

Amplification without inversion: The double- Λ scheme

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We study the propagation of a bichromatic field in three-level and four-level media. In the case of a three-level medium, we consider the Λ configuration and demonstrate that if the upper level is not empty there is a critical value for the low-frequency coherence such that below this threshold and for intense fields, coherent bleaching occurs. This implies a sharp reduction of the field absorption and the fluorescence emission whereas above this threshold a parametric instability takes place. It corresponds to an amplification of the field that does not need a population inversion between the optical transitions. Finally we prove that the field alone cannot create a low-frequency coherence exceeding this threshold. Therefore we analyze the double- Λ configuration involving a four-level medium in which the upper and lower pairs of atomic levels have a low-frequency separation while optical transitions take place between the two pairs of states. An intense bichromatic field that is resonant with one upper state and the lower levels and a weak bichromatic field resonant with the other upper state and the same two lower states are sent into the medium. We analyze the limit in which the low-frequency coherence created between the lower pair of states has a relaxation time which is large. Then there will be population trapping in the lower states leading to coherent bleaching if there is no inversion between the upper levels but there will be amplification if there is inversion between the upper levels. In this way amplification at the optical transitions without population inversion between these states is realized.

I. INTRODUCTION

When an atomic system interacts with light, it can amplify the electromagnetic radiation when gain exceeds loss. This requires that transitions from the upper level to the lower level prevail over the transitions from the lower level to the upper level. If one deals with a two-level system, the only way to obtain amplification is to create a state of population inversion, i.e., the upper level must be more densely populated than the lower level. As a result, it is commonly believed that population inversion is necessary for laser action. This conclusion, however, does not hold in general, when more than two levels are implied in the interaction process. In that case alternative schemes are possible and a mechanism that can be used then is associated with the destructive interference of optical transitions from which an asymmetry between the up- and down-transitions is made possible. Specifically, in the Λ scheme which is displayed on Fig. 1, the field causes transitions between an upper level and a pair of lower levels characterized by a low-frequency separation. Using straightforward quantum mechanics, it can be shown that there exists a linear superposition of the two low-lying states such that no transitions can be induced between the upper state and the superposition state due to destructive interferences.¹⁻⁴ Therefore, this state is called a trapped state. Meanwhile, there is a second linear superposition of the two low-lying states which is orthogonal to the trapped state. If a transition between the upper and lower states is possible at all, transitions between the upper state and this other superposition state will occur. Since the rate of the downward transition is proportional to the upper-level population,

any amount of population in the upper level is sufficient for lasing action. In other words, if the atoms are prepared in the state containing such a trapped state, we must compare the upper-level population not with the total population of the lower levels, but only with the population of the state orthogonal to the trapped state. Hence it is possible to extract the energy from the medium stored in the upper atomic level and to achieve light amplification even when the lower-level population greatly exceeds the upper-level population. In other words, "lasing without inversion" may be possible when part of the atoms are trapped, which removes them from the subspace of states with which the upper level can have transitions.

For example, it was shown by the Pisa group¹ that atoms will be trapped in such a coherent superposition state under the action of two monochromatic fields whose frequencies ω_a and ω_b satisfy the low-frequency (LF) resonance condition $\omega_a - \omega_b = \omega_{21}$. As a result, if the atoms were initially in the ground state, the upper level remained almost empty. Therefore the fluorescence from the medium decreased sharply. Similar results were obtained soon afterwards by Gray *et al.* as well.³

A similar phenomenon occurs when the atoms interact with a periodic train of ultrashort pulses.^{5,6} In this case, LF resonance means that the splitting frequency ω_{21} between the two low-lying states is an integer multiple of the pulse repetition frequency $\omega_{21} = m\Omega$ and the relaxation time τ of LF coherence is large enough: $\tau \gg 2\pi/\Omega$.

The recent interest in these phenomena is due to its many applications in high-resolution spectroscopy,^{5,7} atom cooling,^{8,9} in various schemes of multistage photoionization^{10,11} and optical bistability,^{12,13} in frequency

standards,¹⁴ for laser mode locking,¹⁵ and quenching of spontaneous emission noise in lasers.¹⁶ Some of these possibilities have already been demonstrated experimentally.

