Symmetries in the optical Bloch equations and multiphoton processes

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> The optical Bloch equations exhibit symmetries that are directly related to the probabilities of the processes of absorption and stimulated emission. As a matter of fact, a picture similar to that holding for a two-level system can be recovered also in multilevel systems and, in particular, for any multiphoton process tuned between a given couple of levels. Thus, new quantities, such as the generalized Einstein *B* coefficients, directly related to the probabilities of the involved process, and atomic level indicators, whose inversion signals the existence of gain for that given process, can be defined. It is shown how these lead to valuable conceptual simplifications and to the disclosure of new insights into some popular problems of quantum optics. In particular, the issue of an efficient gain process to be realized within a multilevel atom-field configuration, a problem closely related to the widely investigated problem of lasing configurations without population inversion, can be viewed entirely as a rate problem and therefore analyzed with increased confidence.

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I. INTRODUCTION

The optical Bloch equations describe the interaction of an atomic system, schematically represented with a finite, usually small, number of levels, with coherent electromagnetic radiations. It is therefore the main tool and the most popular model for the investigation of a broad range of phenomena related to the field of quantum optics. One of its distinctive and successful feature is that of including the influence of the dissipative sources in a very simple, though very general, way $[1]$. Therefore, it deals primarily with the density matrix of the atomic system, although its evolution in time can alternatively be viewed as wave function evolution interrupted by stochastic impulsive perturbations having their origin in the dissipation sources interacting with the atomic system $\lceil 2 \rceil$.

One of the first problems where the optical Bloch equations were used was in the calculation of absorption and emission of atomic systems in the nonlinear regime, as for example the laser equations, where the Fermi golden rule could no longer be applied. A similar kind of problem has recently recurred in the context of lasing without inversion where it was pointed out that the scope of the well-known principle of inversion, relating radiation amplification to the existence of a population inversion in the active medium, does not apply to the case of multilevel lasing configurations [3]. In fact, the optical Bloch equations (OBE) for such systems do not explicitly display the symmetries related to absorption and stimulated emission of photons on which the standard principle of inversion rests. Thus, the question of whether such symmetries are instead hidden in the structure of these equations, or not present at all, has received some attention in the past $[4]$. An important step showing their relation with the underlying physical basic mechanisms of absorption and stimulated emission was carried out by means of the quantum jump picture of the dissipation processes [5]. Such a modeling of the dissipation processes, which applies when the dephasing mechanisms are much faster than atomic evolution, has been shown to play an important role in some

stochastic formulations of the quantum mechanics theory of measurement $\vert 6 \vert$. The ideas that will be presented here, showing the connection between the symmetries in the OBE and the probabilities of the active multiphoton processes, are a generalization of those expressed in the previous works [4,5]. Here we establish a set of rate-balance equations where the probabilities of all the involved multiphoton processes appears directly as coefficients in these linear equations. These are of the same kind of the equations first derived in [4] under certain special conditions. In particular, the gain of any multiphoton process can be written in the same way as that of a two-level system where the populations are replaced by new variables which depend only on the atomic level: in this context the generalized Einstein *B* coefficients are introduced. These equations can be solved for the populations or for the level rates beforehand to the calculation of the gain. Thus the analysis of the configuration response to the electromagnetic fields can be set about principally as a rate problem.

This paper is organized in the following way. In Sec. II, a symmetry relation existing in the mathematical structure of the OBE is derived. In Sec. III, a physical picture of the multiphoton processes which relies on the above symmetry relations is presented. The calculations of the *B* coefficients for the two- and three-level configurations are carried out in Sec. IV. In particular, in the case of a three-level system, we give a closed expression that can be applied to several configurations already studied in the framework of lasing without inversion. In Sec. V we present two applications of our equations. The first one is concerned with quite an old problem in quantum optics, i.e., the occurrence of dark resonances in atomic systems. Here it is shown how the well known reduction in absorption that is experienced in this configuration can be related to the quenching of the onephoton absorption probability to the upper state introduced in the present work. In the second example, a Λ and a *V* atomic configuration are investigated in relation to the possibility of providing efficient gain of radiation in the region of short wavelength.

II. SYMMETRIES IN THE OPTICAL BLOCH EQUATIONS

A. The Optical Bloch equation

We write down, in a rather general form, the optical Bloch equation (OBE) for an atomic system with *N* discrete levels as

$$
\rho = i[\rho, H] - \sum_{i \neq j} \gamma_{ij} P_i \rho P_j - \sum_i \gamma_i P_i \rho P_i + \sum_i \Phi_i P_i
$$
 (1)

or equivalently, in a more compact way, as

$$
\dot{\rho} = -i\mathcal{L}\rho + \sum_{i} \Phi_{i}P_{i}.
$$
 (2)

Here, we introduce the the Liouvillian operator $\mathcal L$ which can be expressed in terms of the Hamiltonian operator *H* of the depletion rates of the level i, γ_i , and of the coherence decay γ_{ii} . It is customary to term the equation

$$
\hat{\rho} = -\iota \mathcal{L} \hat{\rho} \tag{3}
$$

as to be the free or coherent evolution because it can usually be made equivalent to the evolution of an effective atomic wave function by means of a proper Schrödinger equation [2]. On the contrary, the second term $\Phi_i P_i$, where P_i is the projection operators in the state $|i\rangle$, is associated to the feeding term of the level *i*. The scalar quantities Φ_i are the level rates, or input flow, and can be written in terms of the level populations as follows:

$$
\Phi_i = \sum_k \; \pi_{i,k}(\gamma_k \rho_{kk}), \qquad (4)
$$

where π_{ik} are the branching probabilities to the level *i* of a decay that occurs from the level *k* and γ_k is the depletion rate of the same level. The above expression for the density matrix ρ can be also given an integral representation as

$$
\rho(t) = \int_{-\infty}^{t} \sum_{i}^{N} \hat{\rho}_i(t - t') \Phi_i(t') dt', \qquad (5)
$$

FIG. 1. Multilevel atom-field configuration without closed loops of the kind discussed in this work. The transition 4–5 has been put in as evidence for purposes related to Sec. III.

where $\hat{\rho}_i(t)$ represents the evolution of $\hat{\rho}$ according to Eq. 3 with the initial condition $\hat{\rho}(0) = |i\rangle\langle i|$. This representation is central to the development of the concepts which will be introduced here.

We will focus here on particular configurations where the rotating wave transformation, followed by the rotating wave approximation, leads to a time-independent Hamiltonian and to a stationary solution of the OBE. It is well known that this becomes possible when the diagrammatic representation of the atom-field configuration presents no closed loops $[7]$. One example of such a configuration is shown in Fig. 1. The stationary condition can be simplified to

$$
\rho = \sum_{i}^{N} \Lambda_{i} \Phi_{i}, \qquad (6)
$$

where

$$
\Lambda_i = \int_0^\infty \hat{\rho}_i(t) dt.
$$
\n(7)

Thus, the quantities Λ_i obey to the equation

$$
P_i = i \mathcal{L} \Lambda_i \tag{8}
$$

which is obtained by integrating Eq. (3) from 0 to ∞ .

B. Symmetry of the coefficients $(\Lambda_i)_{kk}$

There is an interesting symmetry concerning the quantities Λ defined above. This involves the populations and is therefore reminiscent of the original formulation of the principle of inversion in which populations play an important role. However, in the present case the populations are fictitious because Eq. (3) , which governs their evolution, does not contain the feeding terms, which are, instead, present in the complete evolution equation. Nonetheless, we can state that, under such an evolution, the average population in state k , as defined in Eq. (7) , given that the system starts in state i , is the same of that in the state *i*, given that the system initiate in state *k*. Indeed, we can write

$$
(\Lambda_i)_{kk} = (\Lambda_k)_{ii}.
$$
 (9)

The first step necessary to prove this relation concerns the symmetry of the Liouvillan operator \mathcal{L} , i.e., $\mathcal{L} = \mathcal{L}^t$. To this end we remind the reader that the operator $\mathcal L$ is defined on the vector space spanned by the vectors $|k\rangle\langle i| = |k,i\rangle$ as *i* and *k* range over the atomic vector space. In this space the scalar product between two vectors $\psi = |\psi_1, \psi_2\rangle = |\psi_1\rangle \langle \psi_2|$ and ϕ $=|\phi_1, \phi_2\rangle = |\phi_1\rangle \langle \phi_2|$, which are operators in the original atomic space, is given by Tr($\phi^+\psi$), that is,

$$
\langle \phi_1 \phi_2 | \psi_1 \psi_2 \rangle = \langle \phi_1 | \psi_1 \rangle \quad \langle \psi_2 | \phi_2 \rangle. \tag{10}
$$

Now, $\mathcal L$ can be split as $\mathcal L = \mathcal L_H + \mathcal L_D$, where $\mathcal L_D$, which is the term accounting for the dissipation, is surely symmetrical since it is diagonal. The symmetry of the component corresponding to the Hamiltonian evolution, i.e., of \mathcal{L}_H , is then proved as follows. We write

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$$
\langle i',j'| \mathcal{L}_H | i,j \rangle = \langle i' | \{ \mathcal{L}_H | i \rangle \langle j | \} | j' \rangle
$$

$$
= \langle i' | \{ H | i \rangle \langle j | - | i \rangle \langle j | H \} | j' \rangle
$$

$$
= \delta_{j,j'} \langle i' | H | i \rangle - \delta_{i',i} \langle j | H | j' \rangle. \tag{11}
$$

Exchanging indices *i* with i' and *j* with j' the result does not change since $\langle i'|H|i\rangle = \langle i|H|i'\rangle$. This last equality arises from the fact that, in configurations without closed loops, it is possible, by properly choosing the phases of the atomic states, to make all Hamiltonian matrix elements real: thus $\langle i'|H|i\rangle = \langle i|H^{\dagger}|i'\rangle = \langle i|H|i'\rangle$, from which it follows that the Liouvillian operator is symmetrical.

