Quantum-jump approach to dissipative processes: application to amplification without inversion

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Several recent studies have shown that the time evolution of an atom submitted to coherent laser fields and to dissipative processes, such as spontaneous emission of photons or excitation by a broadband incoherent field, can be considered to consist of a sequence of coherent evolution periods separated by quantum jumps occurring at random times. A general statistical analysis of this random sequence is presented for the case in which the number of relevant atomic states is finite and the delay functions giving the distribution of the time intervals between two successive jumps can easily be calculated. These general considerations are then applied to a simple model recently proposed for demonstrating the possibility of amplification without inversion of populations. We show how the quantum-jump approach allows one to calculate the respective contributions of the various physical processes responsible for the amplification or the attenuation of the probe field and to get new insights into the relevant physical mechanisms.

1. INTRODUCTION

During the past few years several experiments have shown that the time evolution of a single atomic system driven by coherent laser fields and subjected to dissipative processes such as spontaneous emission can exhibit discontinuous abrupt changes, also called quantum jumps.[1–3] The analysis of these experiments has stimulated the development of new theoretical approaches to dissipative processes in which the time evolution of the atomic system is pictured as consisting of a series of coherent evolution periods separated by quantum jumps occurring at random times.[4–10] The equivalence between these quantum-jump approaches and the usual description of dissipative processes in terms of master equations for the atomic-density operator has been demonstrated.[7,8] Some connection seems also to exist between these analyses and general stochastic formulations of quantum mechanics.[9–11]

When the number of relevant atomic states involved in dissipative processes is finite and when the Hamiltonian is time independent, the method of delay functions introduced in Refs. 4 and 5 is particularly convenient for performing Monte Carlo simulations of the sequence of quantum jumps.[5,6] In this paper we show how this delay-function approach can be used to derive general statistical properties of the coherent evolution periods taking place between two successive quantum jumps. In order to keep the discussion as physical as possible, we introduce the method by dealing with a simple model that was recently proposed for demonstrating the possibility of an amplification without inversion.[12,13] We show that the quantum-jump approach, based on the delay functions, gives the same results as the master-equation approach followed in Ref. 12 and provides in addition new insights into the underlying physical mechanisms. A brief account of these results, without any demonstration, was presented elsewhere.[14]

The paper is organized as follows. We first give in Section 2 a general qualitative presentation of the method. Starting from the model of Ref. 12, we introduce the basic ideas that are used throughout the paper: quantum jumps associated with dissipative processes, coherent evolution periods between two successive quantum jumps, and physical processes associated with each coherent evolution period. We then introduce in Section 3 the various quantities that fully characterize the stochastic evolution of the atom, in particular the delay functions. Some results of Monte Carlo simulations are also presented. Section 4 is devoted to the derivation of general statistical properties of the sequence of quantum jumps, and we explain how it is possible to calculate various properties of the coherent evolution periods, such as their probabilities or their mean duration. This general method is applied in Section 5 to the model introduced in Section 2, and the probabilities of the various coherent evolution periods are calculated analytically. Finally, the results of this calculation are discussed in Section 6; these allow one to present a detailed analysis of the various competing physical mechanisms. Appendixes A and B are devoted to the derivation of some properties of the coherent evolution periods.

2. PRINCIPLE OF THE METHOD

A. Model

As in Ref. 12, we consider a three-level atom with one excited state e and two lower states g_1 and g_2, forming a Λ configuration (Fig. 1). We denote by ω_{e1} and ω_{e2} the frequencies of the two allowed transitions $g_1 \leftrightarrow e$ and $g_2 \leftrightarrow e$. These two transitions are excited by two laser fields with

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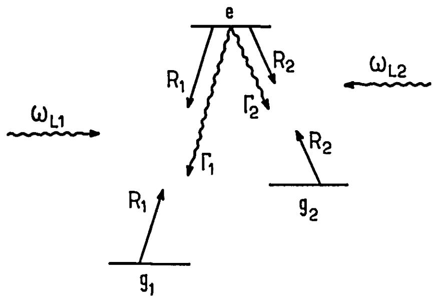

Fig. 1. Three-level atom forming a Λ configuration and subjected to dissipative processes, inducing transitions between the three levels e, g_1, g_2 with rates Γ_1, Γ_2, R_1, R_2 (arrows). The atom is also driven by two laser fields with frequencies ω_{L1} and ω_{L2} that are close, respectively, to the frequencies ω_{e1} and ω_{e2} of the two transitions $g_1 \leftrightarrow e$ and $g_2 \leftrightarrow e$.

frequencies ω_{L1} and ω_{L2} that are close, respectively, to ω_{e1} and ω_{e2}, with the corresponding detunings being

$$\delta_i = \omega_{Li} - \omega_{ei}\,, \qquad i = 1, 2\,. \tag{2.1}$$

By spontaneous emission the atom can decay from e to g_1 or to g_2 with rates equal, respectively, to Γ_1 and Γ_2 (wavy oblique arrows of Fig. 1). The atom is also assumed to be subjected to broadband incoherent fields that induce both absorption and stimulated-emission processes between e and g_1, on the one hand, and e and g_2 on the other hand, with rates equal, respectively, to R_1 and R_2 (straight oblique arrows of Fig. 1).

Such a purely radiative and closed system has been introduced by the authors of Ref. 12 to show that the field ω_{L1}, considered as a weak probe beam, can be amplified for certain values of the parameters, even if the lowest sublevel g_1 contains more than one half of the total population. Such a result is derived in Ref. 12 from the solution of the optical Bloch equations, which describe the evolution of the atomic-density operator driven by the coherent fields ω_{L1} and ω_{L2} and subjected to the dissipative processes described by Γ_1, Γ_2, R_1, and R_2. Rather than solving the optical Bloch equations, we follow here the evolution of the state vector $|\psi\rangle$ of a single atom. The usual results provided by the optical Bloch equations are recovered by averaging over different realizations of the atomic stochastic evolution.

B. Manifolds of Atom + Laser Photon States

It is convenient to use here a quantum description of the two laser fields ω_{L1} and ω_{L2}. If these fields are quasi-resonant ($|\delta_1| \ll \omega_{e1}, |\delta_2| \ll \omega_{e2}$), the states of the total system atom + laser photons are grouped into manifolds $\mathscr{E}(N_1, N_2)$ of three quasi-degenerate states, which become degenerate if $\delta_1 = \delta_2 = 0$ (see Fig. 2):

$$\mathscr{E}(N_1, N_2) = \{|e, N_1, N_2\rangle, |g_1, N_1 + 1, N_2\rangle, |g_2, N_1, N_2 + 1\rangle\}. \tag{2.2}$$

As a result of the above quasi-resonant assumption, the energy distance between two different manifolds is very large compared with the energy splittings within a given manifold.

The atom in g_1 (g_2) can absorb one ω_{L1} (ω_{L2}) photon and be transferred to e, with the corresponding coupling being characterized by the Rabi frequency Ω_1 (Ω_2). More precisely, we have

$$\langle e, N_1, N_2|V_{\mathrm{AL}}|g_1, N_1 + 1, N_2\rangle = \hbar\Omega_1/2\,, \tag{2.3a}$$

$$\langle e, N_1, N_2|V_{\mathrm{AL}}|g_2, N_1, N_2 + 1\rangle = \hbar\Omega_2/2\,, \tag{2.3b}$$

where V_{AL} is the atom–laser interaction Hamiltonian. These couplings are represented by the horizontal arrows of Fig. 2 and exist only within a given manifold. In the absence of dissipative processes, the system, initially in $\mathscr{E}(N_1, N_2)$, would remain forever in the same manifold, and its coherent evolution could be described entirely in terms of Rabi nutations between the three states of $\mathscr{E}(N_1, N_2)$.

C. Quantum Jumps Associated with Dissipative Processes

The system can leave $\mathscr{E}(N_1, N_2)$ only by a quantum jump that brings it into a neighboring manifold (oblique arrows starting from $\mathscr{E}(N_1, N_2)$ in Fig. 2). For example, it can jump from $|e, N_1, N_2\rangle$ into $|g_1, N_1, N_2\rangle$, which belongs to $\mathscr{E}(N_1 - 1, N_2)$, by a spontaneous-emission process or by an incoherent stimulated emission process with a total rate $\tilde{\Gamma}_1$ given by

$$\tilde{\Gamma}_1 = \Gamma_1 + R_1\,. \tag{2.4a}$$

Similar quantum jumps can take place between $|e, N_1, N_2\rangle$ and $|g_2, N_1, N_2\rangle$, which belongs to $\mathscr{E}(N_1, N_2 - 1)$, with a rate

$$\tilde{\Gamma}_2 = \Gamma_2 + R_2\,. \tag{2.4b}$$

The system can also jump from $|g_1, N_1 + 1, N_2\rangle$ ($|g_2, N_1, N_2 + 1\rangle$) into $|e, N_1 + 1, N_2\rangle$ ($|e, N_1, N_2 + 1\rangle$), which belongs to $\mathscr{E}(N_1 + 1, N_2)$ [$\mathscr{E}(N_1, N_2 + 1)$] by an incoherent absorption process with a rate R_1 (R_2).

Figure 2 also represents the quantum jumps bringing the system into $\mathscr{E}(N_1, N_2)$ [oblique arrows arriving in $\mathscr{E}(N_1, N_2)$], from $|g_1, N_1, N_2\rangle$ ($|g_2, N_1, N_2\rangle$) into $|e, N_1, N_2\rangle$ with a rate R_1 (R_2), and from $|e, N_1 + 1, N_2\rangle$ ($|e, N_1, N_2 + 1\rangle$) into $|g_2, N_1 + 1, N_2\rangle$ ($|g_2, N_1, N_2 + 1\rangle$) with a rate $\tilde{\Gamma}_1$ ($\tilde{\Gamma}_2$).

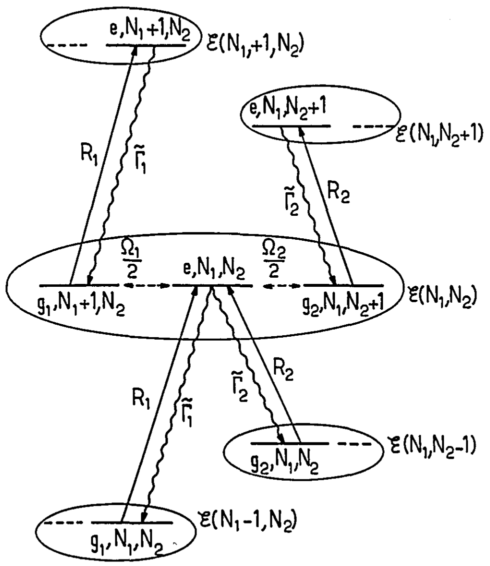

Fig. 2. Manifolds $\mathscr{E}(N_1, N_2)$ of states of the atom + laser photon system. The coherent couplings within a given manifold (horizontal arrows) are characterized by the Rabi frequencies Ω_1 and Ω_2. The oblique arrows represent quantum jumps bringing the system from one manifold to another.