In this paper we focus our attention on the properties of an electromagnetic field propagating in a medium in which population trapping is realized. If the population trapping is due only to the propagating field, no amplification is possible but bleaching of the atomic system can be reached for sufficiently powerful fields.^{6,17} Nevertheless, light amplification without inversion is possible if an external source is used to produce the coherent superposition state. This was shown recently by considering ultrashort pulse propagation in a broadband three-level medium with a Λ configuration when this pulse interacts simultaneously with both optical transitions.¹⁸ The same idea of amplification without inversion was proposed independently by Harris¹⁹ and by Scully *et al.*²⁰ for different situations. In Harris' study, the amplifying medium consists of four-level atoms with two upper levels which are homogeneously broadened and decay to an identical continuum. In the work of Scully *et al.*,²⁰ three-level atoms pass through a resonant cavity and interact with the same monochromatic field at both optical transitions.

Other schemes exploiting the LF coherence can be proposed and two of them are analyzed in this paper, which is therefore divided in two parts. In the first part, which contains Secs. II and III, we shall consider the simple Λ scheme (see Fig. 1). The second part contains the other sections and deals with the double- Λ scheme (see Fig. 2). In the simple Λ scheme, a three-level medium interacts with two monochromatic fields, each field interacting with its own resonant transition only. In Sec. II we study the linear regime of propagation and determine the normal solutions of this problem, i.e., the plane-wave solutions. In particular, we find the dispersion relation of these normal solutions. This leads to the characterization of stable solutions whose energy remains finite (and smaller than the input energy) and unstable solutions whose energy diverges. These unstable solutions are associated with a parametric instability which can occur only when the LF coherence exceeds a critical threshold. In Sec. III the self-consistent nonlinear regime of propagation is considered and it is shown that the bichromatic field cannot create a LF coherence that is large enough to lead to unstable solutions, i.e., to amplification. At most, coherent bleaching, corresponding to a significant decrease of the absorption by the medium, can be reached in suitable conditions. Therefore, we propose in the remainder of this paper the analysis of the double- Λ scheme (see Fig. 2) in which a first bichromatic field (with partial amplitudes α and β) interacts with the low-lying states and one upper state, while a second bichromatic field (with partial amplitudes α' and β') interacts with the same low-lying states and a fourth level. If this second bichromatic field is sufficiently powerful, it can create a large LF coherence that will lead to amplification of the first bichromatic field. In Sec. IV we first make a qualitative analysis of the double- Λ scheme and determine the conditions for amplification of the fields α and β . We

then formulate the full set of field and matter equations required for a systematic study of the model. In Sec. V we study the linear regime of propagation. We find the steady state of the double- Λ system driven by the bichromatic field and show that there is coherent population trapping in the system. When the LF coherence relaxation time is large enough, the populations of all levels remain practically unaffected despite the strong bichromatic field action. The rigorous condition of amplification without inversion is obtained. The laws of pump depletion and amplified field propagation are obtained to first order in the weak amplified fields. It is shown that even when the amplification condition holds for short distances, it can be broken as propagation takes place over longer distances. In Sec. VI we study the behavior of the double- Λ system driven by the pair of bichromatic fields. The explicit solution of the density-matrix equations for the symmetric resonance case is found. We show that even when the intensity of both bichromatic fields diverges, the lower-level populations can, under appropriate conditions, remain practically unchanged: Only population redistribution between the two upper levels takes place. This means that there is coherent population trapping in such a system as well. Section VII is devoted to the investigation of the two bichromatic fields' nonlinear propagation when the LF coherence excitation mediates an interaction between them. It is shown that energy transfer of the pump field into the amplified field with an increase of the frequency is possible and the intensity of the amplified output radiation can exceed significantly the input pump radiation. This process takes place without population inversion at any optical transitions. The only population inversion that is necessary is between the upper levels.

II. PARAMETRIC INSTABILITY IN A THREE-LEVEL MEDIUM

We consider the wave equations for the slowly varying complex amplitudes of a bichromatic field,

$$E = \frac{1}{2} [E_a \exp(-i\omega_a t + ik_a z) + E_b \exp(-i\omega_b t + ik_b z) + c.c.],$$

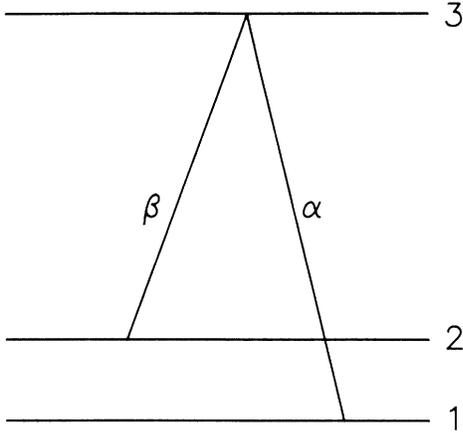
in a three-level medium with a Λ configuration of the energy levels 1, 2, and 3, as shown in Fig. 1:

$$\begin{aligned} \frac{\partial E_a}{\partial z} + c_a^{-1} \frac{\partial E_a}{\partial t} + \kappa_a E_a &= 4\pi i N \omega_a \mu_{13} \sigma_{31} / c_a \epsilon_a, \\ \frac{\partial E_b}{\partial z} + c_b^{-1} \frac{\partial E_b}{\partial t} + \kappa_b E_b &= 4\pi i N \omega_b \mu_{23} \sigma_{32} / c_b \epsilon_b, \end{aligned} \quad (2.1)$$

where σ_{31} and σ_{32} are slowly varying off-diagonal elements of the density matrix

$$\rho_{31} = \sigma_{31} \exp(-i\omega_a t), \quad \rho_{32} = \sigma_{32} \exp(-i\omega_b t).$$

In these expressions, μ_{ij} is the dipole matrix element between levels i and j while N is the atomic density; σ_j , ϵ_j , and $c_j = c / (\epsilon_j)^{1/2}$ (with $j = a$ or b) are, respectively, the ohmic conductivity of the nonresonant medium, the dielectric permittivity, and the velocity of light at the fre-

FIG. 1. The simple Λ scheme.

quency ω_j and wave number $k_j = \omega_j/c_j$. The phenomenological field damping rates are defined by $\kappa_j = 2\pi\sigma_j/c_j\epsilon_j$. In terms of the reduced complex field amplitudes,

$$\alpha = \mu_{31}E_a/2\hbar, \quad \beta = \mu_{32}E_b/2\hbar, \quad (2.2)$$

the propagation equations become

$$\frac{\partial\alpha}{\partial z} + c_a^{-1}\frac{\partial\alpha}{\partial t} + \kappa_a\alpha = 2\pi i N\omega_a |\mu_{31}|^2 \sigma_{31}/c_a\epsilon_a\hbar, \quad (2.3a)$$

$$\frac{\partial\beta}{\partial z} + c_b^{-1}\frac{\partial\beta}{\partial t} + \kappa_b\beta = 2\pi i N\omega_b |\mu_{32}|^2 \sigma_{32}/c_b\epsilon_b\hbar. \quad (2.3b)$$

The equations for the complex amplitudes σ_{31} and σ_{32} in the slowly varying amplitude approximation are

$$\frac{\partial\sigma_{31}}{\partial t} = -\sigma_{31}(\gamma_{31} + i\delta_a) + i\alpha n_{13} + i\beta\sigma_{21}, \quad (2.4)$$

$$\frac{\partial\sigma_{32}}{\partial t} = -\sigma_{32}(\gamma_{32} + i\delta_b) + i\beta n_{23} + i\alpha\sigma_{21}^*,$$

in terms of σ_{21} which characterizes the LF transition (LF coherence)

$$\rho_{21} = \sigma_{21}\exp[-i(\omega_a - \omega_b)t],$$

and the auxiliary functions

$$\begin{aligned} n_{13} &= \rho_{11} - \rho_{33}, & n_{23} &= \rho_{22} - \rho_{33}, \\ \delta_a &= \omega_{31} - \omega_a, & \delta_b &= \omega_{32} - \omega_b. \end{aligned} \quad (2.5)$$

The relaxation rates for the atomic polarizations are γ_{31} and γ_{32} while ρ_{ii} is the occupation probability of the level i .

Let us first study the linear regime of propagation. For this purpose we may assume that n_{13} , n_{23} , and ρ_{21} are constants defined by some external sources and seek the normal solutions of Eqs. (2.3) and (2.4), i.e., plane waves:

$$\alpha, \beta, \sigma_{31}, \sigma_{32} \sim \exp(-i\omega t + ikz). \quad (2.6)$$

This leads to the relations

$$\begin{aligned} \sigma_{31} &= i(\beta\sigma_{21} + \alpha n_{13})/(\gamma_{31} + i\delta_a - i\omega), \\ \sigma_{32} &= i(\beta n_{23} + \alpha\sigma_{21}^*)/(\gamma_{32} + i\delta_b - i\omega), \end{aligned} \quad (2.7)$$

and two algebraic linear equations for the field amplitudes

$$\begin{aligned} [i(k - \omega/c_a) + \kappa_a + g_a n_{13}]\alpha + g_a\sigma_{21}\beta &= 0, \\ g_b\sigma_{21}^*\alpha + [i(k - \omega/c_b) + \kappa_b + g_b n_{23}]\beta &= 0. \end{aligned} \quad (2.8)$$