The next step is now to prove Eq. (9) . By recalling that the coherent evolution with the initial state $|i\rangle$ is given by

$$
\hat{\rho}_i(t) = e^{-i\mathcal{L}t} |i, i\rangle,\tag{12}
$$

it is then sufficient to prove the equality

$$
\langle k,k|e^{-i\mathcal{L}t}|i,i\rangle = \langle i,i|e^{-i\mathcal{L}t}|k,k\rangle.
$$
 (13)

It is to be noted here that, although $\mathcal L$ is a symmetrical operator, it is, indeed, non-Hermitian because some of its diagonal elements may be complex numbers. Thus, its right and left eigenvectors

$$
l_i|l_i\rangle = \mathcal{L}|l_i\rangle,\tag{14}
$$

$$
l_i(l_i) = (l_i | \mathcal{L}, \tag{15}
$$

no longer coincide. However, they satisfy the following relation

$$
|l_i\rangle = |l_i\rangle^*.\tag{16}
$$

This is proved by first taking the adjoint of Eq. (15), $l_i^*|l_i\rangle$ $=\mathcal{L}^{\dagger}|l_i\rangle=\mathcal{L}^{\dagger}|l_i\rangle$, and next its complex conjugate, $l_i|l_i\rangle^*$ $=\mathcal{L}(l_i)^*$. The Equation (16) follows from this equation and from Eq. (14) , in the assumption that the spectrum is not degenerate. We avoid here entering into unnecessary details of degenerate eigenvalues since the final conclusions will remain unchanged regardless. We can now use an eigenvector expansion of Eq. (12) , based on the completeness $\sum_i |l_i\rangle\langle l_i| = \sum_i |l_i\rangle\langle l_i| = id$ and normalization $\langle l_i | l_i \rangle = (l_i | l_i \rangle)$ $= \delta_{i,j}$ relations [8], as given by

$$
\langle k, k | e^{-i\mathcal{L}t} | i, i \rangle = \left\langle k, k | e^{-i\mathcal{L}t} \sum_{s} | l_{s} \rangle (l_{s} | i, i) \right\rangle
$$

$$
= \sum_{s} e^{-il_{s}t} \langle k, k | l_{s} \rangle (l_{s} | i, i) \rangle
$$

$$
= \sum_{s} e^{-il_{s}t} \langle i, i | l_{s} \rangle (l_{s} | k, k) = \langle i, i | e^{-i\mathcal{L}t} | k, k \rangle,
$$
 (17)

where the symmetry of $\mathcal L$ has entered in the third step of the equality above. In fact, by using Eq. (16) we get $\langle k,k|l_{s}\rangle\langle l_{s}|i,i\rangle=\langle l_{s}|k,k\rangle^{*}\langle i,i|l_{s}\rangle^{*}=\langle i,i|l_{s}\rangle\langle l_{s}|k,k\rangle.$ This proves Eq. (13) : by further integrating it over time we get the symmetry relations of Eq. (9) .

III. MULTIPHOTON PROCESSES

We now wish to establish a connection between the results obtained in the previous section and the concept of multiphoton processes. We will first state and later prove a picture that is both simple and extremely intuitive, but which, nonetheless, has never received a rigorous proof. To this end let us consider a given multifield configuration without closed loops as shown in Fig. 1. Any path between two given levels corresponds to a multiphoton process. If we consider a given transition in the structure, as for example transition 5-4 in Fig. 1, there are several multiphoton processes to which it belongs, namely 5-4, 6-4, 7-4, 5-3, 6-3, 7-3, 5-1, etc. To classify these processes, it is convenient to partition all of the levels into absorbing and emitting ones with respect to a given transition. For example, in the case of the transition 5-4, given that the energy of level 5 is greater than that of level 4, levels 7, 6, 5 are emitting whereas levels 4, 3, 2, 1 are absorbing ones. Thus, any given transition will be a segment of each multiphoton process initiating in an emitting level and ending in an absorbing one. The rate of energy in the electromagnetic (EM) field tuned with this transition is found, quite simply, by summing the rates of all the multiphoton processes to which it belongs. Thus, we can write the total photon rate on the transition from 5 to 4 as

$$
\frac{d}{dt}n_{5,4} = \sum_{i',i} \mu_{i',i},\tag{18}
$$

where i' ranges over the emitting nodes and i over the absorbing ones. Moreover, for any multiphoton process its global rate can be written as

$$
\mu_{i',i} = B_{i',i} \quad \left(\frac{\Phi_{i'}}{\gamma_{i'}} - \frac{\Phi_i}{\gamma_i}\right),\tag{19}
$$

where i' stands for the emitting level, i for the absorbing level, and γ_i and γ_i the relative depletion rates. The first term of this expression is identified with the stimulated emission rate and the second with the absorption rate. The coefficients B_i _{*i*,*i*} = B_i _{*i*,*i*} are shown to coincide with the Einstein *B* coefficients in the limiting case of a two-level system and are therefore named in the following generalized Einstein *B* coefficients. It is important to notice that the absorption and stimulated emission rates are proportional to input flow Φ_i of the involved levels: these quantities are merely what is usually known as the level rate. Thus

$$
P_{i,j} = \frac{B_{i,j}}{\gamma_i} \tag{20}
$$

can be interpreted as the probability of the process initiating in level *i* and ending in level *j*. These probabilities satisfy the relation

$$
\frac{P_{i,j}}{P_{j,i}} = \frac{\gamma_j}{\gamma_i},\tag{21}
$$

which was implicitly assumed in $[4]$, in the particular case of $\gamma_i = \gamma_i$, and later proved in [5] in the more general case. In the analysis of the multilevel system we will employ both the expression for the gain in terms of the *B* coefficients and of the probabilities *P*, according to the better convenience. We also note from Eq. (19) that in the case in which the flow in the absorbing and emitting levels is the same, a strong depletion rate of the upper level enhances the stimulated emission, a feature that, as we know, is at the basis of most of the feasible lasing configurations.

A. Generalized Einstein *B* **coefficients**

In this subsection, we focus on the mathematical proof of the picture presented above. To do this, we start from Eq. (3) applied to the most general atomic configuration, with many levels and fields tuned between them, an example of which is shown in Fig. 1. This gives

$$
\hat{\rho}_{qq} = -\gamma_q \hat{\rho}_{qq} + \sum_k \frac{i}{2} \Omega_{qk} (\hat{\rho}_{qk} - \hat{\rho}_{kq}), \qquad (22)
$$

which integrated from 0 to ∞ , with the initial state being $|s\rangle$, yields

$$
(\hat{d}_s)_q = \sum_k (\hat{n}_s)_{k,q} + \delta_{s,q}
$$
 (23)

with the obvious identification $(\hat{d}_s)_q = \gamma_q(\Lambda_s)_{qq}$ and $(\hat{n}_s)_{q,k}$ $=(\iota/2)\Omega_{qk}(\Lambda_{kq}-\Lambda_{qk})$, the sum above ranging over all the levels *k* that are connected to the level *q* with a field Ω_{ak} . For any level different from the initial one, it helps to pictorially associate such an equation with a sink in the level *q*, represented by $(\hat{d}_s)_{q}$, and with currents $(\hat{n}_s)_{k,q}$ going from the neighboring levels *k* to the sink level *q*. Let us now consider a given transition from *i* to *j* and a level *s* which is absorbing with respect to such transition. It is easy to show that

$$
(\hat{n}_s)_{i,j} = -\sum_{s' \in \text{emi}} (\hat{d}_s)_{s'}, \tag{24}
$$

where s' ranges over the emitting levels of the above transition. In fact, by virtue of Eq. (23) and of $(\hat{n}_s)_{q,k}$ $-(\hat{n}_s)_{k,q}$, the sum in the second part of the above equation contains currents that cancel out in any segment except that in the one corresponding to the chosen transition. In a similar way, given an emitting level s', we can write

$$
(\hat{n}_{s'})_{i,j} = \sum_{s \in \text{abs}} (\hat{d}_{s'})_s, \tag{25}
$$

where *s* now ranges over the absorbing levels.

Now, by using Eq. (6) , we can calculate the photon's rate on the given transition *i* to *j* by means of the corresponding off-diagonal matrix element [9]. We thus find

$$
\frac{d}{dt}n_{i,j} = \sum_{s} (\hat{n}_{s})_{i,j} \Phi_{s} + \sum_{s'} (\hat{n}_{s'})_{i,j} \Phi_{s'},
$$
 (26)

where the sum has been split into one over the emitting nodes and the other over the absorbing nodes. Finally we get

$$
\frac{d}{dt}n_{i,j} = -\sum_{s} \sum_{s'} (\hat{d}_{s})_{s'} \Phi_{s} + \sum_{s'} \sum_{s} (\hat{d}_{s'})_{s} \Phi_{s'}
$$

$$
= \sum_{s,s'} (\hat{d}_{s})_{s'} \Phi_{s} - (\hat{d}_{s'})_{s} \Phi_{s'} = \sum_{s',s} \mu_{s',s} \qquad (27)
$$

and

$$
\mu_{s',s} = (\hat{d}_{s'})_s \Phi_{s'} - (\hat{d}_s)_{s'} \Phi_s = B_{s',s} \left(\frac{\Phi_{s'}}{\gamma_{s'}} - \frac{\Phi_s}{\gamma_s} \right) (28)
$$

having defined

$$
B_{s,s'} = \gamma_s \quad (\hat{d}_s)_{s'} = \gamma_s \gamma_{s'} (\Lambda_s)_{s',s'} = \gamma_s \gamma_{s'} (\Lambda_{s'})_{s,s} = B_{s',s}.
$$
\n(29)

The first term of $\mu_{s',s}$ corresponds to the stimulated emission process rate, whereas the second one corresponds to the absorption rate. Moreover, by integrating over the time the equation

$$
\frac{d}{dt}\text{Tr}(\hat{\rho}_i) = -\sum_k \gamma_k(\hat{\rho}_i)_{k,k} \tag{30}
$$

and accounting for the initial condition on $\hat{\rho}$, it results

$$
\sum_{k} \frac{B_{i,k}}{\gamma_i} = \sum_{k} P_{i,k} = 1, \tag{31}
$$

which embodies the probabilistic meaning connected to the *B* coefficients. It is to be pointed out here, in relation to the expansion of these processes in terms of higher order virtual multiphoton processes initiating in *i* and ending in *k*, that B_{ik} corresponds to the complete summation of these latter. Therefore, it takes into account all interferences that may occur between them.

B. Rate-balance equations for the level's populations

Perhaps the most interesting consequence of viewing the optical Bloch equations from the perspective set forth above is that the interaction between atom and field can now be understood in terms of rate processes rather than by means of atomic coherences. Thus it is possible to write a balance equation relating the rates in and rates out for each given atomic level. To show this, let us start from Eq. (6) , take the average over the state *k* and multiply by γ_k ,

$$
\gamma_k \rho_{kk} = \sum_{i \neq k} \gamma_k (\Lambda_i)_{kk} \Phi_i + \gamma_k (\Lambda_k)_{kk} \Phi_k.
$$
 (32)

Upon transforming the last term by means of Eq. (31) , we get

$$
-\gamma_k \quad \rho_{kk} + \Phi_k + \sum_{i \neq k} \mu_{ik} = 0, \tag{33}
$$

whose meaning in terms of photon absorption and stimulated emission arises quite naturally. The first two terms in the above equation represent, respectively, the depletion rates and the input rate of the state *k* due to the dissipation processes only. The coherent fields contribute to this balance with stimulated emission processes ending in state *k* with a rate $P_{i,k}$ $\Phi_i = B_{ik}(\Phi_i / \gamma_i)$ and with absorption processes starting in state *k* with rate $P_{k,i}$ $\Phi_k = B_{ki}(\Phi_k / \gamma_k)$. The equation above can be solved for the populations or, according to the needs, directly for the level rates. In this last case the normalization equation $\Sigma_i \rho_{ii} = 1$ and

$$
\Phi_i = \sum_k \; \pi_{ki}(\gamma_k \rho_{kk}) \tag{34}
$$

can be used to express the populations in term of the rates that are then put back into Eq. (33) .