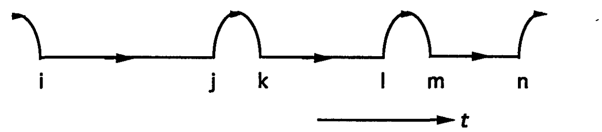

Fig. 3. Sequence of coherent evolution periods ...(i,j), (k,l), (m,n)... separated by quantum jumps from j to k, from l to m, and so on. Each coherent evolution period (i,j) is characterized by the state of entry i and the state of exit j. The duration of each period is a random variable whose distribution is given by the delay functions introduced in the text.

D. Picturing the Time Evolution

Each coherent evolution period (i,j) in a given manifold $\mathscr{E}(N_1,N_2)$ may be characterized by the state i of $\mathscr{E}(N_1,N_2)$ in which the system enters $\mathscr{E}(N_1,N_2)$, after the quantum jump of entry, and by the state j of $\mathscr{E}(N_1,N_2)$ from which the system leaves $\mathscr{E}(N_1,N_2)$, through the quantum jump of exit. To keep the notation as simple as possible, we label the states of $\mathscr{E}(N_1,N_2)$ according to the atomic state $(i,j=1,\ 2,\ \text{or}\ e)$, and we do not write the photon quantum numbers.

The time evolution of the system can thus be pictured as consisting of a series of coherent evolution periods (i,j) (k,l) (m,n) separated by quantum jumps, from the state j of $\mathscr{E}(N_1,N_2)$ to the state k of a neighboring manifold, from l to m, and so on (Fig. 3). In subsequent sections we show how it is possible to calculate the various statistical properties of the random sequence of Fig. 3. Beforehand we now explain why it is interesting to calculate the probabilities of the various periods (i,j) if we want to determine whether the field ω_{L1} is amplified or absorbed during the time evolution.

E. Time Evolution of N_1 and N_2

During a coherent evolution period in a given manifold $\mathscr{E}(N_1,N_2)$ the state vector of the system is in general a linear superposition of the three states of $\mathscr{E}(N_1,N_2)$, so that N_1 and N_2 are not well defined. In contrast, it clearly appears in Fig. 2 that each quantum jump connects two states in which N_1 and N_2 have well-defined values, which are the same just before the jump and just after the jump. Each quantum jump can thus be considered a determination of N_1 and N_2, with each period (i,j) corresponding to well-defined variations ΔN_1 and ΔN_2 of N_1 and N_2 between the jump of entry and the jump of exit.

There are actually four periods (i,j) for which ΔN_1 is not equal to zero, i.e., during which the number of probe photons, N_1, varies. Consider, for example, a period (2,1). The system enters $\mathscr{E}(N_1,N_2)$ in $|2\rangle = |g_2,N_1,N_2+1\rangle$ and leaves $\mathscr{E}(N_1,N_2)$ from $|1\rangle = |g_2,N_1+1,N_2\rangle$, so that

$$\text{period}\ (1,2) \to \Delta N_1 = +1, \Delta N_2 = -1\,. \tag{2.5a}$$

Such a period thus corresponds to a two-photon stimulated Raman process $g_2 \to g_1$, where the field ω_{L2} loses one photon, whereas the field ω_{L1} gains one. The period (1,2) corresponds to the inverse stimulated Raman process $g_1 \to g_2$, where the field ω_{L1} loses one photon, whereas the field ω_{L2} gains one:

$$\text{period}\ (1,2) \to \Delta N_1 = -1, \Delta N_2 = +1\,. \tag{2.5b}$$

We must also consider the periods $(e,1)$, where the system, starting in $|e\rangle = |e,N_1,N_2\rangle$, ends in $|1\rangle = |g_1,N_1+1,N_2\rangle$, which corresponds to the stimulated emission of one photon ω_{L1} without any change of N_2,

$$\text{period}\ (e,1) \to \Delta N_1 = +1, \Delta N_2 = 0\,, \tag{2.5c}$$

and consider the reverse periods $(1,e)$, which corresponds to the absorption of one photon ω_{L1} without any change of N_2,

$$\text{period}\ (1,e) \to \Delta N_1 = -1, \Delta N_2 = 0\,. \tag{2.5d}$$

It may easily be checked that, for the five remaining periods (i,j), $\Delta N_1 = 0$.

If we are able to calculate the relative probabilities of the four periods (2,1), (1,2), $(e,1)$ and $(1,e)$, we can thus determine whether the field ω_{L1} will be amplified or attenuated and identify the respective contributions of the various physical processes, stimulated Raman gain, stimulated Raman loss, induced emission, absorption, which are involved. This is the great advantage of the quantum-jump approach presented here as compared with the optical Bloch equations approach, which gives only the total gain (or loss).

3. CHARACTERIZATION OF THE STOCHASTIC EVOLUTION

In this section, we introduce the various quantities that are needed for characterizing completely the stochastic properties of the random sequence of Fig. 3.

A. Evolution within a Manifold

Consider first a coherent evolution period. We know that the system entered the manifold $\mathscr{E}(N_1,N_2)$ in the state $|i\rangle$ at time t, and we want to study the subsequent time evolution of the projection of the state vector of the system onto $\mathscr{E}(N_1,N_2)$. Since dissipative processes make the system leave the various states $|j\rangle$ of $\mathscr{E}(N_1,N_2)$ with well-defined rates G_j given by (see Fig. 2)

$$G_1 = R_1, \qquad G_2 = R_2, \qquad G_e = \tilde{\Gamma}_1 + \tilde{\Gamma}_2 = \tilde{\Gamma}, \tag{3.1}$$

one can show that the time evolution within $\mathscr{E}(N_1,N_2)$ is governed by an effective non-Hermitian Hamiltonian H_{eff} obtained by adding to the energies of the three states $|j\rangle$ of $\mathscr{E}(N_1,N_2)$ an imaginary part $-i\hbar G_j/2$. One possible method for demonstrating such a result is to study the projection of the resolvent operator onto $\mathscr{E}(N_1,N_2)$ (see, for example, Chap. 3 of Ref. 15 and Sec. 1 of Ref. 16). Using Eqs. (2.1) and (2.3), we thus get

$$H_{\text{eff}} = \hbar\begin{bmatrix} -iG_1/2 + \delta_1 & 0 & \Omega_1/2 \\ 0 & -iG_2/2 + \delta_2 & \Omega_2/2 \\ \Omega_1/2 & \Omega_2/2 & -iG_e/2 \end{bmatrix}. \tag{3.2}$$

From Eq. (3.2) one can calculate the probability amplitude

$$c_{ij}(\tau) = \langle j|\exp(-iH_{\text{eff}}\,\tau/\hbar)|i\rangle \tag{3.3}$$

for the system to be found in the state $|j\rangle$ of $\mathscr{E}(N_1,N_2)$ at time $t+\tau$, when it is known that it started from the state $|i\rangle$ of $\mathscr{E}(N_1,N_2)$ at time t. Multiplying $|c_{ij}(\tau)|^2$ by $G_j\mathrm{d}\tau$ then gives the conditional probability,

$$W_{ij}(\tau)\mathrm{d}\tau = G_j|c_{ij}(\tau)|^2\mathrm{d}\tau, \tag{3.4}$$

that the system leaves $\mathscr{E}(N_1, N_2)$ by a quantum jump from the state $|j\rangle$ between times $t + \tau$ and $t + \tau + d\tau$. The nine functions $W_{ij}(\tau)$, with $i, j = 1, 2, e$, give the distributions of the time intervals spent by the system in a given period (i, j). These delay functions are quite analogous to those introduced previously[4,5] for analyzing the intermittent fluorescence that can be observed on a single trapped ion.[1–3] The delay functions obey the normalization condition

$$\sum_j \int_0^\infty W_{ij}(\tau)d\tau = 1 \qquad \text{for all } i, \tag{3.5}$$

proved in Appendix A, which results from the fact that the system has certainly left $\mathscr{E}(N_1, N_2)$ after an infinite time.

B. Characterization of a Jump

From Fig. 2 one can also find the probabilities π_{kj}, if a quantum jump starts from the state $|k\rangle$ of a manifold, that this quantum jump brings the system into the state $|j\rangle$ of a neighboring manifold:

$$\pi_{1j} = \delta_{ej}, \qquad \pi_{2j} = \delta_{ej}, \qquad \pi_{ej} = \delta_{1j}\frac{\tilde{\Gamma}_1}{\tilde{\Gamma}} + \delta_{2j}\frac{\tilde{\Gamma}_2}{\tilde{\Gamma}}. \tag{3.6}$$

These probabilities are obviously normalized:

$$\sum_j \pi_{kj} = 1 \qquad \text{for all } k. \tag{3.7}$$

Knowing $W_{ik}(\tau)$ and π_{kj}, one can decide randomly the time at which the system will leave $\mathscr{E}(N_1, N_2)$ and the state $|k\rangle$ from which the corresponding jump will occur, as well as the state $|j\rangle$ in which the system will arrive after such a jump, and so on. The stochastic properties of the random sequence of Fig. 2 are thus completely determined by the knowledge of the delay functions $W_{ik}(\tau)$, i.e., by solution (3.3) of the Schrödinger equation corresponding to Eq. (3.2), and by the probabilities π_{kj}. To complete our pictures, we must also give the expression of the state vector of the system between the jump of entry in $|i\rangle$ at time t and the jump of exit from $|j\rangle$. If the jump of exit has not yet occurred at time $t + \tau$, the state vector certainly belongs to $\mathscr{E}(N_1, N_2)$ and is given by the normalized expression

$$|\psi_1(t + \tau)\rangle = \frac{\sum_k c_{ik}(\tau)|k\rangle}{\left(\sum_k |c_{ik}(\tau)|^2\right)^{1/2}}. \tag{3.8}$$

C. Monte Carlo Simulations

In Section 4 we define from $W_{ij}(\tau)$ and π_{kj} a certain number of probabilities for characterizing the mean statistical properties of the random sequence of Fig. 3. Analytical expressions are also derived for these probabilities in some limiting cases in Section 5. Beforehand we think it would be useful to give an example of Monte Carlo simulations of the time evolution of the system, because they provide nice pictorial views of the physical processes. For such simulations one can use the delay functions $W_{ij}(\tau)$ and the probabilities π_{kj} introduced in this paper, which allow one to perform fast numerical calculations. One could also follow the Monte Carlo wave-function approach of Refs. 7 and 8, which requires more computing time but which, on the other hand, is simpler to program. In fact we have used a combination of both methods to generate the stochastic sequence reproduced in Fig. 4. In Fig. 4(a) we plot the number of probe photons as a function of time, each modification in the number of probe photons corresponding to the process indicated above by a small icon. More precisely, the arrows represent stimulated Raman processes, while $\top$ and $\perp$ represent, respectively, absorption and stimulated emission processes of one photon. We notice from this figure that the total number of probe photons is increasing so that the total amplification is positive. A second observation is that this amplification is due mainly to stimulated Raman processes. In fact the stimulated emission of one photon is, for the parameters we have chosen here, a rare event. Since Fig. 4(a) is a little bit misleading because the photon number never has a well-defined value, except at the time of a quantum jump, it is better to plot the photon number as in Fig. 4(b), where, with an enlarged time scale, this value is shown only at the time of a quantum jump. Figure 4(c) shows a further magnification of the temporal sequence of the physical processes changing N_1. In the lower part of Fig. 4(c) we have also indicated the coherent evolution periods by means of oblique lines joining the entrance and