The compatibility condition of the system (2.8) gives the dispersion relation of the normal solutions:

$$\begin{aligned} (k - \omega/c_a)(k - \omega/c_b) - i(k - \omega/c_a)(\kappa_b + g_b n_{23}) \\ - i(k - \omega/c_b)(\kappa_a + g_a n_{13}) + L = 0, \end{aligned} \quad (2.9)$$

where

$$L = g_a g_b |\sigma_{21}|^2 - (\kappa_a + g_a n_{13})(\kappa_b + g_b n_{23}),$$

$$g_a = \frac{2\pi\omega_a |\mu_{31}|^2 N}{\gamma_{31}\epsilon_a c_a \hbar [1 + i(\delta_a - \omega)/\gamma_{31}]},$$

$$g_b = \frac{2\pi\omega_b |\mu_{32}|^2 N}{\gamma_{32}\epsilon_b c_b \hbar [1 + i(\delta_b - \omega)/\gamma_{32}]}.$$

Thus the normal solutions are linear superpositions of plane waves whose amplitudes are coupled to each other by the relation

$$\beta/\alpha = -(g_b\sigma_{21}^*)/[i(k - \omega/c_b) + \kappa_b + g_b n_{23}]. \quad (2.10)$$

In the particular case $c_a = c_b = c'$, when the dispersion of the nonresonant medium is not important, the dispersion equation Eq. (2.9) has the solutions

$$\begin{aligned} k - \omega/c' &= (i/2)\{\Gamma_a + \Gamma_b \pm [(\Gamma_a - \Gamma_b)^2 \\ &\quad + 4g_a g_b |\sigma_{21}|^2]^{1/2}\}, \end{aligned} \quad (2.11)$$

$$\Gamma_a = \kappa_a + g_a n_{13}, \quad \Gamma_b = \kappa_b + g_b n_{23}.$$

These solutions define two dispersion relations for normal waves. Setting ω real leads from (2.11) to two branches of solutions $k_{\pm}(\omega)$. The normal solutions (2.6) will be unstable iff

$$\text{Im}(k) < 0. \quad (2.12)$$

The maximum gain is achieved at line center ($\delta_a = \delta_b = \omega$) and the frequency range of instability is defined by the optical line broadening far enough from the threshold of instability. At line center, the instability condition (2.12) is equivalent to $L > 0$. In the case of lossless propagation ($\kappa_a = \kappa_b = 0$), the condition $L > 0$ takes the form

$$|\sigma_{21}|^2 > n_{13}n_{23}. \quad (2.13)$$

Hence, if the LF coherence is large enough, a parametric instability will develop. This means that both optical fields α and β will increase without bound during the propagation through the inversionless medium. It should be emphasized that condition (2.13) can be fulfilled only

when the upper level is not empty. Indeed, if it is empty, the condition (2.13) takes the form

$$|\rho_{21}|^2 > \rho_{11}\rho_{22},$$

which violates the condition of positive definition of the density matrix. Hence, the amplification is made possible due to the extraction of energy from the medium stored by the atoms in the upper level.

When the atomic polarization relaxes on a time scale much shorter than the field-evolution time scale, we can eliminate adiabatically the atomic polarization at the optical transitions from Eqs. (2.4):

$$\begin{aligned} \sigma_{31} &= i(\beta\sigma_{21} + \alpha n_{13}) / (\gamma_{31} + i\delta_a), \\ \sigma_{32} &= i(\alpha\sigma_{21}^* + \beta n_{23}) / (\gamma_{32} + i\delta_b). \end{aligned} \quad (2.14)$$

Let us substitute (2.14) into (2.3). Then, the field equations becomes

$$\frac{\partial\alpha}{\partial z} + (c')^{-1} \frac{\partial\alpha}{\partial t} + \kappa_a \alpha = -h_a(\beta\sigma_{21} + \alpha n_{13}), \quad (2.15a)$$

$$\frac{\partial\beta}{\partial z} + (c')^{-1} \frac{\partial\beta}{\partial t} + \kappa_b \beta = -h_b(\alpha\sigma_{21}^* + \beta n_{23}), \quad (2.15b)$$

where $h_j \equiv g_j(\omega=0)$. Thus this approximation does not take into account the dispersion of the normal waves (i.e., the ω dependence of α and β). For simplicity, we set $c_a = c_b = c'$. One can show from Eqs. (2.15) that the instability corresponding to the condition (2.12) or (2.13) appears due to the coupling of the partial waves of amplitude α and β which contribute to the energy with different signs. To prove this point, let us multiply (2.15a) by α^* and (2.15b) by β^* . Then, in the case of exact resonance $\omega_a = \omega_{31}$ and $\omega_b = \omega_{32}$, we have the law of energy transfer:

$$\frac{\partial W}{\partial t} + c' \frac{\partial W}{\partial z} = -Q, \quad (2.16)$$

where the energy density W and the loss density Q are defined by

$$W = |\alpha|^2/h_a - |\beta|^2/h_b, \quad (2.17a)$$

$$Q = 2c'[(n_{13} + \kappa_a/h_a)|\alpha|^2 - (n_{23} + \kappa_b/h_b)|\beta|^2]. \quad (2.17b)$$