C. Limiting cases

It is easy to show that in the limit of small fields one recovers the standard picture of absorption and stimulated emission in which the global gain for each process is proportional to the population difference between its initial and final state. Under these conditions, the rates of the multiphoton processes in Eq. (33) are negligible. Thus we have Φ_i $= \gamma_i \rho_{ii}$, and μ_{ik} is given by

$$
\mu_{ik} = B_{ik} \left(\frac{\Phi_i}{\gamma_i} - \frac{\Phi_k}{\gamma_k} \right) = B_{ik} \left(\rho_{ii} - \rho_{kk} \right). \tag{35}
$$

This relation substantiates the naming of generalized Einstein *B* coefficients adopted for the quantities B_{ik} .

Another limiting case concerns the gain of a two-level system. We recover here the usual expression for gain proportional to the population inversion. By indicating with 1 the lower level and with 2 the upper one, the following equations hold:

$$
B\left(\frac{\Phi_2}{\gamma_2} - \frac{\Phi_1}{\gamma_1}\right) = \mu_{21},
$$

$$
-\gamma_2 \rho_{22} + \Phi_2 - \mu_{21} = 0,
$$
 (36)

$$
-\gamma_1\rho_{11}+\Phi_1+\mu_{21}=0.
$$

By multiplying the first one by γ_1 , the second one by γ_2 , and by subtracting the resulting equalities and using the expression found above for μ_{21} , we obtain

$$
\mu_{21} = \frac{B}{1 - \frac{(\gamma_2 + \gamma_1)}{\gamma_1 \gamma_2} B} (\rho_{22} - \rho_{11}).
$$
\n(37)

IV. *B* **COEFFICIENTS IN SOME SIMPLE SPECIAL CASES**

A. Two-level atomic system

We consider now a two-level system driven by a coherent field with lower and upper levels damped at rates γ_1 and γ_2 , respectively. In order to calculate the absorption and stimulated emission, according to Eq. (29) , we must compute $(\Lambda_1)_{22}$. The operator Λ_1 fulfills the following equations (the index 1 has been dropped to simplify the notations), as from Eq. (3) and Eq. (7) :

$$
\frac{i}{2}\Omega^2(\Lambda_{12}-\Lambda_{21})-\gamma_1\Lambda_{11}=-1,
$$

$$
-\gamma_{12}\Lambda_{12}+\frac{i}{2}(\Lambda_{11}-\Lambda_{22})=0,
$$
 (38)

 $\gamma_1\Lambda_{11}+\gamma_2\Lambda_{22}=1$,

where the RW Hamiltonian is given by

$$
H = \begin{pmatrix} \delta & \frac{\Omega}{2} \\ \frac{\Omega}{2} & 0 \end{pmatrix}
$$
 (39)

and the substitutions $\Lambda_{12} \rightarrow \Omega \Lambda_{12}$ and $\gamma_{12} + i \delta \rightarrow \gamma_{12}$ have been made in the original equations. Now, $B = \gamma_1 \gamma_2 \Lambda_{22}$. Thus

$$
B = \frac{1}{\frac{1}{s} + \frac{1}{\gamma_1} + \frac{1}{\gamma_2}},\tag{40}
$$

where

$$
s = \frac{\Omega^2}{4} \left(\frac{1}{\gamma_{12}} + \frac{1}{\gamma_{12}^*} \right) \tag{41}
$$

is the saturation rate. For small fields one has $B = s$. We notice that, although the coefficient *B* is the same, the probabilities for absorption, P_{12} , and stimulated emission, P_{21} , differ as a consequence of the different relaxation constants of the levels 1 and 2 and, in a similar way, the rates of these two processes also differ.

B. Three-level atomic system

We consider a three-level atomic system with states $|1\rangle$, $|2\rangle$, and $|3\rangle$ coupled by coherent fields of Rabi frequences Ω_1 , tuned on the transition 1-3, and Ω_2 , tuned on the transition 2-3. The Hamiltonian, in the rotating wave approximation, is given by

$$
H = \begin{pmatrix} \delta_1 & 0 & \frac{\Omega_1}{2} \\ 0 & \delta_2 & \frac{\Omega_2}{2} \\ \frac{\Omega_1}{2} & \frac{\Omega_2}{2} & 0 \end{pmatrix} . \tag{42}
$$

We need to determine the operator Λ_1 . By dropping the index 1, to simplify the notation, we have

$$
\iota \frac{\Omega_1^2}{2} (\Lambda_{13} - \Lambda_{31}) - \gamma_1 \Lambda_{11} = -1,
$$

\n
$$
\iota \frac{\Omega_2^2}{2} (\Lambda_{23} - \Lambda_{32}) - \gamma_2 \Lambda_{22} = 0,
$$

\n
$$
- \gamma_{12} \Lambda_{12} + \frac{\iota}{2} (\Lambda_{13} - \Lambda_{32}) = 0,
$$
\n(43)

$$
-\gamma_{13}\Lambda_{13} + \frac{i}{2}\Omega_2^2\Lambda_{12} - \frac{i}{2}(\Lambda_{33} - \Lambda_{11}) = 0,
$$

$$
-\gamma_{32}\Lambda_{32} - \frac{i}{2}\Omega_1^2\Lambda_{12} + \frac{i}{2}(\Lambda_{33} - \Lambda_{22}) = 0,
$$

$$
\gamma_1\Lambda_{11} + \gamma_2\Lambda_{22} + \gamma_3\Lambda_{33} = 1,
$$

where the following substitutions have been made to obtain the above equations from the original one derived from Eq. (3) and Eq. (7): $\Lambda_{12} \to \Omega_1 \Omega_2 \Lambda_{12}$; $\Lambda_{32} \to \Omega_2 \Lambda_{32}$; Λ_{13}
 $\to \Omega_1 \Lambda_{13}$; $\gamma_{12} + i(\delta_1 - \delta_2) = \gamma_{12} + i\delta \to \gamma_{12}$; $\gamma_{13} + i\delta_1$ $\gamma_{12} + i(\delta_1 - \delta_2) = \gamma_{12} + i\delta \rightarrow \gamma_{12}$; $\rightarrow \gamma_{13}$; $\gamma_{32} - i \delta_2 \rightarrow \gamma_{32}$. We wish now to determine the quantities Λ_{22} and Λ_{33} that are related, through Eq. (29), to the probabilities of the Raman process, connecting states $|1\rangle$ with $|2\rangle$, and to the one-photon process, connecting states $|1\rangle$ and $|3\rangle$. These equations are solved in Appendix A in the most general case. However, as can be seen, the general expressions are rather complicated and of dubious value. Unless a compact expression can be found, the numerical solution is, at present, to be preferred in the most general case. On the contrary, they lead to simple expressions when Ω_1 is very small. Indeed, we find

$$
P_{12} = F_{13} \quad P_{32} \quad R,\tag{44}
$$

$$
P_{13} + P_{12} = F_{13} = \frac{\Omega_1^2}{2\gamma_1} \left(\frac{1}{\Gamma_{13}}\right)',\tag{45}
$$

where the primed quantities denote the real part of the complex quantity within parentheses and

$$
\Gamma_{13} = \gamma_{13} + \frac{\Omega_2^2}{4\gamma_{12}}.
$$
 (46)

The meaning of the factors appearing in the above expressions will be explained below. These expressions are particularly useful to describe the response of a three-level configuration to a probe field tuned on one of its transitions when another field, usually a strong one, is tuned on the remaining one. A problem that has received considerable attention in the context of the search of new efficient lasing configurations in three levels in V or Λ geometry [10].

*1. Analytical features of B*₂₁ *and B*₁₃

The expressions for the factors introduced above are

$$
B_{23} = \gamma_2 P_{23} = \frac{1}{\frac{1}{s} + \frac{1}{\gamma_2} + \frac{1}{\gamma_3}},\tag{47}
$$

$$
F_{13} = \frac{\Omega_1^2}{2} \left(\frac{1}{\Gamma_{13}}\right)' = \frac{\Omega_1^2}{2\gamma_1\gamma_{13}'} A,
$$
 (48)

$$
R = \left\{ 1 + \frac{\gamma_3}{2 \gamma_{12}^*} \frac{\gamma_{32}}{\gamma_{32}'} \frac{\Gamma_{13}}{\Gamma_{13}'} \right\}',\tag{49}
$$

$$
A = \left(\frac{\gamma'_{13}}{\Gamma'_{13}}\right),\tag{50}
$$

where B_{23} is the *B* coefficient for the transition between 2 and 3 driven at the Rabi frequency Ω_2 in absence of the field on transition 1 to 3, or, in any case, when this field is very small. F_{13} is the total probability of absorption, i.e., the sum of a one-photon process probability P_{13} with the Raman absorption process probability P_{12} . For small driving fields Ω_2 , i.e., when P_{12} is negligible, the one-photon absorption *P*¹³ coincides with *F*¹³ . The Raman factor *R* can be further simplified, such as to be written

$$
R = 1 - \frac{\gamma_3}{2\gamma'_{13}} + \frac{\gamma_3}{2\gamma'_{12}} \left\{ \frac{\overline{\gamma}^2}{\overline{\gamma}^2 + \delta^2} \left[1 + \frac{\gamma'_{12}}{\gamma'_{13}} + \frac{\delta_2^2}{\gamma'_{13}\gamma'_{32}} \left(\frac{\gamma'_{12}}{\overline{\gamma}} \right)^2 \right] + \frac{\overline{\gamma}\delta}{\overline{\gamma}^2 + \delta^2} \frac{\gamma'_{12}}{\overline{\gamma}} \frac{\delta_2}{\gamma'_{32}} \left(1 + \frac{\gamma'_{12}}{\gamma'_{13}} - \frac{\gamma'_{32}}{\gamma'_{13}} \right) \right\}
$$
(51)

which results to be the sum of an absorption and a dispersion profile both centered at the Raman detuning and both having width $\bar{\gamma} = \gamma'_{12} [1 + \Omega_2^2 / 4 \gamma'_{12} \gamma'_{13}]^{1/2}$. In the condition in which γ'_{12} is much smaller than γ_3 , a situation which is found in many experimental occurrences related to this configuration, the peak of the Raman factor at $\delta=0$ is much greater than the unity due to the prefactor γ_3 / γ_{12} . Conversely, the background term is, in these same conditions, almost negligible, as is the prefactor of the dispersion profile. We would like to point out here that the factorization of the Raman probability as it occurs in Eq. (44) , although quite appealing, is almost accidental and we are unable, at the moment, to explain it by using simple and intuitive argu-

FIG. 2. This figure shows the *A* factor, continuous line, and Raman factor *R*, dotted line, for different values of the three-level parameters. The Raman factor is normalized to the quantity R_0 $=\gamma_3 / \gamma_{12}^{\prime}$. In the case (a), $\delta_2=0$ and $\Omega_2=1$; for the case (b), δ_2 = 3 and Ω_2 = 1; and for the case (c), δ_2 = 3 and Ω_2 = 3. EIT corresponds here to the minimum of the *A* profile. The fixed parameters are $\gamma_3 = 1$, $\gamma_2 = 0.001$, $\gamma_1 = 0.01$, and $\gamma_{ij} = (\gamma_i + \gamma_j)/2$.

ments. In any case it is already well known that the Raman probability does not coincide with the product of the probabilities of its segment transitions.