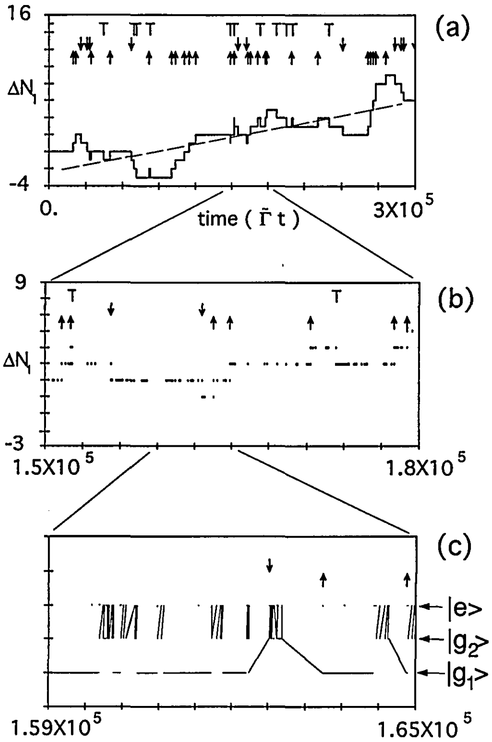

Fig. 4. Stochastic evolution of the number of probe photons obtained by the Monte Carlo simulation explained in the text. The parameters are $\tilde{\Gamma}_1 = 0.25\tilde{\Gamma}$, $\tilde{\Gamma}_2 = 0.75\tilde{\Gamma}$, $R_1 = R_2 = (2.5 \times 10^{-3})\tilde{\Gamma}$, $\Omega_1 = (2.5 \times 10^{-2})\tilde{\Gamma}$, $\Omega_2 = 0.15\tilde{\Gamma}$, $\delta_1 = \delta_2 = 0$. Changes in probe photon number are tagged by the processes that produce them; $\uparrow$ indicates stimulated Raman gain; $\downarrow$ stimulated Raman absorption; $\perp$ one-photon gain; $\top$ one-photon loss. (a) Probe photon number versus normalized time $\tilde{\Gamma}t$. The dashed line represents the mean rate of variation of N_1 computed from Eq. (4.16) below. (b) Enlarged part of the time evolution, with the number of probe photons being represented only at the time of the quantum jumps, where it has a well-defined value. One sees that between two successive changes of N_1 there are several quantum jumps during which N_1 does not change. (c) Further time enlargement allows one to distinguish the coherent evolution periods, which are represented by oblique lines joining their state in and their state out $|g_1\rangle$, $|g_2\rangle$, or $|e\rangle$.

the exit states; these atomic states are represented in the plot at three levels indicated by $|g_1\rangle$, $|g_2\rangle$, and $|e\rangle$. From these lines we can notice that a stimulated Raman gain is complete at the end of a period $(2,1)$ and a stimulated Raman loss at the end of a period $(1,2)$. There are also many transitions taking place between levels g_2 and e without affecting the probe photon number. Finally, it clearly appears that for most of the time the system is in the period $(1,1)$. We show in Subsection 5.D that in such a period the weight of the state g_1 is predominant in the wave function, so that one concludes that the lowest level is on the average the most populated, notwithstanding the positive gain of the system. This very peculiar behavior will be discussed in detail in the next sections.

4. GENERAL STATISTICAL PROPERTIES OF THE SEQUENCE OF QUANTUM JUMPS

A. Probabilities of the Periods (i, j)

Our purpose in the present section is to define some useful quantities relative to the sequence of quantum jumps and to show how, in the particular case we are dealing with, they are amenable to an analytical evaluation. A fundamental quantity here is the probability $\mathcal{P}(i,j)$ of a period (i,j), i.e., the probability that a random choice among all periods of the stochastic sequence gives as a result the period (i,j). Note that such a random choice is made independently of the duration of the period, each period being characterized only by the state of entry $|i\rangle$ and the state of exit $|j\rangle$. Since each period corresponds to a well-defined change in the photon numbers, the probabilities $\mathcal{P}(i,j)$ are directly connected to the absorption and the amplification processes. Making use of Eqs. (2.5), we can for example explicitly write the mean change of the number of probe photons per period:

$$\langle \Delta N_1\rangle = \mathcal{P}(2,1) + \mathcal{P}(e,1) - \mathcal{P}(1,2) - \mathcal{P}(1,e), \tag{4.1}$$

where the roles of the amplification periods $(2,1)$ and $(e,1)$ and of the absorption period $(1,2)$ and $(1,e)$ clearly appear. It has been also pointed out in Section 2 that during the other periods the number of probe photons does not change.

The probabilities $\mathcal{P}(i,j)$ are linked to the probabilities $\mathcal{P}(i)$, that a randomly chosen period starts in the state $|i\rangle$, by the relation

$$\mathcal{P}(i,j) = \mathcal{P}(i)\mathcal{P}(j/i), \tag{4.2}$$

where $\mathcal{P}(j/i)$ is the conditional probability that, given that the period has started in $|i\rangle$, it ends in the state $|j\rangle$. According to Eq. (3.4) we can write

$$\mathcal{P}(j/i) = \int_0^\infty W_{ij}(\tau)\mathrm{d}\tau = G_j\int_0^\infty \mathrm{d}\tau|c_{ij}(\tau)|^2, \tag{4.3}$$

with

$$\sum_j \mathcal{P}(j/i) = 1 \qquad \text{for all } i, \tag{4.4}$$

making use of Eq. (3.5).

Equation (4.3) allows us to derive an interesting relation between the conditional probabilities $\mathcal{P}(j/i)$ and $\mathcal{P}(i/j)$. In Appendix B it is shown that for an effective non-Hermitian Hamiltonian such as in Eq. (3.2), which satisfies $(H_{\text{eff}})^* = (H_{\text{eff}})^\dagger$, the following relation applies: $c_{ij}(\tau) = c_{ji}(\tau)$. Thus in Eq. (4.3) the time integrals for the two conditional probabilities are equal, and we get

$$\frac{\mathcal{P}(j/i)}{G_j} = \frac{\mathcal{P}(i/j)}{G_i}. \tag{4.5a}$$

The conditional probability of a given period is proportional to the dissipative departure rate from the final state of that period. Using Eqs. (4.2) and (4.5a), one can also derive the exact equation

$$\frac{\mathcal{P}(i,j)}{\mathcal{P}(j,i)} = \frac{\mathcal{P}(i)}{\mathcal{P}(j)}\frac{G_j}{G_i}. \tag{4.5b}$$

The probability of a given period is thus proportional to the probability of starting in the initial state and to the dissipative departure rate from the final state.

We see from Eq. (4.2) that, once the coefficients c_{ij} are derived from Eq. (3.3), it is necessary to calculate the probabilities $\mathcal{P}(i)$ if we want to determine $\mathcal{P}(i,j)$. In the hypothesis of a stationary process, $\mathcal{P}(i)$ is time independent and is related to the conditional probability $Q(\text{in: } j/\text{in: } i)$ to start a period in the state $|j\rangle$, given that the previous period has started in the state $|i\rangle$, by the equation

$$\mathcal{P}(j) = \sum_i \mathcal{P}(i)Q(\text{in: } j/\text{in } i). \tag{4.6}$$

These conditional probabilities can be written as

$$Q(\text{in: } j/\text{in: } i) = \sum_k \mathcal{P}(k/i)\pi_{kj}, \tag{4.7}$$

where the normalization condition (3.7) entails that

$$\sum_j Q(\text{in: } j/\text{in: } i) = 1 \qquad \text{for all } i. \tag{4.8}$$

This allows us to show that the homogeneous system (4.6) has a nonzero solution, which can be normalized in such a way that

$$\sum_i \mathcal{P}(i) = 1. \tag{4.9}$$

To sum up, solving the Schrödinger equation associated with Eq. (3.2) gives c_{ij} [see Eq. (3.3)] and then $\mathcal{P}(j/i)$, from Eqs. (4.3) and (3.1). One then calculates $\mathcal{P}(i)$, using Eqs. (4.6), (4.7), (3.6), and (4.9), which gives according to Eq. (4.2) the probabilities $\mathcal{P}(i,j)$.

B. Average Statistical Quantities

Several average statistical quantities can now be calculated from the probabilities introduced above. Interesting quantities are, for instance, the average duration, $T(i,j)$, of a period (i,j) and the average time, T, between two consecutive quantum jumps, given, respectively, by

$$T(i,j) = \frac{\int_0^\infty \tau W_{ij}(\tau)\mathrm{d}\tau}{\int_0^\infty W_{ij}(\tau)\mathrm{d}\tau} = \frac{G_j\int_0^\infty \tau|c_{ij}(\tau)|^2\mathrm{d}\tau}{\mathcal{P}(j/i)}, \tag{4.10}$$

$$T = \sum_{i,j} T(i,j)\mathcal{P}(i,j), \tag{4.11}$$

while the probability that, at a given randomly chosen time, the atom is found in the period (i, j) is given by

$$\Pi(i,j) = \frac{\mathcal{P}(i,j)T(i,j)}{T}. \tag{4.12}$$

In a collection of atoms this quantity defines the fraction of them in the period (i, j).

We also derive here an expression for the populations of the atomic levels, with it understood that a similar procedure can be followed for other physical observables as well. Before starting this derivation, we write Eq. (3.8) (with $t = 0$) in a more physically meaningful way, i.e.,

$$|\psi_i(\tau)\rangle = \frac{\sum_k (c_{ik}(\tau)|k\rangle}{[N_i(\tau)]^{1/2}}, \tag{4.13}$$

where we have introduced the probability $N_i(\tau)$ that no quantum jump has occurred in the time interval $[0,\tau]$ for a coherent evolution period starting at time $\tau = 0$ in $|i\rangle$:

$$N_i(\tau) = \sum_k |c_{ik}(\tau)|^2 = 1 - \int_0^\tau \sum_k W_{ik}(\tau')\mathrm{d}\tau'. \tag{4.14}$$

In deriving expression (4.14) for $N_i(\tau)$, use has been made of relation (A6) below. The average population of the state $|k\rangle$ is calculated by integration of its instantaneous value $|\langle k \mid \psi_i(\tau)\rangle|^2$ over the time $T_\infty = nT$, corresponding to a very large number ($n \to \infty$) of coherent evolution periods. The number $\mathrm{d}n_i(\tau)$ of periods starting in $|i\rangle$ and lasting a time between τ and $\tau + \mathrm{d}\tau$ is $\mathrm{d}n_i(\tau) = n\mathcal{P}(i)\Sigma_j W_{ij}(\tau)\mathrm{d}\tau$. The contribution to the average population of level $|k\rangle$ coming from a coherent evolution period that started in $|i\rangle$ is $\int_0^\tau |\langle k \mid \psi_i(\tau')\rangle|^2 \mathrm{d}\tau'$. We then obtain for the average population

$$\Pi_k = \sum_{i,j} \frac{\mathcal{P}(i)}{T} \int_0^\infty W_{ij}(\tau) \left[\int_0^\tau |\langle k \mid \psi_i(\tau')\rangle|^2 \mathrm{d}\tau' \right] \mathrm{d}\tau$$
$$= \sum_i \frac{\mathcal{P}(i)}{T} \int_0^\infty |c_{i,k}(\tau)|^2 \mathrm{d}\tau \tag{4.15}$$

after having performed an integration by parts and having used relation (4.14) for $N_i(t)$.

