizing state. For small fields the populations are given by

$$\rho_{00} = \gamma \pi_0 \mathcal{P}_{00} = \pi_0, \quad \rho_{11} = \gamma \pi_1 \mathcal{P}_{11} = \pi_1 \frac{\gamma}{\Gamma+\gamma}, \tag{30}$$

where the expression for ρ_{11} simply reflects the fact that population in the upper states is shared between the autoionizing state and the continuum in the ratio γ/Γ. This explains the necessity of a large pumping rate in the upper autoionizing state in spite of the small population in this state. Such a large rate is a signature of the considerable amount of energy that has to be pumped in the autoionizing state and ultimately into the whole system. This is not a negligible one, as instead is the population in the upper state, but, on the contrary, it is even larger than the power required by the pumping process in a similar two-level system with no losses into the continuum. As a matter of fact, this last system would require, at the threshold, the same rate in the upper and in the lower level.

We can generalize the results above for any number of autoionizing states. In fact, we can prove that the threshold population in the ground state must always be less than half of the total population. Thus, rewriting the first of Eq. (28) and recalling that $\mathcal{P}_{00} = (1 - W_a/\hbar\omega)/\gamma$ and $\mathcal{P}_{10} = W_e/\hbar\omega\gamma$ we obtain with the condition of zero gain

$$\rho_{00} = \pi_0 \left(1 - \frac{W_a}{\hbar\omega}\right) + \pi_1 \frac{W_e}{\hbar\omega} = \pi_0 < \frac{1}{2}, \tag{31}$$

thus proving our point. The same can be said for the system with two autoionizing states. In this case the population in the ground state can be written as

$$\rho_{00} = \gamma(\pi_0 \mathcal{P}_{00} + \pi_1 \mathcal{P}_{10} + \pi_2 \mathcal{P}_{20}) \tag{32}$$

that in the condition of zero gain

$$\pi_0 W_a = \pi_1 W_{e1} + \pi_2 W_{e2} \tag{33}$$

becomes $\rho_{00} = \gamma \pi_0$. Now the condition above implies that $\pi_0 < \frac{1}{2}$: if it were not so the left-hand side of Eq. (33) would be greater than $W_a/2$ while the right hand side would be less than this same value by virtue of the fact that $\pi_1 < \frac{1}{2}$, $\pi_2 < \frac{1}{2}$ and $W_a \geqslant W_{e1} + W_{e2}$.

B. Efficiency of the lasing schema

So far two points have been established by our calculations: (i) Absorption is always larger than stimulated emission and (ii) at threshold atoms in the ground state are always less than half of the total. Point (i) implies that to have gain the rate in the upper level must be greater than that in the ground state. This indicates that the requirement for obtaining gain is in any case a demanding one, in spite of a very small population in the upper state. Point (ii) is rather marginal in our argument, since it refers to populations, but it should be one more confirmation that the autoionizing system needs to be driven far away from equilibrium if gain is to be obtained. Of course this has its costs in terms of energy pumped into the system.

We believe that these points are a sufficient basis to argue that the involved configuration is far from being an efficient one. Usually, in laser designing, efficiency is understood as the ratio between the energy transformed in coherent radiation and that required for the pumping process. This can be further specialized in terms of the threshold pumping power and of the differential efficiency, i.e., the change in output lasing power produced by increasing of one unity the pumping power. Obviously the theoretical limit for the lasing threshold rate is set by the rate at which atoms must be pumped to the upper level of the lasing configuration. When this rate is prohibitively high amplification is not practically feasible. Thus the autoionizing states' configuration that we have discussed here has a lower efficiency than that of its companion configuration in which the upper level is not coupled to a continuum, i.e., of a simple two-level system. In fact in this latter system the threshold rate in the upper level

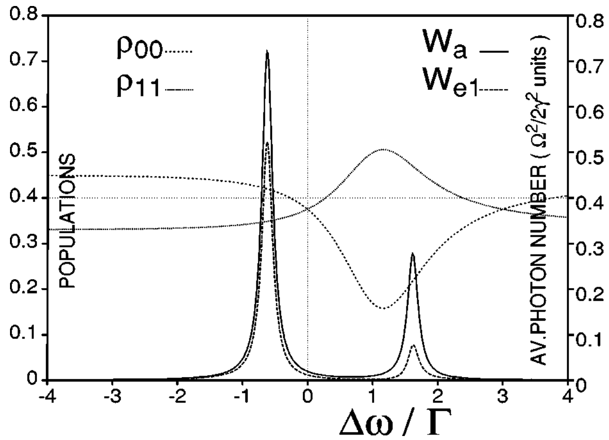

FIG. 5. Absorption W_a, stimulated emission W_{e1} from state $|1\rangle$, ground-state population ρ_{00}, and excited-state population ρ_{11} for an atomic system with two upper levels, as a function of the detuning from the bare energy of level $|1\rangle$ in units of Γ. Parameters are $\gamma=0.1$, $\Gamma=2$, $\Omega_1=\Omega=0.001$, and $\Delta_{12}=1$.

must be the same as the rate in the lower level, whereas, in the autoionizing states configuration the former must exceed the latter.

The considerations made above lead one to rethink the role of the level's populations as meaningful parameters that determine the gain. From a theoretical point of view the picture of individual trajectories helps us in doing so. The essential point here is that both the gain and the pumping are rate processes; we usually think of gain in terms of the photon's rate and of the pumping mechanisms in terms of pumping rates. On the contrary, population is an average over a long period of time in atomic evolution. Thus the photon's emission and absorption are more naturally related to the level's rates rather than to atomic populations. Thus, in this framework, the relation between population and gain that holds in a two-level system can be considered just as a remarkable exception. Indeed, population is related to gain only and exclusively in the case of a two-level system insofar as it establishes a condition at the atomic level to reach the lasing threshold, and this with no regard to the physical feasibility of this condition. As a matter of fact, not all the lasing configurations working with the standard schema have the same efficiency while they all satisfy to the same condition of population inversion. What instead makes the difference is the rate in the upper level required for the lasing threshold.