In Fig. 2, the quantities *R* and *A* are shown for different values of the configuration's parameters. The factor *A* displays the peculiar doublet structure of the Autler-Townes absorption. In fact, $1/\Gamma_{13}$ possesses two simple poles in the complex plane of the variable δ which can be interpreted as light shifts, or dynamical Stark shift, of the upper dressed states $|2\rangle$ and $|3\rangle$ induced by the strong driving field. The position of the resulting peaks can be identified approximately, when the real part is a slowly varying function of δ , with the vanishing of the imaginary part of Γ_{13} . This latter can be explained as a function of δ in the following way:

$$
\Gamma_{13} = \gamma'_{13} + i \delta_1 + \frac{\Omega_2^2}{4(\gamma'_{12} + i \delta)}
$$

= $\gamma'_{13} + i \delta_1 + \frac{\Omega_2^2}{4(\gamma'_{12} + \delta^2)} (\gamma'_{12} - i \delta)$ (52)

and the condition mentioned above can be expressed in terms of the ratio between the derivative of the real part and imaginary part of the above expression resulting in the inequality $2|\delta |\gamma'_{12}| |\gamma'_{12}^2 - \delta^2| \ll 1$. This condition is satisfied when δ $\gg \gamma_{12}$ and the peaks are located at detunings which obey the relation

$$
\delta_1 \delta \approx \frac{\Omega_2^2}{4}.\tag{53}
$$

For very strong driving fields, this yields $\delta_1 \approx \delta \approx \pm \Omega/2$ whereas the peak maximum value is given by $A \approx \gamma'_{13}/(\gamma'_{13})$ $+\gamma_{12}^{\prime}$. Different kind of information can be gathered by calculating the poles of $1/\Gamma_{13}$. By assuming that γ'_{12} is the smallest time constant in the system such as to be neglected in the above expressions, $\Omega_2 \ll \gamma'_{13}$ and upon defining $\hat{\gamma}$ $= \gamma'_{13} + i \delta_2$, the above poles result to be $\delta_+ = i \hat{\gamma}$ and $\delta_ = i(\Omega_2^2/4 \hat{\gamma})$, respectively. We find

$$
\frac{1}{\Gamma_{13}} = \frac{A_+}{\delta - \delta_+} + \frac{A_-}{\delta - \delta_-} = \frac{1}{\gamma_{13}} - \frac{\Omega_2^2}{4\hat{\gamma}^2} \frac{1}{\frac{\Omega_2^2}{4\hat{\gamma}} + i\delta}.
$$
 (54)

The first term of this expression corresponds to the two-level absorption of the probe field on the 1-3 transition in the absence of the driving field. The second one gives rise to a dip in this profile when the detuning δ_2 is within the onephoton linewidth, and to a well-distinguished peak when δ_2 falls outside of it, as can be shown in Figs. $2(b)$ and $2(c)$. These peaks occurring at the Raman detuning can be interpreted both on the basis of Eqs. (45) and on the presence of a Raman signature in *P*¹³ due Raman resonant multiphoton processes having the same initial and final state of the onephoton process corresponding to P_{13} .

Now, as it concerns the features of the Raman probability P_{12} , Eq. (44) tells us that in order to have the maximum value of this quantity the peaks in the Raman factor and in the *A* factor must match. To this end it is important to observe that the Raman factor *R* is generally peaked at $\delta=0$, i.e., at Raman resonance: indeed, within the approximation made here the term $F_{32}R$ is given by $F_{32}R = [\gamma_2/(\gamma_2$ + γ_3) $\left[\frac{1}{2}\right]$ ($\frac{1}{2}$ /($\frac{1}{2}$ + δ ²) with $\frac{1}{2}$ = $\frac{1}{2}$ /2 $\sqrt{\frac{1}{2}}$ / $\frac{1}{2}$ / $\frac{1}{2}$. On the contrary, the position of the A peaks obeys Eq. (53) according to which greater values of δ_1 correspond to smaller δ values and, by consequence, to better matching. This tells merely that off-resonant Raman probability is greater as compared with the in-resonance probability.

V. APPLICATIONS TO SOME POPULAR PROBLEMS IN QUANTUM OPTICS

We present now some physical situation where the concepts above can be applied. The first one concerns the problem of dark resonances, i.e., the disappearance of fluorescence and absorption that occurs in a three-level Λ atomic system when the detunings satisfy the Raman condition, first reported in $[11]$. This is at the basis of a wealth of phenomena in quantum optics ranging from sub-Doppler laser cooling $\lceil 12 \rceil$ up to the the most recent observation of the anomalous reduction of the speed of light in laser cooled and noncooled media $[13]$. We also briefly show how the electromagnetic induced transparency (EIT) can be interpreted in terms of the multiphoton process probabilities introduced here. Finally, we discuss the issues of efficiency and profitability of some three-level configurations designed to provide efficient gain of radiation by means of an additional driving field, a subject that has close relations with the investigations on lasing configurations operating without population inversion $[3]$.

A. Dark resonances

In a three-level system in the Λ configuration and for values of the detuning corresponding to Raman resonance, the population of the upper state vanishes and the system becomes transparent to the radiation. It is possible to gain some insight into this behavior also in the framework of the rate-balance equations that we have developed here. To carry out our calculations, we assume the presence of a spontaneous emission with rate Γ from the upper state $|3\rangle$ with equal branching ratios to the states $|1\rangle$ and $|2\rangle$ which, in turn, are thermalized at a small rate γ . Thus, the level flow schema is $\Phi_1 = (\Gamma/2)\rho_{33} + \gamma \rho_{22}$, $\Phi_2 = (\Gamma/2)\rho_{33} + \gamma \rho_{11}$, $\Phi_3 = 0$, and the balance-rate equation for the upper state can be written as

$$
-\Gamma \rho_{33} - P_{13} \Phi_1 + P_{23} \Phi_2 = 0. \tag{55}
$$

This, solved for ρ_{33} , gives

$$
\rho_{33} = \frac{\gamma}{\Gamma} \frac{P_{13}\rho_{22} + P_{23}\rho_{11}}{1 - \frac{P_{13} + P_{23}}{2}}.
$$
\n(56)

Now, for very small γ and at Raman resonance where δ_1 $= \delta_2$, it is possible to calculate the absorption probabilities by using the expressions in Appendix B; then we find

$$
P_{13} = \frac{\Omega_1^2}{\Omega_1^2 + \Omega_2^2}, \qquad P_{23} = \frac{\Omega_2^2}{\Omega_1^2 + \Omega_2^2} \tag{57}
$$

and consequently,

$$
\rho_{33} = 2\frac{\gamma}{\Gamma} \frac{\Omega_2^2 \rho_{11} + \Omega_1^2 \rho_{22}}{\Omega_1^2 + \Omega_2^2},
$$
\n(58)

from which it is clear that as $\gamma \rightarrow 0$, $\rho_{33} \rightarrow 0$, thus accounting for the vanishing of the upper state population at Raman resonance $[14]$. Thus, the existence of the dark resonance comes as a result of the quenching of the one-photon absorption probabilies P_{13} and P_{23} , as compared to their value close to the unity when the two fields act separately, see Eq. (40) , or are detuned from Raman resonance. In Appendix B, a useful expression for P_{13} and P_{12} is obtained under the conditions of very small $\gamma_1 = \gamma_2 = \gamma$, equal Rabi frequencies $\Omega_1 = \Omega_2 = \Omega$, $\gamma \ll \Omega^2/\Gamma \ll \Gamma$, and detunings much smaller than Γ , i.e., δ_1 , $\delta_2 \ll \Gamma$. These are given by

FIG. 3. Raman and one-photon *P* probabilities for a three-level system in Λ configuration, with equal Rabi fields, as a function of the detuning $\delta = \delta_1$ with $\delta_2 = 0$. Parameter values are $\Omega_1 = \Omega_2$ $= \Omega = 0.1$ and $\gamma = 10^{-4}$. These are all normalized to the decay rate Γ of the upper state and ρ_{33} is normalized to $(\Omega/\Gamma)^2$.

$$
P_{13} = \frac{1 + \frac{\Omega^2}{\Gamma} \left(\frac{1}{\gamma_{12}}\right)' + \frac{\gamma \Gamma}{\Omega^2}}{1 + 2\frac{\Omega^2}{\Gamma} \left(\frac{1}{\gamma_{12}}\right)' + 2\frac{\gamma \Gamma}{\Omega^2}},
$$
(59)

$$
P_{12} = \frac{\Omega^2}{2\Gamma} \frac{\left(\frac{1}{\gamma_{12}}\right)'}{1 + 2\frac{\Omega^2}{\Gamma} \left(\frac{1}{\gamma_{12}}\right)' + 2\frac{\gamma\Gamma}{\Omega^2}},
$$
(60)

and are shown in Fig. 3. They have Lorentzian shapes whose width is approximately $\delta_{\rm B}=\Omega\sqrt{2\gamma/\Gamma}$. The populations can now be calculated by using Eq. (56) and taking into account that, in the conditions contemplated here, P_{13} and P_{23} are equal. By denoting with *P* their common value, we get

$$
(1 - P)\rho_{33} = \frac{\gamma}{\Gamma} P(\rho_{11} + \rho_{22}), \tag{61}
$$

which, in the limit of small Rabi fields such as $\rho_{11} + \rho_{22}$ \approx 1, leads to

$$
\rho_{33} = \frac{\gamma}{\Gamma} + \left(\frac{\Omega}{\Gamma}\right)^2 \left\{ 1 - \frac{\left(\frac{\Omega^2}{\Gamma}\right)^2}{\delta^2 + \left(\frac{\Omega^2}{\Gamma}\right)^2} \right\}
$$
(62)

with δ being the Raman detuning and $(\Omega/\Gamma)^2$ the ρ_{33} population free from interference. The results obtained here are, of course, very well known in the literature. However, the interesting point of this approach is that of making explicit their connection with the quenching of the absorption probabilities. This can be interpreted as due to the interference of the one-photon process 1-3 with at least a three-photon resonant with the states 1-3-2-3 in sequence.

B. Electromagnetic induced transparency

It is interesting to recognize that, within our framework, the conditions for electromagnetic induced transparency (EIT) | 15 , obtained with increasing driving fields, coincides with a strong reduction of the *B* coefficients with respect to their small fields value. In this situation, no transitions between the prescribed levels takes place and the medium becomes transparent. By inspecting the expressions for P_{21} and *P*₁₃ given above in Eq. (44) and Eq. (45), as $\Omega_2 \rightarrow \infty$, we notice that their vanishing is to be traced back uniquely to the vanishing of the factor *A*. Indeed, in a three-level Λ system, where the depletion of the lower state is very small such as all atoms remains in their lower state ($\rho_{11}=1$), we have $\Phi_1 = \gamma_1$ with negligible stimulated emission and the absorption being equal to $\vec{n} = F_{13} = (\Omega_1^2/2\gamma_{13})A$ $=\Omega_1^2/2(1/\Gamma_{13})'$. To discuss EIT line-shape features $1/\Gamma_{13}$ can be transformed as follows:

$$
\frac{1}{\Gamma_{13}} = \frac{\gamma'_{12} + i\delta}{(\gamma'_{12} + i\delta)(\gamma'_{13} + i\delta + i\delta_2) + \frac{\Omega_2^2}{4}}
$$

$$
= \frac{\gamma'_{12} + i\delta}{\frac{\Omega_2^2}{4}} \left(1 - \frac{(\gamma'_{12} + i\delta)(\gamma'_{13} + i\delta + i\delta_2)}{\frac{\Omega_2^2}{4}} + \cdots \right)
$$
(63)

and its real part written as

$$
\left(\frac{1}{\Gamma_{13}}\right)' = \frac{16}{\Omega_2^4} \left\{ (2\gamma'_{12} + \gamma'_{13}) \delta^2 + 2\gamma'_{12} \delta_2 \delta \right.+ \gamma'_{12} \left(\frac{\Omega_2^2}{4} - \gamma'_{12} \gamma'_{13} \right) + \cdots \right\}
$$
(64)

which can be viewed as a function of δ whose minimum occurs at

$$
\delta_{\min} = -\delta_2 \frac{\gamma'_{12}}{\gamma'_{13} + 2\gamma'_{12}} \tag{65}
$$

away from the Raman resonance. The results above are, of course, subjected to the validity of the expansion of Eq. (63) , which holds as the modulus of the second term within the parenthesis is much smaller than the unity. This condition is, as one can easily see, detuning dependent. As a matter of fact, EIT occurs principally in the frequency region between the two peaks of the F_{13} probability.