One final quantity to be calculated is the average rate of increase of the number of probe photons, N_1. First, it is clear from Fig. 4(a) that the increase (or decrease) of N_1 takes place only through random quantum jumps. Thus, for the time evolution of a single atom, such as that pictured in Fig. 4(a), $\mathrm{d}N_1/\mathrm{d}t$ cannot be defined. We may define only a coarse-grained rate of variation, averaged over a very large number of coherent evolution periods, n, lasting a time $T_\infty = nT$. Because of the random occurrence of the quantum jumps, such a coarse-grained rate of variation of N_1 may be approximated as the ratio between the variation in the probe photon number and the time. The average change in the photon number N_1 during each coherent evolution period is given by Eq. (4.1), so that the coarse-grained rate of variation of N_1 is equal to

$$\left\langle \frac{\mathrm{d}N_1}{\mathrm{d}t} \right\rangle = \frac{n\langle \Delta N_1 \rangle}{T_\infty} = \frac{\langle \Delta N_1 \rangle}{T}$$
$$= \frac{\mathcal{P}(2,1) + \mathcal{P}(e,1) - \mathcal{P}(1,2) - \mathcal{P}(1,e)}{\sum_{i,j} T(i,j)\mathcal{P}(i,j)}. \tag{4.16}$$

In Fig. 4 the value of $\langle \mathrm{d}N_1/\mathrm{d}t \rangle$ given by this expression is represented by the slope of the dashed line passing through the photon-number values.

5. APPLICATION TO AMPLIFICATION WITHOUT INVERSION

The determination of the delay functions introduced in Section 3 and the use of these functions in the equations for the probabilities and conditional probabilities derived in Section 4 allow one to calculate the average variation in the photon number, ΔN_1, the average time between quantum jumps, and through Eq. (4.16) the amplification coefficient corresponding to the scheme of Fig. 1. In this section such a determination will be performed analytically in the limiting case of a weak probe field and a not-too-strong pump field.

A. Assumptions

The following relations are assumed to be valid for the analytical calculations:

$$\Omega_1, \Omega_2 \ll \tilde{\Gamma}, \tag{5.1a}$$

$$R_1, R_2 \ll \Gamma_2' = \Omega_2^2/\Gamma, \tag{5.1b}$$

$$\Omega_1 \ll \Omega_2, R_1, R_2, \Gamma_2'. \tag{5.1c}$$

Combination of inequalities (5.1a) and (5.1b) leads to

$$R_1, R_2 \ll \Gamma_2' \ll \tilde{\Gamma}. \tag{5.1d}$$

These assumptions correspond to well-defined conditions to be realized in experimental configurations. Inequality (5.1a) for the Rabi frequencies expresses a weak-excitation condition, meaning that the two transitions are not saturated. Inequality (5.1b) expresses that the absorption rate Γ_2' of ω_{L2} photons from the g_2 level is larger than the absorption rate R_1 and R_2 of incoherent radiation from levels g_1 and g_2, respectively. Finally, according to inequality (5.1c), the field at frequency ω_{L1} is considered a weak probe field, as is usually done near the threshold for lasing. From these assumptions it follows that the populations of the three levels are not modified by the application of the ω_{L1} field. In contrast, owing to inequality (5.1b), the populations of levels g_2 and e are modified appreciably by the application of the ω_{L2} field. Note that while the population level g_1 is not affected by the probe field ω_{L1}, owing to the depletion of level g_2 by the pump field ω_{L2}, the population of level g_1 can become larger than the population of level g_2, so that we can realize a condition of no inversion between two levels connected by a Raman transition. In summary, the threshold conditions for the amplification of the ω_{L1} field are investigated with the assumption that we apply a field ω_{L2} strong enough to modify the population of g_2 and at the same time weak enough not to saturate the transition $g_2 \leftrightarrow e$.

Two more assumptions will be used in this section. The first one,

$$\delta_2 = 0, \tag{5.2}$$

is not essential, but allows one to write simpler analytical expressions. The second one,

$$0 \le |\delta_1| \ll \Gamma, \tag{5.3}$$

expresses that the frequency ω_{L1} can be modified for scanning the Raman resonance, with the frequency ω_{L2} remaining in resonance with the transition $g_2 \leftrightarrow e$. No hypothesis is introduced concerning the relative magnitudes of the pumping rate Γ_2' and the frequency detuning δ_1.

B. Calculation of the Relevant Probabilities

The Schrödinger equation associated with the effective Hamiltonian (3.2) leads to the following differential equations for the amplitudes $c_{ij}(\tau)$ introduced in Eq. (3.3) and describing the coherent evolution within the manifold $\mathscr{E}(N_1, N_2)$ for a system starting from the state $|i\rangle$ at time $\tau = 0$:

$$|\psi_i(\tau)\rangle = c_{ie}(\tau)|e, N_1, N_2\rangle + c_{i1}(\tau)|g_1, N_1 + 1, N_2\rangle + c_{i2}(\tau)|g_2, N_1, N_2 + 1\rangle, \tag{5.4}$$

$$\dot{c}_{ie}(\tau) = -\frac{\tilde{\Gamma}}{2}c_{ie}(\tau) - i\frac{\Omega_1}{2}c_{i1}(\tau) - i\frac{\Omega_2}{2}c_{i2}(\tau), \tag{5.5a}$$

$$\dot{c}_{i1}(\tau) = -i\frac{\Omega_1}{2}c_{ie}(\tau) - \left(\frac{R_1}{2} + i\delta_1\right)c_{i1}(\tau), \tag{5.5b}$$

$$\dot{c}_{i2}(\tau) = -i\frac{\Omega_2}{2}c_{ie}(\tau) - \frac{R_2}{2}c_{i2}(\tau), \tag{5.5c}$$

$$c_{ik}(\tau = 0) = \delta_{ik}. \tag{5.5d}$$

In these equations, use has been made of the resonant condition $\delta_2 = 0$ for the pump field at frequency ω_{L2}. Since by assumption the rate $\tilde{\Gamma}$ defines the fastest time constant, we are allowed to perform, at times $t > 1/\tilde{\Gamma}$, an adiabatic elimination of the variable c_{ie} in terms of the slow variables c_{i1} and c_{i2}. In the resulting equations, owing to inequalities (5.1), $\Omega_1^2/\tilde{\Gamma}$ can be neglected with respect to R_1, and R_2 can be neglected with respect to Γ_2', so that the following equations are obtained (for times $t > 1/\tilde{\Gamma}$):

$$\dot{c}_{ie}(\tau) = -i\frac{\Omega_1}{\tilde{\Gamma}}c_{i1}(\tau) - i\frac{\Omega_2}{\tilde{\Gamma}}c_{i2}(\tau), \tag{5.6a}$$

$$\dot{c}_{i1}(\tau) = -\left(\frac{R_1}{2} + i\delta_1\right)c_{i1}(\tau) - \frac{\Omega_1\Omega_2}{2\tilde{\Gamma}}c_{i2}(\tau), \tag{5.6b}$$

$$\dot{c}_{i2}(\tau) = -\frac{\Omega_1\Omega_2}{2\tilde{\Gamma}}c_{i1}(\tau) - \frac{\Gamma_2'}{2}c_{i2}(\tau). \tag{5.6c}$$

On the other hand, the initial conditions of Eqs. (5.5) are modified in a negligible way under the assumptions specified above. Solving Eqs. (5.6), with the proper initial conditions, and substituting the corresponding results into the relation (4.3) allow one to derive the conditional probabilities $\mathscr{P}(j/i)$. In what follows, the conditional probabilities for the four processes contributing in Eq. (4.1) to the variation of the probe photon number will be explicitly derived.

1. $g_2 \rightarrow g_1$ *Stimulated Raman Gain*

According to Eq. (2.5a) the stimulated Raman gain takes place during the periods (2, 1), whose probability $\mathscr{P}(2,1)$ depends, according to Eq. (4.2), on the conditional probability $\mathscr{P}(1/2)$. For one to calculate

$$\mathscr{P}(1/2) = R_1\int_0^\infty d\tau|c_{21}(\tau)|^2, \tag{5.7}$$

Eqs. (5.6) have to be solved with $i = 2$. Using inequalities (5.1) and neglecting terms containing $1/\Gamma_2'$ with respect to those containing $1/R_1$, we obtain the following expression:

$$\mathscr{P}(1/2) = \frac{\Omega_1^2}{\Omega_2^2}\frac{1}{1 + (2\delta_1/\Gamma_2')^2}. \tag{5.8}$$

$\mathscr{P}(1/2)$ has a maximum value Ω_1^2/Ω_2^2 at $\delta_1 = 0$ and tends to zero when the detuning δ_1 increases from zero to values larger than the linewidth of the Raman transition between levels g_2 and g_1, $(\Gamma_2' + R_1)/2 \simeq (\Gamma_2')/2$.

2. $g_1 \rightarrow g_2$ *Stimulated Raman Loss*

The stimulated Raman loss takes place during the periods (1, 2), with conditional probability $\mathscr{P}(2/1)$, to be calculated through

$$\mathscr{P}(2/1) = R_2\int_0^\infty d\tau|c_{12}(\tau)|^2. \tag{5.9}$$

The symmetry between the amplitudes $c_{12}(\tau)$ and $c_{21}(\tau)$ (see Appendix B) allows one to simplify this derivation. Making use of Eqs. (4.5a) and (3.1), we obtain

$$\mathscr{P}(1/2) = \frac{R_2}{R_1}\frac{\Omega_1^2}{\Omega_2^2}\frac{1}{1 + (2\delta_1/\Gamma_2')^2}. \tag{5.10}$$

The frequency dependence of this probability is equal to that discussed for the conditional probability of the Raman gain process.

3. $e \rightarrow g_1$ *Stimulated-Emission Gain*

Following Eq. (2.5c), we have to study the periods $(e, 1)$ and calculate

$$\mathscr{P}(1/e) = R_1\int_0^\infty d\tau|c_{e1}(\tau)|^2. \tag{5.11}$$

The calculation of $c_{e1}(\tau)$ or $c_{1e}(\tau)$ will be presented in the following paragraph. We just note here that, owing to Eqs. (4.5a) and (3.1), the following relation applies:

$$\mathscr{P}(1/e) = (R_1/\tilde{\Gamma})\mathscr{P}(e/1), \tag{5.12}$$

so that it results from inequality (5.1d) that

$$\mathscr{P}(1/e) \ll \mathscr{P}(e/1). \tag{5.13}$$

We can thus neglect stimulated-emission gain $e \rightarrow g_1$ in comparison with absorption loss $g_1 \rightarrow e$.