Along the same line, an approach for systems working with bound states and where the role of level rates has been put in due relevance in the physical equations that determine the gain has been developed by the author. Here the contribution of each multiphoton process adds to the total gain while the gain of each process depends on the difference between the rate of the initial and final level [12,13]. Thus a useful criterion for the lasing threshold condition still exists for more complex systems if one formulates it in terms of rates. By pursuing such an approach one combines the symmetry of the resulting equations with a formulation in terms of quantities, the rates, which are directly involved in the assessment of the efficiency of a given configuration.

C. Similarities with the behavior of a discrete system

To conclude our discussion we stress again the similarity initially proposed between the level structure of an autoionizing state atomic configuration and that of a bound state atomic configuration. We would like to show that the same behavior, concerning the possibility of lasing without population inversion in the autoionizing configuration, can be found even when the upper level manifold contains just two states $|e_1\rangle$ and $|e_2\rangle$. This result is of course well known, but it is worth emphasizing in this context. Here the laser field couples only state $|g\rangle$ with state $|e_1\rangle$, the target state, to which atoms are pumped with a rate $\phi_e=\gamma\pi_e$. The modeling Hamiltonian can thus be written as

$$H=\hbar\begin{pmatrix} \Delta\omega & -\dfrac{\Omega_1}{2} & 0 \\ -\dfrac{\Omega_1}{2} & 0 & \dfrac{\Gamma}{2} \\ 0 & \dfrac{\Gamma}{2} & \Delta_{12} \end{pmatrix}, \tag{34}$$

where the off-diagonal term Γ ensures that both $|e_i\rangle$ are not eigenstates of the bare Hamiltonian. This is the common point with the autoionizing state configuration since the autoionizing state is not a stationary state. We have already seen in Sec. II C 1 that the emission from any one of the upper manifold states is necessarily smaller than the absorption from the ground state. Thus, in order to have gain, the rate $\phi_e=\gamma\pi_e$ of the excited state must exceed that of the ground state $\phi_g=\gamma\pi_g$. This also means that, for small fields, the total population of the upper states must exceed that in the ground state, in fact, in this case the first of Eq. (30) still holds. However, the population ρ_{11} of the target state $|e_1\rangle$ shows a different behavior. To check this we numerically solve the equation for the populations. The result is shown in Fig. 5 where the absorption and stimulated emission from level $|e_1\rangle$ are shown together with ρ_{11} and ρ_{00}, populations of $|e_1\rangle$ and $|g\rangle$, respectively, corresponding to the lasing threshold. One notices that, for negative detuning, ρ_{11} is less than the ground-state population ρ_{00}, thus showing a striking similarity with the autoionizing state configuration. Here too the same basic mechanism described in the previous section is operative by providing oscillations or transfer of the population between the upper states. It is, in fact, easy to show that for small fields we have

$$\rho_{11}=\gamma\pi_1\mathcal{P}_{11}=\pi_1\left(1-\frac{\Gamma^2/2}{\Gamma^2+\gamma^2+\Delta_{12}^2}\right), \qquad \rho_{22}=\gamma\pi_1\mathcal{P}_{12}=\pi_1\frac{\Gamma^2/2}{\Gamma^2+\gamma^2+\Delta_{12}^2}. \tag{35}$$

Thus, in a situation such as that corresponding to Fig. 5, where for negative detuning absorption and emission are almost equal, the threshold population in the upper states must be almost equal to that in the lower one. However, the way of partitioning the upper manifold population among the two upper levels as indicated by Eq. (35) (for the parameters corresponding to Fig. 5 it results that $\rho_{22}\approx\frac{2}{5}$ of the total upper state population) allows one to have gain without population inversion. Needless to say, the efficiency here is lower than that of a simple two-level atomic system.

V. CONCLUSIONS

We have shown that the original proposal for lasing without inversion using autoionizing states should not be identified with the proposal for an efficient lasing configuration. Although we have studied here a particular and simple model of dissipation and, from the strict logical point of view, the conclusions derived should pertain only to this model, we have reasons to believe, reasons also supported by our previous work, that the above conclusions are rather general. On the basis of our calculations, two results are in open contrast with the existing point of view on these physical mechanisms. The first one relates to the idea that interference would prevent absorption, but not stimulated emission, i.e., that it would break the symmetry inherent in these two processes: This was found to have no support in our calculations, nor in the very simple energy arguments used to complement them. The second one concerns the choice of the population inversion as an indicator of efficient gain implicitly made in the previous works, on these and alike configurations. The rather ambiguous role that populations play in the autoionizing states' configurations, and in the three-level system discussed here to complement our findings, leads us to support the conclusion that they are often a rather uncertain indicator of gain efficiency and, more generally, on the basis of their microscopical meaning, that they are not directly correlated to the processes of emission and absorption. This, in a certain sense, should be rather evident and easily understood also from the following considerations. To our knowledge there is no proof that the principle of population inversion can be extended to configurations more complex than a two-level system. Thus, one should not be surprised to find situations where gain is not constrained by population inversion. It is, however, a completely different issue to associate the existence of such gain with the efficiency of the amplification process, an issue of which we have tried to make a critical examination in the present work.

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