C. Profitability of multilevel lasing configurations

Here we review some of the simplest atomic lasing configurations. This analysis has gained momentum in recent years in connection with the lasing without inversion problem $[3,10,16]$, thus reviving some prospects for efficient amplification in the region of short wavelengths. However, some examples in which the absence of population inversion did not warrant the optimal conversion efficiency were subsequently brought about $\lfloor 17 \rfloor$. This was a stimulus for going back to the analysis of these mechanisms by focusing more on the efficiency of the process that transforms atomic excitation into coherent radiation rather than on the absence of population inversion. The point of view of the atomic response to coherent radiation introduced here provides a framework where these issues can be more suitably phrased and assessed. Thus, the question is if there is any advantage in pursuing radiation amplification by means of three-level configurations, where both the one-photon process and the Raman process are simultaneously active, instead of through the more simple and conventional two-level system. Before entering into the details of the more complicated three-level systems in Λ and V configuration, we review some concepts relative to a two-level system within the framework of ideas developed here.

1. Lasing efficiency in a two-level system

The gain in a two-level system, with 1 as lower level and 2 as upper one, can be derived from Eq. (36) by assuming, quite reasonably, $\Phi_1 = \gamma_2 \rho_{22}$ and $\Phi_2 = \gamma_1 \rho_{11}$. In the condition of negligible fields, i.e., when *s* is the smallest rate in the system, we obtain $\Phi_1 = \Phi_2$, $B = s$. If, in addition, $\gamma_1 \ll \gamma_2$, we have $\rho_{11} \approx 1$, and $n = -s$, which is related to the natural absorption cross section $\sigma=3\lambda^2/2\pi$. On the contrary, if $\gamma_2 \ll \gamma_1$ we obtain $n = s$ which represents the upper limit for the gain. By exactly solving Eq. (36) , one instead gets *n* $= (\gamma_1 - \gamma_2)s/(\gamma_1 + \gamma_2 + 2s).$

Now, as far as lasing efficiency is concerned, a lasing system presents some similarities to a thermal engine where microscopic disordered energy is transformed into macroscopic energy. Quite similarly to the thermal engine, where not all the work done by the highest temperature source is available work, here not all the photons emitted in the stimulated emission process are useful photons because a fraction of them is absorbed back in the medium. Thus, we can define the atomic conversion efficiency as $\eta_a = \frac{n}{P_2} \Phi_2$ which, as from Eq. (36), can be written as $\eta_a = 1 - (t_1 / t_2)$, where the quantities $t_i = \Phi_i / \gamma_i$ play the same role of the temperature in a thermal engine. Another important indicator of the lasing efficiency is the pump conversion efficiency η_p . It accounts for the fraction of energy provided by the pumping process that is transformed into stimulated emission radiation. The overall conversion efficiency is given, now, by the product $\eta = \eta_a \eta_p$. In the ideal case, which occurs only when all the pumped atoms are engaged in the pumping cycle through the upper level, this ratio becomes $\eta_p = P_{21}\Phi_2 / \Phi_2 = P_{21}$, i.e., it does coincide with the stimulated emission probability from the upper level. The pumping efficiency is a decreasing function of the pumping rate and for small intensities, in resonance and at threshold, i.e., where $\gamma_1 = \gamma_2 = \Gamma$, it is given by $P_{21} = s/\gamma_2 = s/\Gamma = \Omega^2/2\Gamma^2$. In the case of a very strong spontaneous emission Γ , such conversion efficiency becomes very low. This is, indeed, in relation to the well known limitations connected to gain at small wavelengths, although it is not often pointed out that in these conditions the excess of energy above the threshold is mostly dissipated and only a small fraction of it is converted into coherent radiation. It is trivial to see that $P_{21} \propto 1/|\rho|^2$, where $|\rho|$ is the electric dipole moment of the transition, whereas the stimulated emission rate at threshold, given by $\Omega^2/4\Gamma$, is indepen-

FIG. 4. Three-level system in Λ configuration, case (a) and V configuration, case (b).

dent of this quantity. The conclusion is that the decreasing of the transition dipole moment increases the efficiency and soften the threshold requirement by leaving unaltered the stimulated emission rate at threshold. The above considerations suggest that a metastable state, weakly coupled with an interacting state in such a way as to give rise to a stationary state with a small electric dipole moment, will present a favorable condition of small threshold and high pumping efficiency even within the two-level configuration scheme. There is nothing new in this, but mentioning it here will help one to understand the true scope of the improvement, if any, obtained with the more complicated configurations, which will be analyzed in the following.

2. Three-level Λ *configuration*

We consider a Λ atomic schema with a lower state $|1\rangle$ and two upper states, $|2\rangle$ and $|3\rangle$, closely spaced as shown in Fig. $4(a)$. The probe field acts on the transition 1-3 whereas the driving field operates on the transition 2-3. In order emphasize that we discuss a case where the upper level 3 is a strongly decaying one, we indicate its decay rate by Γ instead of γ_3 . The performance of this system largely depends on the relaxation mechanisms adopted. For instance, with the schema $\Phi_3 = \gamma_1 \rho_{11}$, $\Phi_2 = \Gamma \rho_{33}$, and $\Phi_1 = 0$, absorption does not take place because no atoms start their evolution in the state $|1\rangle$, $\Phi_1=0$, and the top performance can be obtained. Needless to say, such performance would never come about in practice because the relative relaxation schema is physically unfeasible. It is also possible to imagine that the upper level 3 decays to 1 and 2, with branching ratios π_1 $\approx \pi_2$, respectively, as has been done in [5]. This configuration also provides a very good performance. However, considering that the two upper levels are closely spaced, it can be realistic only when the transition 1-3 is a very weak one. If this is the case, i.e., if a slowly decaying and nearly metastable upper lasing level is available, is there any advantage in using this configuration instead of a two-level configuration operating on the weakly coupled transition 1-3 that could instead benefit from the improved performance as outlined in the preceding section? We will try to answer to this question in this subsection. However, in the meantime, we want to start our discussion by dealing with a strong probe transition 1-3 that corresponds to short wavelengths. Since it is expected that the Raman process will be the dominant one for the gain, we could take the most advantage of it by adopting a direct pumping mechanism from level 1 to level 2 with no decay from level 3 to 2 being allowed. Thus, we have

FIG. 5. One-photon absorption, P_{13} , and Raman stimulated gain, P_{21} , normalized to the standard two level absorption probability Ω_1^2/Γ^2 . We have set $\Gamma = 1.0$, $\gamma_2 = 10^{-4}$, $\gamma_1 = 0.01$, δ_2 = 5.0, Ω_1 = 10⁻³, γ_{ii} = (γ_i + γ_i)/2. In the inset (a) the driving field is Ω_2 =0.05, while in the inset (b) Ω_2 =0.6. The emerging of *F*₁₃ absorption peak, as Ω_2 is increased, can be noted.

$$
\Phi_1 = \Gamma \rho_{33} + \gamma_2 \rho_{22}, \quad \Phi_2 = \gamma_1 \rho_{11}, \quad \Phi_3 = 0.
$$
 (66)

In the condition in which the probe field is negligible, the balance equations are

$$
-\gamma_1 \rho_{11} + \Phi_1 = 0,
$$

$$
-\gamma_2 \rho_{22} + \Phi_2 - P_{23} \Phi_2 = 0,
$$
 (67)

$$
-\Gamma \rho_{33} + P_{23} \Phi_2 = 0,
$$

which, together with Eqs. (66), allow one to prove that Φ_1 $=$ Φ_2 . Using the condition $\rho_{11} + \rho_{22} + \rho_{33} = 1$ one gets

$$
\Phi_2 = \Phi_1 = \frac{1}{\frac{1}{\gamma_1} + \frac{1}{\gamma_2} + P_{23} \left(\frac{1}{\Gamma} - \frac{1}{\gamma_2} \right)}.
$$
(68)

The total gain is then given by

$$
\dot{n}_{31} = \mu_{21} + \mu_{13} + \mu_{12}
$$
\n
$$
= B_{21} \left(\frac{\Phi_2}{\gamma_2} - \frac{\Phi_1}{\gamma_1} \right) - B_{13} \frac{\Phi_1}{\gamma_1}
$$
\n
$$
= \Phi_2 \left\{ B_{21} \left(\frac{1}{\gamma_2} - \frac{1}{\gamma_1} \right) - B_{13} \frac{1}{\gamma_1} \right\}
$$
\n
$$
= (P_{21} - F_{13}) \Phi_2.
$$
\n(69)

It is now clear that the threshold requires that $\gamma_1 \geq \gamma_2$. Thus, only if $\gamma_2 \ll \Gamma$ we can expect a consistent lowering of the lasing threshold, thus making an almost metastable state a good candidate to suit this purpose. Beams of metastable He atoms and pulsed pump field has been experimentally investigated in $[18]$ to provide vuv amplification. However, because of the destabilization of the mestastable state due to the collisions, its realization might not be simple in bulk gaseous matter.