4. $g_1 \rightarrow e$ *Absorption Loss*

Following Eq. (2.5d), we have to study the periods $(1, e)$ and calculate

$$\mathscr{P}(e/1) = \tilde{\Gamma}\int_0^\infty d\tau|c_{1e}(\tau)|^2. \tag{5.14}$$

Equations (5.5), with $i = 1$, determine $c_{1e}(\tau)$. At times $t > 1/\tilde{\Gamma}$, where an adiabatic elimination of the variable c_{1e} is performed, Eqs. (5.6b) and (5.6c) for c_{11} and c_{12} are solved, and the solution is substituted into Eq. (5.6a). The final result for c_{1e}, with the usual approximations, is

$$c_{1e}(\tau) = \frac{\Omega_1}{\tilde{\Gamma}}\frac{1}{\delta_1 + i(\Gamma_2'/2)}\left\{\frac{\Gamma_2'}{2}\exp\left(-\frac{\Gamma_2'}{2}\tau\right) - i\delta_1\exp\left[-\left(\frac{R_1}{2} + i\delta_1\right)\tau\right]\right\}. \tag{5.15}$$

In Eq. (5.15) two separate terms contribute to c_{1e} and to $\mathcal{P}(e/1)$, one of them being zero at resonance ($\delta_1 = 0$). The general expression for $\mathcal{P}(e/1)$ results:

$$\mathcal{P}(e/1) = \frac{\Omega_1^2}{\Omega_2^2}\frac{(\Gamma_2'/2)^2}{(\Gamma_2'/2)^2 + \delta_1^2}\left[1 + \frac{4\delta_1^2}{R_1\Gamma_2'} - \frac{4\delta_1^2}{(\Gamma_2'/2)^2 + \delta_1^2}\right], \tag{5.16a}$$

with the limiting cases at resonance ($\delta_1 = 0$),

$$\mathcal{P}(e/1) = \frac{\Omega_1^2}{\Omega_2^2}, \tag{5.16b}$$

and far from resonance ($|\delta_1| \gg \Gamma_2'$),

$$\mathcal{P}(e/1) = \frac{\Omega_1^2}{\tilde{\Gamma}R_1} = \frac{\Omega_1^2}{\Omega_2^2}\frac{\Gamma_2'}{R_1}. \tag{5.16c}$$

Because of inequality (5.1b), the conditional probability for absorption $\mathcal{P}(e/1)$ takes its minimum value at resonance.

5. Probabilities $\mathcal{P}(i)$ and $\mathcal{P}(i, j)$

The next step before deriving the probabilities $\mathcal{P}(i, j)$ is to calculate the probabilities $\mathcal{P}(i)$ that a randomly chosen period starts in the state $|i\rangle$. Since all the probabilities $\mathcal{P}(j/i)$ just determined and contributing to the modification of the probe photon number ΔN_1 are proportional to Ω_1^2, it is sufficient here to calculate $\mathcal{P}(i)$ to order 0 in Ω_1.

It is clear from Fig. 2 that the system enters into levels g_1 and g_2 only through quantum jumps from $|e\rangle$, either in spontaneous-emission processes or in emission processes stimulated by the incoherent radiation. The relative probabilities of entering into $|g_1\rangle$ and $|g_2\rangle$ are proportional, respectively, to $\tilde{\Gamma}_1/\tilde{\Gamma}$ and $\tilde{\Gamma}_2/\tilde{\Gamma}$. Thus the ratio between the probabilities $\mathcal{P}(1)$ and $\mathcal{P}(2)$ of starting a period in states $|1\rangle$ and $|2\rangle$ is given exactly by

$$\frac{\mathcal{P}(1)}{\mathcal{P}(2)} = \frac{\tilde{\Gamma}_1}{\tilde{\Gamma}_2}. \tag{5.17}$$

An alternative method for determining the probabilities $\mathcal{P}(i)$ is to make use of Eqs. (4.6) and (4.9), the conditional probabilities $Q(\text{in: } j/\text{in: } i)$ being derived from Eq. (4.7). This derivation will be performed here within the assumptions specified above. In this limit we notice from Fig. 2 that, if a period starts in $|g_1\rangle$, the following period always starts from $|e\rangle$. This implies that

$$Q(\text{in: } j/\text{in: } 1) = \delta_{ej}. \tag{5.18a}$$

If instead a period starts in $|e\rangle$, at the lowest order in $\Gamma_2'/\tilde{\Gamma}$, the probabilities of quantum jumps to levels g_1 and g_2 are larger than the probability for a coherent evolution toward $|2\rangle$ within the manifold $\mathcal{E}(N_1, N_2)$ followed by a jump from this level. Thus we may write

$$Q(\text{in: } j/\text{in: } e) \simeq \frac{\tilde{\Gamma}_1}{\tilde{\Gamma}}\delta_{1j} + \frac{\tilde{\Gamma}_2}{\tilde{\Gamma}}\delta_{2j}. \tag{5.18b}$$

Finally, for a period starting in $|g_2\rangle$, in the limit $\Gamma_2' \gg R_2$, the most probable process is a coherent evolution toward the state $|e\rangle$ of the same manifold $\mathcal{E}(N_1, N_2)$ followed by a quantum jump toward g_1 or g_2. Thus we may write

$$Q(\text{in: } j/\text{in: } 2) \simeq \frac{\tilde{\Gamma}_1}{\tilde{\Gamma}}\delta_{1j} + \frac{\tilde{\Gamma}_2}{\tilde{\Gamma}}\delta_{2j}. \tag{5.18c}$$

From relations (5.18) it follows that

$$Q(\text{in: } e/\text{in: } i) \simeq \delta_{i1}. \tag{5.19}$$

Applying Eq. (4.6) to the state $|j\rangle = |e\rangle$ and using relation (5.19), we derive

$$\mathcal{P}(e) \simeq \mathcal{P}(1). \tag{5.20}$$

Finally, from Eq. (5.17) and relation (5.20), and from the normalization condition given by Eq. (4.9), we get

$$\frac{\mathcal{P}(1)}{\tilde{\Gamma}_1} = \frac{\mathcal{P}(2)}{\tilde{\Gamma}_2} = \frac{\mathcal{P}(e)}{\tilde{\Gamma}_1} = \frac{1}{2\tilde{\Gamma}_1 + \tilde{\Gamma}_2}, \tag{5.21}$$

which leads to

$$\mathcal{P}(1) = \frac{\tilde{\Gamma}_1}{2\tilde{\Gamma}_1 + \tilde{\Gamma}_2}, \qquad \mathcal{P}(2) = \frac{\tilde{\Gamma}_2}{2\tilde{\Gamma}_1 + \tilde{\Gamma}_2},$$
$$\mathcal{P}(e) = \frac{\tilde{\Gamma}_1}{2\tilde{\Gamma}_1 + \tilde{\Gamma}_2}. \tag{5.22}$$

At this point Eqs. (5.8), (5.10), (5.12), (5.16), and (5.22) represent the elements required for calculation of the probabilities of the periods contributing to a variation in the probe photon number. For the stimulated Raman processes we get

$$\mathcal{P}(2, 1) = \mathcal{P}(2)\mathcal{P}(1/2) = \frac{\Omega_1^2}{\Omega_2^2}\frac{1}{1 + (2\delta_1/\Gamma_2')^2}\frac{\tilde{\Gamma}_2}{2\tilde{\Gamma}_1 + \tilde{\Gamma}_2}, \tag{5.23}$$

$$\mathcal{P}(1, 2) = \mathcal{P}(1)\mathcal{P}(2/1) = \frac{R_2}{R_1}\frac{\Omega_1^2}{\Omega_2^2}\frac{1}{1 + (2\delta_1/\Gamma_2')^2}\frac{\tilde{\Gamma}_1}{2\tilde{\Gamma}_1 + \tilde{\Gamma}_2}, \tag{5.24}$$

while for the one-photon processes at resonance ($\delta_1 = 0$) we have

$$\mathcal{P}(e, 1) = \mathcal{P}(e)\mathcal{P}(1/e) = \frac{R_1}{\tilde{\Gamma}}\frac{\Omega_1^2}{\Omega_2^2}\frac{\tilde{\Gamma}_1}{2\tilde{\Gamma}_1 + \tilde{\Gamma}_2}, \tag{5.25a}$$

$$\mathcal{P}(1, e) = \mathcal{P}(1)\mathcal{P}(e/1) = \frac{\Omega_1^2}{\Omega_2^2}\frac{\tilde{\Gamma}_1}{2\tilde{\Gamma}_1 + \tilde{\Gamma}_2}, \tag{5.26a}$$

and far from resonance ($|\delta_1| \gg \Gamma_2'$)

$$\mathcal{P}(e, 1) = \mathcal{P}(e)\mathcal{P}(1/e) = \frac{\Omega_1^2}{\tilde{\Gamma}^2}\frac{\tilde{\Gamma}_1}{2\tilde{\Gamma}_1 + \tilde{\Gamma}_2}, \tag{5.25b}$$

$$\mathcal{P}(1, e) = \mathcal{P}(1)\mathcal{P}(e/1) = \frac{\Omega_1^2}{\tilde{\Gamma}R_1}\frac{\tilde{\Gamma}_1}{2\tilde{\Gamma}_1 + \tilde{\Gamma}_2}. \tag{5.26b}$$

Comparison of Eqs. (5.23) and (5.24) shows that

$$\frac{\mathcal{P}(1, 2)}{\tilde{\Gamma}_1 R_2} = \frac{\mathcal{P}(2, 1)}{\tilde{\Gamma}_2 R_1}. \tag{5.27}$$

In fact, such a relation could have been derived directly from the exact equations (4.2), (4.5b), and (5.17), which shows that Eq. (5.27) remains valid even if the assumptions made in Subsection 5.A were not valid. We return to the physical meaning of Eq. (5.27) in Subsection 6.A.

C. Condition for Amplification

By substitution of the probabilities for the four coherent evolution periods modifying the probe photon number into

Eq. (4.16), or more simply into Eq. (4.1), the conditions required for realizing amplification may be determined. We examine the condition $\langle \Delta N_1 \rangle > 0$ for the case $\delta_1 = 0$, where, according to Eq. (5.23), the probability $\mathcal{P}(2,1)$ of the Raman gain process is maximum, whereas, according to Eqs. (5.26), the probability of the one-photon loss process is minimum. Making use of Eqs. (5.23)–(5.27) with $\delta_1 = 0$, we obtain

$$\langle \Delta N_1 \rangle = \frac{1}{2\tilde{\Gamma}_1 + \tilde{\Gamma}_2} \frac{{\Omega_1}^2}{{\Omega_2}^2} \frac{1}{R_1} \times \left(\tilde{\Gamma}_2 R_1 + \tilde{\Gamma}_1 R_1 \frac{R_1}{\tilde{\Gamma}} - \tilde{\Gamma}_1 R_2 - \tilde{\Gamma}_1 R_1 \right). \quad (5.28)$$

The second term in parentheses on the right-hand side of this equation, i.e., the one-photon process gain $\mathcal{P}(e,1)$, can be neglected because it is much smaller than the other terms. Thus the amplification condition $\langle \Delta N_1 \rangle > 0$ may be written as

$$\frac{\tilde{\Gamma}_2}{\tilde{\Gamma}_1} > 1 + \frac{R_2}{R_1}. \quad (5.29)$$

We may also consider separately the condition for having the Raman gain larger than the Raman losses, $\mathcal{P}(2,1) > \mathcal{P}(1,2)$:

$$\frac{\tilde{\Gamma}_2}{\tilde{\Gamma}_1} > \frac{R_2}{R_1}. \quad (5.30)$$

Comparison, in inequalities (5.30) and (5.29), of the two terms appearing on the right-hand side identifies the contributions to the losses that are due to the one-photon and the Raman processes.

D. Calculation of Average Populations

The occurrence of amplification without inversion also entails an average population in level g_1 larger than the sum of populations in levels g_2 and e. Because we are interested in the conditions for reaching amplification without inversion in threshold conditions, we need the populations Π_k of levels $|k\rangle$ in the absence of the field ω_{L1}, i.e., to order 0 in Ω_1. To perform a complete analysis, we calculate the populations for both $\Omega_2 = 0$ and $\Omega_2 \neq 0$. Equations (4.15) represent the exact approach to the determination of the populations. To order 0 in Ω_1 and Ω_2 we have $|c_{ik}(\tau)|^2 = \delta_{ik} \exp(-G_i \tau)$, and from Eqs. (4.15) we deduce

$$\Pi_1 = \frac{\mathcal{P}(1)}{TR_1}; \qquad \Pi_2 = \frac{\mathcal{P}(2)}{TR_2}; \qquad \Pi_e = \frac{\mathcal{P}(e)}{T\tilde{\Gamma}}. \quad (5.31)$$

Making use of Eq. (5.21) and of the normalization condition for the populations,

$$\sum_k \Pi_k = 1, \quad (5.32)$$

we obtain

$$\frac{\Pi_1}{(\tilde{\Gamma}_1/R_1)} = \frac{\Pi_2}{(\tilde{\Gamma}_2/R_2)} = \frac{\Pi_e}{(\tilde{\Gamma}_1/\tilde{\Gamma})} = \frac{1}{(\tilde{\Gamma}_1/R_1) + (\tilde{\Gamma}_2/R_2) + (\tilde{\Gamma}_1/\tilde{\Gamma})}. \quad (5.33)$$

Owing to the hypothesis $\tilde{\Gamma} \gg R_1, R_2$, the population Π_e remains negligible in comparison with Π_1 and Π_2.

For Ω_2 different from zero and satisfying inequalities (5.1), with Ω_1 still equal to zero, the result of the first of Eqs. (5.31), for Π_1, remains the same. For the other populations we note that the eigensolutions of Eqs. (5.5) are some states $|\overline{2}\rangle$ and $|\overline{e}\rangle$ containing mainly states $|2\rangle$ and $|e\rangle$, with decay rates respectively equal to Γ_2' and $\tilde{\Gamma}$. If a period starts in state $|2\rangle$, then state $|\overline{2}\rangle$ is mainly occupied, and the integral appearing in Eq. (4.15) for Π_2 gives $1/\Gamma_2'$. On the other hand, if a period starts in state $|e\rangle$, then state $|\overline{e}\rangle$ is mainly occupied, and the integral appearing in Eq. (4.15) for Π_e gives $1/\tilde{\Gamma}$. Thus we deduce

$$\Pi_1 = \frac{\mathcal{P}(1)}{TR_1}, \qquad \Pi_2 = \frac{\mathcal{P}(2)}{T\Gamma_2'}, \qquad \Pi_e = \frac{\mathcal{P}(e)}{T\tilde{\Gamma}}. \quad (5.34)$$

Making use of Eq. (5.21) and of the normalization condition (5.32), we obtain for the populations at the lowest order in Ω_2

$$\frac{\Pi_1}{(\tilde{\Gamma}_1/R_1)} = \frac{\Pi_2}{(\tilde{\Gamma}_2/\Gamma_2')} = \frac{\Pi_e}{(\tilde{\Gamma}_1/\tilde{\Gamma})} = \frac{1}{(\tilde{\Gamma}_1/R_1) + (\tilde{\Gamma}_2/\Gamma_2') + (\tilde{\Gamma}_1/\tilde{\Gamma})}. \quad (5.35)$$

For this case the population in $|e\rangle$ is also negligible. Furthermore, if $\tilde{\Gamma}_1$ and $\tilde{\Gamma}_2$ are comparable, the condition $R_1 \ll \Gamma_2'$ [see inequality 5.1(b)] entails that Π_1 be larger than Π_2, so that the population of state g_1 is the largest one, in agreement with the results of the Monte Carlo simulations of Fig. 4(c).

Note that when these results for the populations are combined with Eqs. (5.34) and (5.22) the following expression for the average time between two consecutive quantum jumps, T, is obtained:

$$T = \frac{\mathcal{P}(1)}{\Pi_1 R_1} = \frac{\mathcal{P}(2)}{\Pi_2 \Gamma_2'} = \frac{\mathcal{P}(e)}{\Pi_e \tilde{\Gamma}} = \frac{\tilde{\Gamma}_1}{2\tilde{\Gamma}_1 + \tilde{\Gamma}_2} \left(\frac{1}{R_1} + \frac{1}{\tilde{\Gamma}} \right) + \frac{\tilde{\Gamma}_2}{2\tilde{\Gamma}_1 + \tilde{\Gamma}_2} \frac{1}{\Gamma_2'}. \quad (5.36)$$

E. Condition for Noninversion

Using the expressions derived in the previous subsection for the populations of the different states, we may now determine the conditions to be satisfied in order to realize noninversion between states g_1 and g_2, i.e., $\Pi_1 > \Pi_2$. Making use of Eqs. (5.35), for the case in which $\Omega_1 = 0$ and $\Omega_2 \neq 0$ and at the lowest order in this parameter, we obtain for the noninversion condition

$$\frac{\Gamma_2'}{R_1} > \frac{\Gamma_2}{\Gamma_1}. \quad (5.37)$$

For the case $\Omega_2 = 0$ and $\Omega_1 = 0$, the noninversion condition results from Eq. (5.33):

$$\frac{R_2}{R_1} > \frac{\Gamma_2}{\Gamma_1}. \quad (5.38)$$

One must compare these conditions for noninversion with that for amplification, inequality (5.29), for one to see whether they are compatible. It is obvious from inspection of inequalities (5.29) and (5.37) that these conditions, valid for $\Omega_2 \neq 0$, are compatible and may be combined into the double inequality

$$\frac{\Gamma_2'}{R_1} > \frac{\Gamma_2}{\Gamma_1} > 1 + \frac{R_2}{R_1}. \tag{5.39}$$

On the other hand, inequalities (5.29) and (5.38) are not compatible, i.e., for $\Omega_2 = 0$ a population inversion is realized between states g_2 and g_1. It may be noticed that in the treatment of lasers based on stimulated Raman gain the populations are usually calculated for $\Omega_1 = \Omega_2 = 0$. In contrast, in the scheme discussed here, the noninversion condition is valid only when the field at frequency ω_{L2} is applied. The reason for this difference is that in the usual stimulated Raman gain fields at both frequencies ω_{L1} and ω_{L2} are far from resonance, so that the populations, calculated for $\Omega_1 = 0$, are equal for $\Omega_2 = 0$ or $\Omega_2 \neq 0$. In contrast, in the scheme of Ref. 12 that is considered here the application of a resonant field at frequency ω_{L2} introduces radiative loss mechanisms that modify both the gain condition and the population distribution.

It has been pointed out[17] that introducing a direct decay rate between states g_2 and g_1, even a small one between those two states with the same parity, leads to a condition of amplification without inversion that is valid for both $\Omega_2 = 0$ or $\Omega_2 \neq 0$, as can be verified by application of the present quantum-jump approach.[18] Also, for this configuration the radiative losses introduced by the resonant field at frequency ω_{L2} drastically modify the gain conditions.

F. Agreement with the Optical Bloch Equations

We have confirmed that complete agreement exists among the expressions we have derived, through the quantum jump approach, for the gain condition, the noninversion condition, the gain per unit time, and the results obtained from the solution of optical Bloch equations for the density matrix of the three-level system. We have performed this comparison by solving the optical Bloch equations numerically for different sets of atom and laser parameters and by testing the equality between the gain per unit time derived from such a numerical solution with that obtained with Eq. (4.16). Furthermore, it has been verified that the conditions for gain and noninversion derived above coincide with those derived in Ref. 12 by a solution of the density-matrix equations within the assumptions specified in Subsection 5.A.

6. PHYSICAL DISCUSSION

In this section we discuss the physical content of the results derived in the previous section, and we show how the quantum-jump approach provides interesting physical insights into the physical mechanisms at the origin of amplification without population inversion.

A. Amplification Mechanism: Raman Gain versus Raman Loss

The analysis of Subsection 2.E shows that there are two physical processes permitting the probe field to be amplified: two-photon stimulated Raman processes $g_2 \to g_1$ and one-photon stimulated-emission processes $e \to g_1$. We have seen in Section 5 that the one-photon processes $e \to g_1$ make a negligible contribution if the assumptions of Subsection 5.A are fulfilled. This shows that stimulated Raman processes $g_2 \to g_1$ play a key role in determination of the amplification of the three-level system.

The balance between stimulated Raman gain and stimulated Raman loss is determined by the ratio between $\mathcal{P}(2,1)$ and $\mathcal{P}(1,2)$, which can be calculated from the exact equation (5.27). One obtains

$$\frac{\mathcal{P}(2,1)}{\mathcal{P}(1,2)} = \frac{R_2}{R_1}\frac{\tilde{\Gamma}_1}{\tilde{\Gamma}_2} = \frac{(\text{rate out of 2}) \times (\text{rate in 1})}{(\text{rate out of 1}) \times (\text{rate in 2})}. \tag{6.1}$$

It thus appears that the dissymmetry between the two stimulated inverse Raman processes $g_2 \to g_1$ and $g_1 \to g_2$ can come only from the dissymmetry between the rates of the quantum jumps through which the system enters and leaves these two states. The dissymmetry is completely independent of the amplitudes of the two laser fields, i.e., for Ω_1 and Ω_2. This problem of the dissymmetry between the transition probabilities of two inverse processes has been discussed largely in the context of amplification without inversion.[13] In fact, if the incoherent fields responsible for the rates R_1 and R_2 are thermal fields with temperature Θ_1 and Θ_2, we have

$$R_i = \frac{\Gamma_i}{\exp(\hbar\omega_{ei}/k_B\Theta_i) - 1}, \tag{6.2}$$

with $i = 1, 2$, so that condition $R_1\tilde{\Gamma}_2 > R_2\tilde{\Gamma}_1$ implies that $\Theta_1 > \Theta_2$, which is just the inversion condition for the incoherent fields introduced in Ref. 12.

B. QUENCHING OF ABSORPTION

To have the probe field amplified, it is not sufficient to have stimulated Raman gain larger than stimulated Raman loss. The one-photon absorption loss $g_1 \to e$, described by $\mathcal{P}(1,e)$, must be weak enough that $\langle \Delta N_1 \rangle$, given by Eq. (4.1), remains positive.

In this respect it is important to note the small value of $\mathcal{P}(1,e)$, which implies a quenching of the absorption process, and to understand the physical origin of this quenching. Consider for example the value of $\mathcal{P}(e/1)$, which, according to Eq. (5.16b), is equal to Ω_1^2/Ω_2^2 at resonance ($\delta_1 = 0$). A naïve argument would give, at resonance, an absorption rate from g_1 to e equal to $\Gamma_1' = \Omega_1^2/\tilde{\Gamma}$, equivalent to the rate $\Gamma_2' = \Omega_2^2/\tilde{\Gamma}$ given in relation (5.1b). Multiplying this rate Γ_1' by the mean lifetime $T_1 = 1/R_1$ of g_1 would then lead to a value of $\mathcal{P}(e/1)$ equal to

$$[\mathcal{P}(e/1)]_{\text{naïve}} = \frac{\Omega_1^2}{\tilde{\Gamma}}\frac{1}{R_1} = \frac{\Omega_1^2}{\Omega_2^2}\frac{\Gamma_2'}{R_1}, \tag{6.3}$$

which, according to inequality (5.1d), is much larger than the true value Ω_1^2/Ω_2^2. If Eq. (6.3) were true, the one-photon absorption loss would be too large to be overcome by stimulated Raman gain.