In Fig. 5, the quantities P_{21} and F_{13} , which serve as a basis for the calculation of gain in the Λ configuration discussed here, are shown. In order to obtain the maximum

Raman probability detuned fields must be employed because they allow the best matching between the factors *A* and *R*. Both the driving and probe field tuned on the respective transitions represents, in this respect, the worst matching condition, as it can be gathered from Fig. 2. For the parameters in inset (a) the total absorption F_{13} is negligible and the gain is totally due to Raman stimulated emission. The system thus operates in a very efficient way because absorption is nearly eliminated, but, as noticed above, this depends, on the basis of Eq. (69), solely from the smallness of the γ_2 decay. However, as the gain is concerned, this is still small because Φ_2 is also small. This happens because values of Ω_2 , such as those in inset (a) , do not produce an efficient mixing between states $|2\rangle$ and $|3\rangle$ such that the decay to the lower level is dominated by the rate γ_2 . In fact, most of the population is in state $|2\rangle$ and the rates $\Phi_2 = \Phi_1$ are approximately equal to γ_2 , i.e., to a very small quantity. As a matter of fact, we find $n_{31} = P_{21} \Phi_2 \approx 71. \times 0.123 \times 10^{-3} = 0.008$, in units of the natural absorption, by reading the value of P_{21} from inset (a) in Fig. 5 . In the case of inset (b) the driving field is increased such as to allow an efficient mixing between the two upper levels. This increases the rates $\Phi_1 = \Phi_2$ because of the fast decay back to state $|1\rangle$ that occurs from state $|3\rangle$. Thus, most of the atoms are found in the lower state and the level rates become approximately equal to γ_1 . For $\Omega_2=0.6$ we obtain $\Phi_2 = 0.267 \times 10^{-2}$ and, by using the value of $P_{21} - F_{13}$ \approx 48, gathered from inset (b) at the detuning where this difference is the greatest one, we calculate the overall gain to be approximately n_{31} =0.12 times the natural absorption. This is pedagogical in showing that having most of the atoms in the lower state helps improve the gain via an increase of the upper level rate, a mechanism that is active in most of the schemes for lasing without inversion $[3,5,10]$. Unfortunately the driving field cannot be increased indefinitely because a drastic diminishing of the Raman stimulated emission probability, caused by a mismatch between *A* and *R*, corresponds only to a modest increase of Φ_2 which, on the other hand, presents a saturating behavior as a function of Ω_2 . A comparison with a two level system on the transition 1-3 is now possible. To obtain the same gain with a two level system operating between level 1 and 3 one would need, according to the expression $(\gamma-\Gamma)/(\gamma+\Gamma)$ of the normalized gain, a pumping rate $\gamma=1.25$ instead of $\gamma_1=0.01$, which is that used in inset (b). Thus, an improvement in the threshold of a factor of about 100 times has been obtained.

However, in spite of this, it appears evident that this system operates less efficiently than the individual Raman process because the one-photon process adds extra absorption. Moreover, one might ask whether a one-photon two-level process between the same initial and final states 1 and 2 could provide an even better performance of the Raman process alone. Indeed, by performing this comparison one must notice that the atomic efficiency of both processes is the same, i.e., $\eta_a = 1 - (\gamma_2/\gamma_1)$. On the contrary, the pump efficiency of the Raman process will be given by

$$
P_{21} = \frac{\gamma_1}{\gamma_2} P_{12} = \frac{\gamma_1}{\gamma_2} F_{13} P_{32} R \le \frac{\gamma_1}{\gamma_2} \frac{\Omega_1^2}{2 \gamma_1} \left(\frac{1}{\Gamma_{13}}\right)' \frac{\gamma_2}{\gamma_3} \frac{\gamma_3}{\gamma_1} = \frac{\Omega_1^2}{\Gamma \gamma_1}.
$$
\n(70)

Here we have used the fact that $F_{32} \leq \gamma_2 / \gamma_3$ and $R \leq \gamma_3 / \gamma_1$ for small γ_2 . We also recall that the pump efficiency of a two-level system operating on 1-2 transition is P_{21}^0 $= \Omega_{12}^2/\gamma_1\gamma_2$. In addition, because levels 2 and 3 are closely spaced, the relation $\Omega_{12}/\gamma_2 \approx \Omega_1/\Gamma$ holds. Thus, inserting this in Eq. (70), we obtain $P_{21}^0 \ge P_{21}$ [19]. To complete the comparison we notice that only for strong driving fields can the rate Φ_2 be as high as the equivalent one in the two-level system between levels 1 and 2. Thus, considering the difficulties intrinsic in the experimental implementation of the multilevel lasing configurations, as enlisted in the recent review work of Mompart and Corbalan $[20]$, the simple twolevel configuration results, between the two, in a better overall performance.

3. Three-level V configuration

We consider 1 to be the upper lasing level, whereas 2 and 3 are a couple of closely spaced lower levels, 1-3 is the probe transition, and 2-3 the driver transition, all as shown in Fig. $4(b)$. The flow schema

$$
\Phi_1 = \gamma_2 \rho_{22} + \gamma_3 \rho_{33}, \quad \Phi_2 = 0, \quad \Phi_3 = \Gamma \rho_{11}, \quad (71)
$$

with $\Phi_2=0$, implies that no Raman absorption takes place in spite of stimulated Raman emission being active: it can, therefore, be considered as an optimal one for the purpose of obtaining gain. We also assume that pumping to the upper state takes place both from level 2 and level 3. The rate equations in the limit of small probe field are

$$
-\Gamma \rho_{11} + \Phi_1 = 0,
$$

$$
-\gamma_2 \rho_{22} + P_{32} \Phi_3 = 0,
$$
 (72)

$$
-\gamma_3 \rho_{33} + \Phi_3 - P_{32} \Phi_3 = 0,
$$

from which, after trivial manipulations, the following results:

$$
\Phi_3 = \Phi_1 = \frac{1}{\frac{1}{\Gamma} + \frac{1}{\gamma_3} + P_{32} \left(\frac{1}{\gamma_2} - \frac{1}{\gamma_3} \right)}.
$$
(73)

Thus, the gain of the probe field can be written as

$$
n_{31} = \Phi_1 P_{13} + \Phi_1 P_{12} - \Phi_3 P_{31} = \Phi_1 (F_{13} - P_{31}).
$$
 (74)

Now, this configuration, contrary to the Λ configuration, has a better performance over the single one-photon process between 1 and 3 because it can benefit from the additional gain due to the Raman process. Thus making the Raman stimulated emission as big as possible would provide the maximum benefit in terms of lasing gain. A similar effect can be induced, on the basis of Eq. (21), by increasing γ_2 .

We now investigate both the threshold condition and the magnitude of the gain obtained with this system. We thus consider the ratio

$$
r_{\rm th} = \frac{F_{13}}{P_{31}} = \frac{P_{12} + P_{13}}{P_{31}} = \frac{\gamma_3}{\gamma_1} \left(\frac{1}{1 - F_{32}R} \right) \tag{75}
$$

FIG. 6. Maximum gain of the V configuration in the space of the parameters δ_1 , δ_2 , and Ω_2 as a function of γ_2 with $\gamma_3=10^{-4}$, continuous line, and $\gamma_3=0.01$, dotted line. The inset (a) shows the gain peak corresponding to $\gamma_2=0.5$. In this case $\Omega_2=67.25$, δ_2 =41.92, and the maximum occurs at the detuning δ_1 =60.6. It is possible to verify that these values satisfy the relation $\delta_1(\delta_1-\delta_2)$ $\approx (\Omega_2/2)^2$. The inset (b) shows the values of Ω_2 and δ_2 corresponding to the maximum gain as a function of γ_2 . We have set $\Gamma = 1.0$ and $\gamma_3 = 10^{-4}$ in both (a) and (b).

showing that the most favorable condition occurs for detuning close to the Raman resonance where *R* is peaked. This ratio becomes, in the limit of strong driving fields, equal to $r_{\text{th}}=[(\gamma_2+\gamma_3)/\Gamma][(\Gamma+\gamma_2)/\Gamma]$, how it can be gathered from Eqs. (49) and (47) , indicating that gain in this condition can be obtained only if the lower state relaxations are close to the upper state relaxation Γ , a result that is not surprising. However, we point out that for reasonable relaxation parameters this condition is fulfilled for Ω_2 ranging around rather prohibitive values of about some hundreds of Γ . Indeed we have numerically verified that the gain condition can be met even by decreasing γ_2 and γ_3 to very small values, given that Ω_2 and the detunings δ_2 are further increased. Thus we can conjecture that, no matter how small the decay rates of the lower levels are, it is always possible, by increasing the driving field intensity and detuning, to make $r_{th} > 1$. However, if we look at the probe gain profile near the Raman condition, this appears flat and, in particular, undistinguishable from zero. In other words, such gain takes place at frequencies where the medium is already transparent and absorption can hardly be distinguished from gain. This behavior is related to the shape of F_{13} profile as discussed in Sec. IV B 1. We recall here that F_{13} is almost zero everywhere except for two peaks located at detunings related by $\delta_1(\delta_1 - \delta_2) \approx \Omega_2^2/4$. Thus, when Ω_2 is strong the above detunings depart from the Raman condition and the overall stimulated emission, given by F_{13} , together with absorption *P*³¹ , becomes very small.

However, for more reasonable values of Ω_2 , but still not too small, the location of the *A* peak can be made to fall within the Raman detuning range that, in turns, widens as Ω_2 increases. Here, as from our initial assumption of strong decay of the upper state, we assume a very small decay γ_3 of the lower state ($\gamma_3 \ll \gamma_1 = \Gamma = 1$). In this way a competition between the Raman stimulated emission and the one-photon absorption takes place. We then calculate numerically the maximum gain by means of a gradient algorithm that operates the space of the parameters δ_1 , δ_2 , and Ω_2 . The results, plotted in Fig. 6 as a function of γ_2 , is, in this way, independent of all the system parameters and has a universal character. We notice that, for values of γ_2 approximately below $0.1 \times \Gamma$, the gain obtained can be considered rather negligible. Indeed, by identifying the effective threshold as the intercept on the coordinate axis of the straight line tangent to the upper rectilinear portion of this curve we obtain a threshold value for $\gamma_2 \approx 0.3 \times \Gamma$. In the inset (a) of Fig. 6 it is shown how the gain profile looks like as a function of the probe detuning. The gain reported here as a function of γ_2 corresponds to the tracking of this peak in the parameter space of the V configuration. Inset (b) of this same figure shows how the driving field parameters Ω_2 and δ_2 must be changed in order to fulfill the maximum gain condition.

The conclusion is that the condition for obtaining gain in this V configuration is just a little better than that corresponding to the gain in a simple two-level atomic system, but does not differ too much from it. However, it must be pointed out that all this occurs in the presence of a rather artificial pumping schema that trades off its physical feasibility for efficiency. On the other hand, the other gain regime also reported here, which can be obtained with much lower decay rates of the lower states, needs extremely high and, almost certainly, unfeasible values of the driving field and are so small as to be practically indistinguishable from the condition of transparency that the bare medium displays.

4. Multilevel configurations

Considering the disappointing results obtained for the three-level systems, in terms of efficiency and physical feasibility of the gain process, we would be tempted to extend such findings also to the case of a more general multilevel configuration. However, no rigorous proof that excludes possible favorable cases can be given here. All we can do is only a very approximate reasoning. We simply notice that our equations deal with the level rates: these are essentially flows or currents which obey conservation laws as in the case of the currents in an electric circuit. In turn, the presence of strong driving fields modifies the prexisting branching geometry, as can be gathered from Eq. (33) . However, by considering a group of upper levels and one of lower ones, with driving fields connecting only levels in the same group, it remains substantially true that the current rising to a set of upper levels comes also back down. A more delicate question concerns how the flows are branched within the group of upper and lower levels. If there are no strong imbalances and the flows to the levels are roughly the same, only small decay rates of the upper levels will give a chance for the quantities $t_i = \Phi_i / \gamma_i$ to be inverted in order to provide gain. Thus, the simple rule of thumb at the basis of the conventional laser operations of the two-level system turns out to hold, probably also for the operations of more complicated multilevel systems.