Actually, the naïve argument leading to Eq. (6.3) is not correct, because it neglects interference effects between the two absorption amplitudes starting from g_1 and g_2. The absorption from g_1 to e cannot be determined independently of the presence of the field at frequency ω_{L2} that induces transitions from g_2 to e. Such an interference between two absorption amplitudes, which represents the basis of the phenomenon of coherent population trapping,[19] to be discussed in what follows, is essential for realizing amplification without inversion. More precisely, let us introduce the following linear combinations of the two states $|g_1, N_1+1, N_2\rangle$ and $|g_2, N_1, N_2+1\rangle$ belonging to the manifold $\mathscr{E}(N_1, N_2)$:

$$|\psi_{NC}\rangle = (\Omega_2/\Omega)|g_1, N_1+1, N_2\rangle - (\Omega_1/\Omega)|g_2, N_1, N_2+1\rangle, \tag{6.4a}$$

$$|\psi_C\rangle = (\Omega_1/\Omega)|g_1, N_1+1, N_2\rangle + (\Omega_2/\Omega)|g_2, N_1, N_2+1\rangle, \tag{6.4b}$$

where

$$\Omega = (\Omega_1^2 + \Omega_2^2)^{1/2} \simeq \Omega_2, \tag{6.5}$$

with the last relation resulting from inequality (5.1c). In the state $|\psi_{NC}\rangle$, the two absorption amplitudes from $|g_1, N_1+1, N_2\rangle$ and $|g_2, N_1, N_2+1\rangle$ toward $|e, N_1, N_2\rangle$ interfere destructively.[19] Using Eqs. (2.3), we get

$$\langle e, N_1, N_2|V_{AL}|\psi_{NC}\rangle = 0. \tag{6.6}$$

In contrast, these absorption amplitudes interfere constructively in the coupled state $|\psi_C\rangle$, since

$$\langle e, N_1, N_2|V_{AL}|\psi_C\rangle = \hbar\Omega/2 \simeq \hbar\Omega_2/2. \tag{6.7}$$

Inverting Eqs. (6.4), one gets the following relation between the state $|g_1, N_1+1, N_2\rangle$ and the states $|\psi_{NC}\rangle$ and $|\psi_C\rangle$:

$$|g_1, N_1+1, N_2\rangle = (\Omega_2/\Omega)|\psi_{NC}\rangle + (\Omega_1/\Omega)|\psi_C\rangle. \tag{6.8}$$

Since $\Omega_1 \ll \Omega_2 \simeq \Omega$, Eq. (6.8) shows that $|g_1, N_1+1, N_2\rangle$ nearly coincides with $|\psi_{NC}\rangle$. Thus it is only because $|g_1, N_1+1, N_2\rangle$ is slightly contaminated by $|\psi_C\rangle$ (with a weight Ω_1^2/Ω_2^2) that a small absorption can take place from state g_1. One understands in this way why the absorption of one photon ω_{L1} from g_1 is quenched.

In the previous discussion we have supposed the two detunings δ_1 and δ_2 to be equal to zero. In such a case the two states $|g_1, N_1+1, N_2\rangle$ and $|g_2, N_1, N_2+1\rangle$ are degenerate in the manifold $\mathscr{E}(N_1, N_2)$. It follows that the two linear combinations $|\psi_{NC}\rangle$ and $|\psi_C\rangle$ are, as $|g_1, N_1+1, N_2\rangle$ and $|g_2, N_1, N_2+1\rangle$, eigenstates of the unperturbed atom–laser photon Hamiltonian H_0 (i.e., without the interaction Hamiltonian V_{AL}). Consequently, the state $|\psi_{NC}\rangle$ is, at resonance, a stationary state with respect to H_0. Such a state thus is not only insensitive to V_{AL} [see Eq. (6.6)] but also does not evolve under the effect of H_0. If δ_1 is no longer equal to zero, the two states $|g_1, N_1+1, N_2\rangle$ and $|g_2, N_1, N_2+1\rangle$, which are still eigenstates of H_0, are no longer degenerate, so that $|\psi_{NC}\rangle$ is no longer stationary with respect to H_0. There is a nonzero off-diagonal element of H_0 between $|\psi_{NC}\rangle$ and $|\psi_C\rangle$, which is of the order of $\hbar\delta_1$. As a consequence of this coupling, a system initially in $|\psi_{NC}\rangle$ can be transferred by H_0 into $|\psi_C\rangle$, from where it can be excited to $|e, N_1, N_2\rangle$ by V_{AL} [see relation (6.7)]. Out of resonance ($\delta_1 \neq 0$), an atom in the state $|\psi_{NC}\rangle$ can thus absorb the probe field ω_{L1}. The critical value of δ_1, characterizing the breaking of the quenching of absorption, is such that the off-diagonal coupling $\hbar\delta_1$ induced by H_0 between $|\psi_{NC}\rangle$ and $|\psi_C\rangle$ is of the order of the radiative width $\hbar\Gamma_2'$ of $|\psi_C\rangle$. One understands in this way why, when $|\delta_1| \gg \Gamma_2'$, the correct value of $\mathscr{P}(e/1)$, given by Eq. (5.16c), coincides with the value deduced from the naïve argument neglecting interference effects and leading to Eq. (6.3).

These general considerations are confirmed by the results of numerical calculations of $\mathscr{P}(1,e)$ and $\mathscr{P}(2,1)$ versus δ_1, represented in Fig. 5. In Fig. 5(a), δ_2 is supposed to be equal to zero. Near $\delta_1 = 0$, in a very narrow interval with a width of the order of Γ_2', $\mathscr{P}(1,e)$ is strongly quenched, whereas $\mathscr{P}(2,1)$ is enhanced. If δ_2 is no longer equal to zero [Fig. 5(b)], the same effect (enhancement of the Raman gain and quenching of the absorption loss) still exists, but it now occurs near $\delta_1 = \delta_2$, i.e., near the resonance condition for the Raman processes.

We conclude this subsection with two remarks:

(i) A diagrammatic approach for understanding the quenching of absorption in the Λ configuration of Fig. 1 was recently introduced.[20] At the lowest order in Ω_1 there are two interfering diagrams, allowing the atom to reach e from g_1 (see Fig. 5 of Ref. 20): the first diagram corresponds to the direct one-photon absorption process $g_1 \to e$; the second corresponds to a three-photon process that consists of a two-photon stimulated Raman process $g_1 \to g_2$ followed by a one-photon absorption process $g_2 \to e$. These two ways of reaching e in fact correspond to the two terms on the right-hand sides of Eqs. (5.6a) and (5.15). If the broad level e is considered a continuum, the first and second interfering diagrams correspond, respectively, to a direct transition to the continuum and to an indirect transition through the narrow discrete level g_2 with a width Γ_2'. In this way one can interpret the narrow structures appearing for $\mathscr{P}(1,e)$ in Fig. 5 as being characteristic of a Fano profile.

Another example of quenching of absorption by destructive interference between two physical processes was recently proposed for explaining the physical origin of amplification without inversion near the central resonance of the Mollow absorption spectrum.[21]

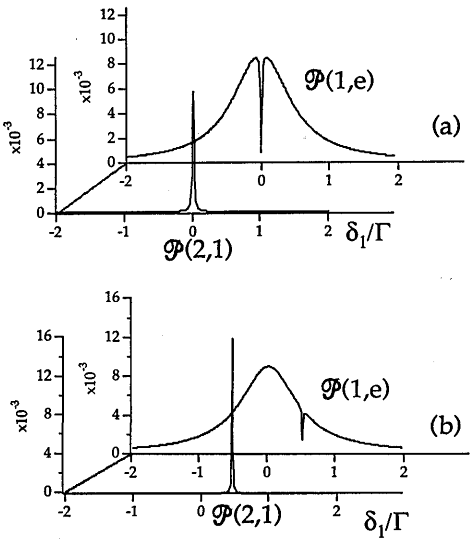

Fig. 5. Probabilities $\mathscr{P}(2,1)$ and $\mathscr{P}(1,e)$ for the coherent evolution periods (2, 1) and (1, e) contributing to the gain or loss of the probe photon number versus the detuning δ_1 of the probe laser at frequency ω_{L1}. The parameters are the same as in Fig. 4, with (a) $\delta_1 = 0$ and (b) $\delta_1 = -0.5\Gamma$.

(ii) The quantum interference effects discussed in this subsection play an equivalent role in the absorption and stimulated-emission processes between e and g_1. In fact the stimulated-emission process from $|e, N_1, N_2\rangle$ to $|g_1, N_1 + 1, N_2\rangle$ depends on the contamination of $|\psi_C\rangle$ in the linear combination of Eqs. (6.4), exactly as in the absorption process. Another simple way of getting this result is to use Eq. (5.12), which shows that $\mathcal{P}(1/e)$ and $\mathcal{P}(e/1)$ have the same δ_1 dependence, reflecting the presence of quantum interference effects. Note also that $\mathcal{P}(1, e)$ and $\mathcal{P}(e, 1)$ are not connected by an equation similar to Eq. (6.1), because $\mathcal{P}(1)$ and $\mathcal{P}(e)$ are not related by an exact equation analogous to Eq. (5.17).

C. Dissymmetry between the One-Photon Absorption Processes from g_1 and g_2

In the previous subsection we have seen that the absorption of the ω_{L1} field is quenched by the presence of the ω_{L2} field. There is no similar effect for the absorption of the ω_{L2} field, which is not quenched by the presence of the ω_{L1} field. Such a dissymmetry arises because we have supposed that $\Omega_1 \ll \Omega_2$, so that the equation equivalent to Eq. (6.8),

$$|g_2, N_1, N_2 + 1\rangle = (\Omega_2/\Omega)|\psi_C\rangle - (\Omega_1/\Omega)|\psi_{NC}\rangle, \qquad (6.9)$$

shows that $|g_2, N_1, N_2 + 1\rangle$ nearly coincides with the coupled state $|\psi_C\rangle$. There is therefore a one-photon absorption rate from g_2 that is correctly described by the parameter Γ_2' introduced in inequality (5.1b).

The dissymmetry between $\mathcal{P}(1, e)$ and $\mathcal{P}(2, e)$ is important for achieving amplification without inversion. We have seen in Subsection 6.A that the balance between stimulated Raman gain and stimulated Raman loss is independent of the amplitude of the two laser fields, i.e., of Ω_1 and Ω_2. When Ω_1 and Ω_2 are different from zero, with $\Omega_1 \ll \Omega_2$, the population of g_1 is not modified appreciably because of the quenching of absorption from g_1 discussed in Subsection 6.B. In contrast, if $\Gamma_2' \gg R_2$, as we have supposed in inequality (5.1b), the absence of quenching of absorption from g_2 results in the fact that the population of g_2 is considerably reduced from its value for $\Omega_2 = 0$, so that g_2 becomes less populated than g_1.