VI. CONCLUSIONS

In this paper we have studied the symmetries in the optical Bloch equations which are associated with the absorption and stimulated emission processes. This has allowed us to establish a set of balance-rate equations which are useful for describing the response of an atomic system to coherent radiations and, more importantly, to recover the symmetries between absorption and stimulated emission known to hold for a two-level system. In particular, it is found that for any single multiphoton process the gain is depends on the difference of the quantities $t_i = \Phi_i / \gamma_i$ between the initial and final levels of the process. Physically meaningful parameters, such as the generalized Einstein *B* coefficients or the probabilities of multiphoton processes P_{ij} , appear directly in these equations, thus making the analysis of the atomic response conceptually more simple and physically intuitive. In fact, here the coherence-related effects that have played such a central role in the investigation of some configurations operating without population inversion, are already included in the calculation of the the *B* coefficients and have, therefore, the same effects on both absorption and stimulated emission processes. What instead determines the emergence of the lasing condition is to be searched for in the rate processes governed by the equations that we have introduced here. Important issues, such as the efficiency of a given lasing configuration, are thus more easily tackled. Regarding the two applications of the concepts presented here, the analysis of the dark resonance reveals, quite explicitly, through the use of the ratebalance equations, how it can be considered the result of the quenching of the one-photon absorption probabilities due to the interference between virtual multiphoton processes connecting the lower to the upper state. In the other application, the analysis of the Λ configuration reported here shows a dramatic increase of the efficiency over the standard twolevel configuration. However, this is not really a substantial improvement since it can be shown to depend entirely on the presence of a slowly decaying upper state present in the configuration. Thus, an even better performance can be obtained more directly by using this last state as upper state of a two-level configuration. A similar analysis, carried out for the V configuration, reveals that the threshold condition can be made only slightly better than that corresponding to the two-level configuration but not yet enough to represent a substantial improvement toward devising feasible lasing operations at short wavelengths. This points to the conclusion that, perhaps, the conventional concepts at the basis of the two-level lasing operations represent a more viable alternative at these wavelengths. A possibility of this kind could be given by a two-level system which operates on a weakly coupled transition obtained, for example, by a Stark coupling of a metastable state. However, this may not be easy in bulk gaseous matter due to the collisions that make these states highly unstable. On the contrary, a collection of cold metastable atoms, such as those obtained nowadays in several laboratories where cooling techniques are successfully applied, could be more suited to this purpose.

APPENDIX A: SOLUTION OF OBEs IN A THREE-LEVEL SYSTEM

We report here the equations relative to a three-level system as they were written down in Sec. IV B,

$$
\begin{aligned}\n\Omega_1^2 & (\Lambda_{13} - \Lambda_{31}) - \gamma_1 \Lambda_{11} = -1, \\
\frac{\Omega_2^2}{2} & (\Lambda_{23} - \Lambda_{32}) - \gamma_2 \Lambda_{22} = 0, \\
& -\gamma_{12} \Lambda_{12} + \frac{l}{2} (\Lambda_{13} - \Lambda_{32}) = 0, \\
& -\gamma_{13} \Lambda_{13} + \frac{l}{2} \Omega_2^2 \Lambda_{12} - \frac{l}{2} (\Lambda_{33} - \Lambda_{11}) = 0, \\
& -\gamma_{32} \Lambda_{32} - \frac{l}{2} \Omega_1^2 \Lambda_{12} + \frac{l}{2} (\Lambda_{33} - \Lambda_{22}) = 0, \\
& \gamma_1 \Lambda_{11} + \gamma_2 \Lambda_{22} + \gamma_3 \Lambda_{33} = 1.\n\end{aligned}
$$
\n(A1)

These can be solved by first eliminating the coherences which can be expressed in terms of the diagonal elements in the equations above,

$$
\Lambda_{13} = \frac{i}{2\Delta} \left\{ \left(\gamma_{32} + \frac{\Omega_1^2}{4\gamma_{12}} \right) (\Lambda_{11} - \Lambda_{33}) + \frac{\Omega_2^2}{4\gamma_{12}} (\Lambda_{33} - \Lambda_{22}) \right\},\tag{A2}
$$

$$
\Lambda_{32} = \frac{i}{2\Delta} \left\{ \frac{\Omega_1^2}{4\gamma_{12}} (\Lambda_{11} - \Lambda_{33}) + \left(\gamma_{13} + \frac{\Omega_2^2}{4\gamma_{12}} \right) (\Lambda_{33} - \Lambda_{22}) \right\},\
$$

where

$$
\Delta = \gamma_{13}\gamma_{32} + \frac{\Omega_1^2}{4\gamma_{12}}\gamma_{13} + \frac{\Omega_2^2}{4\gamma_{12}}\gamma_{32}.
$$
 (A3)

By inserting these equations in the first two, we obtain the following equations:

$$
-\frac{\Omega_1^2}{\gamma_1} \left\{ B + \Omega_2^2 \right\} C \left\} P_{11} + \left\{ 1 + \frac{(\Omega_1 \Omega_2)^2}{\gamma_2} \right\} P_{12} + \left\{ 1 + \frac{\Omega_1^2}{\gamma_3} [B + C(\Omega_1^2 - \Omega_2^2)] \right\} P_{13} = 0,
$$
\n
$$
\frac{(\Omega_1 \Omega_2)^2}{\gamma_1} P_{11} - \left\{ 1 + \frac{\Omega_2^2}{\gamma_2} (A + C\Omega_2^2) \right\} P_{12}
$$
\n
$$
+ \frac{\Omega_2^2}{\gamma_3} \left\{ A + C(\Omega_2^2 - \Omega_1^2) \right\} P_{13} = 0,
$$
\n(A4)

in which the following quantities have been defined:

$$
A = \frac{1}{2} \left(\frac{\gamma_{13}}{\Delta} \right)' = \frac{1}{2|\Delta|^2} (\gamma_{13}^* \Delta)',
$$

\n
$$
B = \frac{1}{2} \left(\frac{\gamma_{32}}{\Delta} \right)' = \frac{1}{2|\Delta|^2} (\gamma_{32}^* \Delta)',
$$

\n
$$
C = \frac{1}{2} \left(\frac{1}{4 \gamma_{12} \Delta} \right)' = \frac{1}{8|\Delta|^2} \left(\frac{\Delta}{\gamma_{12}^*} \right)',
$$

\n(A5)

In solving the above equations one could follow the suggestions of $[21]$. One thus finds

$$
P_{11} = \left\{ 1 + \Omega_2^2 \left(\frac{1}{\gamma_2} + \frac{1}{\gamma_3} \right) A + \frac{\Omega_1^2}{\gamma_3} B + \left[\frac{1}{\gamma_3} (\Omega_1^2 - \Omega_2^2)^2 + \frac{1}{\gamma_2} \Omega_2^4 \right] C + \frac{(\Omega_1 \Omega_2)^2}{\gamma_2 \gamma_3} P \right\} / \Sigma,
$$

\n
$$
P_{12} = \frac{(\Omega_1 \Omega_2)^2}{\gamma_1} \left\{ \frac{P}{\gamma_3} + C \right\} / \Sigma,
$$

\n
$$
P_{13} = \frac{\Omega_1^2}{\gamma_1} \left\{ \frac{\Omega_2^2}{\gamma_2} P + A + C(\Omega_1^2 - \Omega_2^2) \right\} / \Sigma, \quad (A6)
$$

with

$$
P = AB + \Omega_1^2 AC + \Omega_2^2 BC \tag{A7}
$$

and

$$
\Sigma = 1 + \Omega_2^2 \left(\frac{1}{\gamma_2} + \frac{1}{\gamma_3} \right) (A + C\Omega_2^2)
$$

+
$$
\Omega_1^2 \left(\frac{1}{\gamma_1} + \frac{1}{\gamma_3} \right) (B + C\Omega_1^2) + (\Omega_1 \Omega_2)^2 \left\{ \left(\frac{1}{\gamma_1 \gamma_2} + \frac{1}{\gamma_1 \gamma_3} + \frac{1}{\gamma_2 \gamma_3} \right) P - 2\frac{C}{\gamma_3} \right\}.
$$
 (A8)

In evaluating the factor *P*, one can keep in mind the following equality:

$$
\sum_{i} \text{ Re}\left(\frac{a_{i}}{\sum a_{s}b_{s}}\right) \text{Re}\left(\frac{b_{i}}{\sum a_{s}b_{s}}\right) = \sum_{i} \frac{\text{Re}(a_{i})\text{Re}(b_{i})}{|\sum a_{s}b_{s}|^{2}}.
$$
\n(A9)

It is not difficult now, to provide an expression for P_{12} and P_{13} in the condition of small Ω_1 . In the case of Σ the last two terms vanish and $A + C \Omega_2^2 = \frac{1}{2} (1/\gamma_{32})'$. Thus

$$
\Sigma = 1 + s \quad \left(\frac{1}{\gamma_2} + \frac{1}{\gamma_3}\right),\tag{A10}
$$

where $s = \Omega_2^2/2(1/\gamma_{32})'$ is the saturation rate in the transition 2-3. Now, P_{12} is found to be

$$
P_{12} = \frac{\Omega_1^2 \Omega_2^2}{4 \gamma_1 \gamma_3} \left\{ \frac{\left(\frac{1}{\gamma_{32}}\right)' + \frac{\gamma_3}{2} \frac{1}{\gamma_{12} \gamma_{32}}}{\Gamma_{13}} \right\} \frac{1}{\Sigma} \tag{A11}
$$

whereas P_{13} is given by

$$
P_{13} = \frac{\Omega_1^2}{2\gamma_1} \left(\frac{1}{\Gamma_{13}}\right)' - P_{12}.
$$
 (A12)

By further manipulating the above expression, one gets

$$
P_{12} = F_{13} \t F_{32} \left\{ 1 + \frac{\gamma_3}{2 \gamma_{12}^*} \frac{\gamma_{32}}{\gamma_{32}'} \frac{\Gamma_{13}}{\Gamma_{13}'} \right\}', \quad \text{(A13)}
$$

where $F_{32} = P_{32} = s / \gamma_3 \Sigma$ coincides with the probability of the transition from 3 to 2 while F_{13} is given by

$$
F_{13} = \frac{\Omega_1^2}{2\,\gamma_1} \left(\frac{1}{\Gamma_{13}}\right)'
$$
 (A14)

with

$$
\Gamma_{13} = \gamma_{13} + \frac{\Omega_2^2}{4 \gamma_{12}}.
$$
 (A15)

The last factor in Eq. $(A13)$ can be further transformed to yield

$$
R = 1 - \frac{\gamma_3}{2\gamma'_{13}} + \frac{\gamma_3}{2\gamma'_{12}} \left\{ \frac{\hat{\gamma}^2}{\hat{\gamma}^2 + \delta^2} \right\} 1 + \frac{\gamma'_{12}}{\gamma'_{13}} + \frac{\delta_2^2}{\gamma'_{13}\gamma'_{32}} \left(\frac{\gamma'_{12}}{\hat{\gamma}} \right)^2 \right\} + \frac{\hat{\gamma}\delta}{\hat{\gamma}^2 + \delta^2} \frac{\gamma'_{12}}{\hat{\gamma}} \frac{\delta_2}{\hat{\gamma}} \left(1 + \frac{\gamma'_{12}}{\gamma'_{13}} \right) - \frac{\gamma'_{32}}{\gamma'_{13}} \left\{ 1 + \frac{\gamma'_{32}}{\gamma'_{13}} \right\} \tag{A16}
$$

with

$$
\hat{\gamma} = \gamma'_{12} \left(1 + \frac{\Omega_2^2}{4 \gamma'_{12} \gamma'_{13}} \right)^{1/2}.
$$
 (A17)