7. CONCLUSION

In this section we summarize the main results, which have been obtained by applying a quantum-jump approach to the model of reference.[12] The respective contributions of the various physical processes responsible for the absorption or the amplification of the probe field ω_{L1} have been identified. Analytical expressions have been obtained for the probabilities of these processes in threshold conditions for the field ω_{L1} and in the limit where the field ω_{L2} is strong enough for modifying the population of level g_2 but also weak enough for not saturating the transition $g_2 \leftrightarrow e$.

In such conditions amplification is due to the two-photon stimulated Raman processes $g_2 \to g_1$, which predominate over the inverse processes $g_1 \to g_2$ if there is a proper dissymmetry between the rates in and out of g_1 and g_2 [see Eq. (6.1)]. Quantum interference effects play an essential role in the limit $\Omega_1 \ll \Omega_2$ by quenching the one-photon absorption processes $g_1 \to e$. Within the same limit they do not change the one-photon absorption processes $g_2 \to e$, which can thus deplete g_2, since we have assumed that the corresponding coherent absorption rate Γ_2' is larger than the pumping rate R_2 of the incoherent fields. One can thus understand in this way how the atom can spend most of its time in g_1, which then becomes more populated than g_2, without the introduction of too-large absorption losses that otherwise would prevent the amplification of the field ω_{L1}.

In this problem interference effects do not introduce a dissymmetry between the one-photon absorption processes $g_1 \to e$ and the reverse stimulated processes $e \to g_1$. They do not modify $\mathcal{P}(e, 1)/\mathcal{P}(1, e)$. In contrast, they do introduce a dissymmetry between the one-photon absorption processes $g_1 \to e$ and $g_2 \to e$ starting, respectively, from g_1 and g_2. They modify $\mathcal{P}(1, e)/\mathcal{P}(2, e)$.

The quantum approach presented in this paper could be extended in several directions. We focused our attention here on average quantities such as the mean rate of increase of N_1 represented by the dotted line of Fig. 4(a). Using Monte Carlo simulations or analytical calculations, one could investigate fluctuations around mean value and related physical effects such as laser linewidth and time correlations. Including, in the Monte Carlo simulations, the variations of the Rabi frequency Ω_1 that are due to the increase of N_1, one could also explore nonlinearities, for example in the laser startup. Finally, regimes other than the one considered here (with both detunings δ_1 and δ_2 different from zero, saturation of the transition $g_2 \leftrightarrow e$, and so on) could be analyzed for determination of the optimal conditions for experimental investigations.

APPENDIX A: NORMALIZATION OF THE DELAY FUNCTIONS

In this appendix we prove Eq. (3.5), which is the normalization condition for the delay functions. By use of Eq. (3.4), that condition may be written as

$$\sum_j \int_0^\infty W_{ij}(\tau)\mathrm{d}\tau = \sum_j G_j \int_0^\infty |c_{ij}(\tau)|^2 \mathrm{d}\tau = 1 \qquad \text{for all } i. \quad \text{(A1)}$$

The coefficients c_{ij} obey differential equations derived from the Schrödinger equation corresponding to the effective Hamiltonian (3.2). For the off-diagonal elements of that Hamiltonian, which are the Rabi frequencies associated with the Hermitian atom–laser interaction operator, the following notation is introduced:

$$\left\langle j \left| \frac{H_{\text{eff}}}{\hbar} \right| k \right\rangle = \frac{\Omega_{jk}}{2} = \left\langle k \left| \frac{H_{\text{eff}}}{\hbar} \right| j \right\rangle^* = \frac{\Omega_{kj}{}^*}{2}. \qquad \text{(A2)}$$

The equations for the coefficients c_{ij} may be written as

$$\dot{c}_{ij}(\tau) = -\left(\frac{G_j}{2} + i\delta_j\right)c_{ij} - i\sum_k \frac{\Omega_{jk}}{2} c_{ik} \qquad \text{with } j = 1, 2, e, \qquad \text{(A3)}$$

where we have introduced $\delta_e = 0$. The initial conditions $c_{ij}(0)$ depend on the coherent evolution period, but they satisfy the normalization condition

$$\sum_j |c_{ij}(0)|^2 = 1. \qquad \text{(A4)}$$

Using Eq. (A3) and its complex conjugate and summing over the index j, we obtain

$$\frac{\mathrm{d}}{\mathrm{d}\tau}\sum_j |c_{ij}(\tau)|^2 = -\sum_j G_j|c_{ij}(\tau)|^2 - i\sum_{j,k}\left[c_{ij}^*(\tau)\frac{\Omega_{jk}}{2}c_{ik}(\tau) - c_{ik}^*(\tau)\frac{\Omega_{kj}^*}{2}c_{ij}(\tau)\right] = -\sum_j G_j|c_{ij}(\tau)|^2. \tag{A5}$$

We have used Eq. (A2) to replace Ω_{kj}^* and exchanged the j and k indices to find that the second term on the right-hand side vanishes. Integration of Eq. (A5) on the $(0,\tau)$ interval then leads to

$$\sum_j |c_{ij}(\tau)|^2 = \sum_j |c_{ij}(0)|^2 - \sum_j G_j\int_0^\tau |c_{ij}(\tau')|^2\mathrm{d}\tau' = 1 - \sum_j G_j\int_0^\tau |c_{ij}(\tau')|^2\mathrm{d}\tau', \tag{A6}$$

where use has been made of Eq. (A4). If we let τ tend to ∞ and if we use the fact that $c_{ij}(+\infty) = 0$ for all j, because at $\tau = +\infty$ the system has certainly left the manifold $\mathscr{E}(N_1, N_2)$, we get Eq. (A1).

APPENDIX B: SYMMETRY OF THE c_{ij} COEFFICIENTS

In this appendix we prove that, if the expansion of $|\psi(t)\rangle$ on an orthonormal basis $|j\rangle$ is used,

$$|\psi(t)\rangle = \sum_j c_{ij}(t)|j\rangle, \tag{B1}$$

with the initial conditions

$$c_{ij}(t=0) = \delta_{ij} \tag{B2}$$

in order to describe the time evolution under a Hamiltonian represented by a matrix H such that

$$(H)^* = (H)^\dagger, \tag{B3}$$

then we have the following symmetry relation:

$$c_{ij}(t) = c_{ji}(t). \tag{B4}$$

Owing to Eq. (3.3), this symmetry relation may also be written as

$$\langle j|\exp(-iHt/\hbar)|i\rangle = \langle i|\exp(-iHt/\hbar)|j\rangle. \tag{B5}$$

In our analysis the effective Hamiltonian H_{eff}, given by Eq. (3.2), has for off-diagonal elements the Rabi frequencies associated with the Hermitian atom–laser interaction operator, as in Eq. (A2). The diagonal elements of the effective Hamiltonian

$$\left\langle i\left|\frac{H_{\text{eff}}}{\hbar}\right|i\right\rangle = \delta_i - i\hbar\frac{G_i}{2} \tag{B6}$$

contain an imaginary part describing the departure rate G_i from state $|i\rangle$ that is due to the dissipative processes. Thus the Hamiltonian H_{eff} satisfies the conditions expressed by Eq. (B3).

Let $|\phi_\alpha\rangle$ be the eigenvector of H with eigenvalue E_α:

$$H|\phi_\alpha\rangle = E_\alpha|\phi_\alpha\rangle. \tag{B7}$$

Since H is not Hermitian, its eigenvalues E_α are not real and its eigenvectors $|\phi_\alpha\rangle$ are not orthogonal.

We may use an expansion of $|\phi_\alpha\rangle$ on the orthonormal basis $|i\rangle$ with coefficients $c_\alpha{}^i$:

$$|\phi_\alpha\rangle = \sum_i c_\alpha{}^i|i\rangle. \tag{B8}$$

From this expansion we may introduce a new vecotr $|\overline{\phi}_\alpha\rangle$, defined as

$$|\overline{\phi}_\alpha\rangle = \sum_i (c_\alpha{}^i)^*|i\rangle. \tag{B9}$$

This vector has several interesting properties. Inserting expansion (B8) into Eq. (B7) and taking the complex conjugate, we get

$$H^*|\overline{\phi}_\alpha\rangle = E_\alpha{}^*|\overline{\phi}_\alpha\rangle. \tag{B10}$$

From the adjoint of this relation and from Eq. (B2), we derive

$$\langle\overline{\phi}_\alpha|H = E_\alpha\langle\overline{\phi}_\alpha|. \tag{B11}$$

The dual set of vectors $|\phi_\alpha\rangle$ and $|\overline{\phi}_\alpha\rangle$ is called a biorthogonal set.[22] It is always possible to normalize $|\phi_\alpha\rangle$ in such a way that

$$\langle\overline{\phi}_\alpha|\phi_\alpha\rangle = 1. \tag{B12}$$

We now demonstrate that, for nondegenerate eigenvalues E_α and E_β, we have

$$\langle\overline{\phi}_\beta|\phi_\alpha\rangle = \delta_{\alpha\beta}. \tag{B13}$$

Consider $\langle\overline{\phi}_\beta|H|\phi_\alpha\rangle$. Applying Eqs. (B7) and (B11), we get

$$\langle\overline{\phi}_\beta|H|\phi_\alpha\rangle = E_\alpha\langle\overline{\phi}_\beta|\phi_\alpha\rangle = E_\beta\langle\overline{\phi}_\beta|\phi_\alpha\rangle. \tag{B14}$$

The equality between the last two terms shows that if $E_\alpha \neq E_\beta$ then $\langle\overline{\phi}_\beta|\phi_\alpha\rangle = 0$, which proves Eq. (B13).

We show now that the operator $\Sigma_\beta|\phi_\beta\rangle\langle\overline{\phi}_\beta|$ is the identity operator I. Applying this operator to any eigenvector $|\phi_\alpha\rangle$, we obtain

$$\left(\sum_\beta |\phi\beta\rangle\langle\overline{\phi}_\beta|\right)|\phi_\alpha\rangle = \sum_\beta |\phi_\beta\rangle\delta_{\alpha\beta} = |\phi_\alpha\rangle. \tag{B15}$$

If the $|\phi_\alpha\rangle$'s form a complete set, Eq. (B15) implies that

$$\sum_\beta |\phi_\beta\rangle\langle\overline{\phi}_\beta| = I. \tag{B16}$$

Making use of Eqs. (B7) and (B16), we get

$$H = \sum_\alpha E_\alpha|\phi_\alpha\rangle\langle\overline{\phi}_\alpha|, \tag{B17}$$

$$\exp(-iHt/\hbar) = \sum_\alpha \exp(-iE_\alpha t/\hbar)|\phi_\alpha\rangle\langle\overline{\phi}_\alpha|. \tag{B18}$$

Let us now calculate the amplitude $\langle j|\exp(-iHt/\hbar)|i\rangle$. Using Eqs. (B18), (B8), and (B9), we derive

$$\begin{aligned}\langle j|\exp(-iHt/\hbar)|i\rangle &= \sum_{\alpha}\exp(-iE_{\alpha}t/\hbar)\langle j\,|\,\phi_{\alpha}\rangle\langle\overline{\phi}_{\alpha}|i\rangle \\ &= \sum_{\alpha}\exp(-iE_{\alpha}t/\hbar)c_{\alpha}{}^{j}c_{\alpha}{}^{i} \\ &= \langle i|\exp(-iHt/\hbar)|\,j\rangle, \qquad \text{(B19)}\end{aligned}$$

since i and j play a symmetric role in the second line. This proves Eqs. (B5) and (B4).

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