APPENDIX B: THREE-LEVEL SYSTEM IN A CONFIGURATION AND DARK RESONANCE

In the case in which the upper state decays with a spontaneous emission rate Γ_1 to the lower state $|1\rangle$ and with a rate Γ_2 to the lower state $|2\rangle$ and the lower levels have decays $\gamma_1 = \gamma_2 = \gamma$, the transverse relaxation rates are

$$
\gamma_{13} = \frac{\Gamma}{2} + i \delta_1, \qquad \gamma_{32} = \frac{\Gamma}{2} - i \delta_2,
$$

$$
\gamma_{12} = \gamma + i(\delta_1 - \delta_2) = \gamma + i \delta
$$
 (B1)

with $\Gamma = \Gamma_1 + \Gamma_2 + \gamma$. In this case the quantities *A*, *B*, *C*, and *P* are given by

$$
A = \frac{1}{2|\Delta|^2} \left\{ \left[\left(\frac{\Gamma}{2} \right)^2 + \delta_1^2 \right] \left[\frac{\Gamma}{2} + \frac{\Omega_1^2}{4} \left(\frac{1}{\gamma_{12}} \right)' \right] + \frac{\Omega_2^2}{4} \left(\frac{1}{\gamma_{12}} \right)' \left[\left(\frac{\Gamma}{2} \right)^2 - \delta_1 \delta_2 - \frac{\delta(\delta_1 + \delta_2)}{\gamma} \frac{\Gamma}{2} \right] \right\},
$$
\n(B2)

$$
B = \frac{1}{2|\Delta|^2} \left\{ \left[\left(\frac{\Gamma}{2} \right)^2 + \delta_2^2 \right] \left[\frac{\Gamma}{2} + \frac{\Omega_2^2}{4} \left(\frac{1}{\gamma_{12}} \right)' \right] + \frac{\Omega_1^2}{4} \left(\frac{1}{\gamma_{12}} \right)' \left[\left(\frac{\Gamma}{2} \right)^2 - \delta_1 \delta_2 + \frac{\delta(\delta_1 + \delta_2)}{\gamma} \frac{\Gamma}{2} \right] \right\},
$$
(B3)

$$
C = \frac{1}{8|\Delta|^2} \left(\frac{1}{\gamma_{12}}\right)' \left\{ \left(\frac{\Gamma}{2}\right)^2 + \delta_1 \delta_2 + \frac{\Gamma}{2\gamma} \left(\frac{\Omega_1^2 + \Omega_2^2}{4} - \delta^2\right) \right\},\tag{B4}
$$

$$
P = \frac{1}{4|\Delta|^2} \left\{ \left(\frac{\Gamma}{2}\right)^2 + \frac{\Omega_1^2 + \Omega_2^2}{4} \left(\frac{1}{\gamma_{12}}\right)^T \frac{\Gamma}{2} \right\}.
$$
 (B5)

In the condition of $\Omega_1 = \Omega_2 = \Omega$ and $\delta_2 = 0$ one finds

$$
A = \frac{1}{2|\Delta|^2} \left(\frac{\Gamma}{2}\right)^3 \left\{ 1 + \frac{\Omega^2}{2\Gamma} \left(\frac{1}{\gamma_{12}}\right)' \left(2 + \frac{4\delta^2}{\Gamma_2} - \frac{2\delta^2}{\gamma \Gamma} \right) + \frac{4\delta^2}{\Gamma^2} \right\},
$$

\n
$$
B = \frac{1}{2|\Delta|^2} \left(\frac{\Gamma}{2}\right)^3 \left\{ 1 + \frac{\Omega^2}{2\Gamma} \left(\frac{1}{\gamma_{12}}\right)' \left(2 + \frac{2\delta^2}{\gamma \Gamma} \right) \right\},
$$

\n
$$
C = \frac{1}{8|\Delta|^2} \left(\frac{\Gamma}{2}\right)^2 \left(\frac{1}{\gamma_{12}}\right)' \frac{1}{\gamma} \left\{ \gamma + \left(\frac{\Omega^2}{\Gamma} - \frac{2\delta^2}{\Gamma} \right) \right\},
$$

\n
$$
P = \frac{1}{4|\Delta|^2} \left(\frac{\Gamma}{2}\right)^2 \left\{ 1 + \frac{\Omega^2}{\Gamma} \left(\frac{1}{\gamma_{12}}\right)' \right\}.
$$
 (B6)

Now, in order to calculate the B_{13} through Eq. (A6) in the conditions of $\gamma \rightarrow 0$, we must calculate *A*, *B*, and *C* up to the zero order in γ and *P* to the first order in γ . By doing so we get

$$
A = \frac{1}{2|\Delta|^2} \left(\frac{\Gamma}{2}\right)^3 \left(1 + 4\frac{\delta^2}{\Gamma^2}\right),
$$

\n
$$
B = \frac{1}{2|\Delta|^2} \left(\frac{\Gamma}{2}\right)^3,
$$

\n
$$
C = \frac{1}{2|\Delta|^2} \left(\frac{\Gamma}{2}\right)^3 \frac{1}{\gamma^2 + \delta^2} \left(\frac{\Omega^2}{2\Gamma^2} - \frac{\delta^2}{\Gamma^2}\right),
$$

\n
$$
P = \frac{1}{2|\Delta|^2} \left(\frac{\Gamma}{2}\right)^2 \left[1 + \frac{\Omega^2}{\Gamma}\left(\frac{1}{\gamma_{12}}\right)'\right].
$$
 (B7)

Thus we find

$$
P_{13} = \frac{P + \frac{\gamma}{\Omega^2}A}{\frac{\Gamma + \gamma}{\Gamma - \gamma}P + 2\gamma C + \frac{\gamma}{\Omega^2}(A + B)}
$$
(B8)

from which, making the assumption of small detuning δ , the first of the two expressions in Eq. (60) can be obtained.

- [1] R.K. Wangness and F. Bloch, Phys. Rev. 89, 728 (1953); C. Cohen Tannoudji, *Frontiers in Laser Spectroscopy*, Les Houches XXVII, edited by R. Balian, S. Haroche, and S. Liberman (North-Holland, Amsterdam, 1977).
- @2# P. Zoller, M. Marte, and D.F. Walls, Phys. Rev. A **35**, 198 (1987); G. Nienhuis, J. Stat. Phys. 53, 417 (1988); J. Dalibard, Y. Castin, and K. Mølmer, Phys. Rev. Lett. **68**, 580 (1992).
- [3] A.S. Zibrov *et al.*, Phys. Rev. Lett. **75**, 1499 (1995); G.G. Padmabandu *et al.*, *ibid.* **76**, 2053 (1996), and references therein.
- [4] B. Zambon, Phys. Rev. A 47, R38 (1993); B. Zambon, Phys. Lett. A 172, 426 (1993).
- [5] C. Cohen Tannoudji, B. Zambon, and E. Arimondo, J. Opt. Soc. Am. B 10, 2107 (1993).
- [6] L. Fonda, G.C. Ghirardi, and A. Rimini, Nuovo Cimento Soc. Ital. Fis., B 18, 1 (1973); N. Gisin, Phys. Rev. Lett. **52**, 1657 (1994)
- @7# P. Visser and G. Nienhuis, Quantum Semiclassic. Opt. **9**, 621 $(1997).$
- @8# P.M. Morse and H. Feshbach, *Methods in Theoretical Physics* (McGraw-Hill, New York, 1953).
- [9] In the equivalent quantum jump picture approach, similar to that used in $[5]$, the absorption and emission are calculated by appealing to the quantum nature of the EM field. Here, by contrast, we depict this latter as being classical and work entirely within the framework of the OBE.
- [10] O. Kocharovskaya and P. Mandel, Phys. Rev. A 42, 523 $(1990).$
- [11] G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, Nuovo Cimento Soc. Ital. Fis., B **36**, 5 ~1976!; G. Orriols, *ibid.* **53**, 1 $(1979).$
- [12] J. Lawall, S. Kulin, B. Saubamea, N. Bigelow, M. Leduc, and C. Cohen-Tannoudji, Phys. Rev. Lett. **75**, 4194 (1995), and references therein.
- [13] M.O. Scully, Phys. Rev. Lett. **67**, 1855 (1991); L.V. Hau, S.E. Harris, Z. Dutton, and C.H. Behroozi, Nature (London) 397, 594 (1999); D.F. Phillips, A. Fleischhauer, A. Mair, R.L. Walsworth, and M.D. Lukin, Phys. Rev. Lett. **86**, 783 (2001).
- $[14]$ The *P* probabilities above can also easily be determined by means of the approach in $[5]$ where Eq. (3) is made equivalent to a Schrödinger evolution with Hamiltonian H_{eff} and P_{ij} $= \gamma_j \int_0^{\infty} |\langle j| e^{-iH_{\text{eff}}t} |i \rangle|^2 dt$. For a two-level system with zero damping of the lower state, we have $P_{22}=0$ and $P_{12}=1$ as a result of Eq. (31). For the three-level system with zero damping of the lower state, by introducing the states $|a\rangle = \Omega_1|1\rangle$ $+\Omega_2|2\rangle$ and $|d\rangle = \Omega_2|1\rangle - \Omega_1|2\rangle$ as the absorbing state and the dark state, respectively, we have $P_{a,3}=1$ and $P_{d,3}=0$ as a consequence of the fact that the dark state is completely decoupled from the upper state $|3\rangle$, i.e., $H_{\text{eff}}|d\rangle=0$. Thus expressing $|1\rangle = \Omega_1|a\rangle + \Omega_2|d\rangle$, we find Eq. (57).
- [15] S.E. Harris, Phys. Today **50**(7), 36 (1997), and references therein.
- [16] P.B. Sellin, C.C. Yu, J.R. Bochinski, and T.W. Mossberg, Phys. Rev. Lett. **78**, 1432 (1997).
- [17] B. Zambon, Phys. Rev. A **58**, 690 (1997).
- [18] B. Cheng, M. Francesconi, F. Gianmanco, F. Strumia, O. Tommasi, and M. Tosetti, Opt. Commun. 97, 183 (1993).
- [19] The much larger value of the Raman stimulated emission probability P_{21} with respect to the bare one-photon stimulated

emission probability, this last given by Ω_1^2/Γ , is to be attributed to a small decay of the coherence between $|1\rangle$ and $|2\rangle$ with respect to the coherence decay between $|1\rangle$ and $|3\rangle$.

- [20] J. Mompart and R. Corbalan, J. Opt. B: Quantum Semiclassical Opt. 2, R7 (2000).
- [21] J. Schnakenberg, Rev. Mod. Phys. 48, 571 (1976).