Hence we see that the field α has a positive contribution to the energy (2.17a) whereas the field β has a negative contribution to the energy. This property explains the possibility of a parametric instability which requires that the energies of both optical waves of amplitudes α and β will increase without violating the law of energy conservation.²¹ Furthermore, the normal waves have opposite signs of energy; indeed, using the definition (2.17a) for the energy and the coupling relation (2.10), we obtain

$$W_{\pm} = \frac{2|\alpha|^2(\Gamma'_b - \Gamma'_a)}{h_a \{ \Gamma'_b - \Gamma'_a \mp [(\Gamma'_b - \Gamma'_a)^2 + 4h_a h_b |\sigma_{21}|^2]^{1/2} \}}, \quad (2.18)$$

$$\Gamma'_a = \Gamma_a/h_a, \quad \Gamma'_b = \Gamma_b/h_b,$$

from which it is obvious that $W_+ W_- < 0$.

The conclusion of this section is that the physical mechanism that leads to amplification without inversion in the case of the Λ scheme is the parametric instability of two waves which contribute with opposite signs to the total field energy and propagate in a resonant three-level medium possessing LF coherence and partial occupation of the upper level. This raises the question of how to create a LF coherence large enough to fulfill the instability condition (2.13). This problem will be analyzed in Sec. IV and subsequent sections.

III. COHERENT BLEACHING OF A THREE-LEVEL MEDIUM

In Sec. II we made a linear analysis of the propagation equations. In this section we undertake a nonlinear analysis of the propagation equations to verify that the LF coherence created by a bichromatic field (i) cannot sustain the field amplification in a self-consistent way and (ii) leads essentially to a decrease of the field absorption, i.e., to coherent bleaching of the medium.

Let us investigate the propagation of a bichromatic field through a three-level medium in a general case when the population of the upper level before interacting with the field is not zero (when it is zero, amplification is obviously impossible). Because we want to solve the problem in a self-consistent way, we must consider the sets of equations (2.3) and (2.4) together with the equations for the LF coherence σ_{21} and the population differences n_{13} and n_{23} :

$$\begin{aligned} \frac{\partial\sigma_{21}}{\partial t} + \sigma_{21}[\gamma_2 + i(\delta_a - \delta_b)] &= i(\beta^*\sigma_{31} - \alpha\sigma_{32}^*), \\ \frac{\partial n_{13}}{\partial t} + \gamma_3(\alpha_1 + w_1 n_{13} + w_2 n_{23}) &= 2 \operatorname{Im}(2\alpha\sigma_{13} + \beta\sigma_{23}), \\ \frac{\partial n_{23}}{\partial t} + \gamma_3(\alpha_2 + w_1^2 n_{13} + w_2^2 n_{23}) &= 2 \operatorname{Im}(2\beta\sigma_{23} + \alpha\sigma_{13}). \end{aligned} \quad (3.1)$$

In these equations γ_2 is the relaxation rate of the LF coherence, γ_3 is the relaxation rate of the population differences at the optical transitions 1-3 and 2-3, γ_1 will denote the relaxation rate of n_{21} , and we have introduced the notations

$$\begin{aligned} \alpha_i &= \frac{1}{3}[3\rho_{33}^{(0)} + (-1 + \gamma_1/\gamma_3)\rho_{kk}^{(0)} - (2 + \gamma_1/\gamma_3)\rho_{ii}^{(0)}], \quad i \neq k \\ w_i^k &= -\alpha_k - (\gamma_1/\gamma_3)\rho_{kk}^{(0)} + \rho_{33}^{(0)}, \\ w_i^i &= -\alpha_i + 2\rho_{33}^{(0)} + (\gamma_1/\gamma_3)\rho_{kk}^{(0)}, \end{aligned}$$

where i and $k = 1, 2$. The superscript 0 refers to the state of the system in the absence of the external fields but un-

der the simultaneous action of the pump (if any), the spontaneous emission, and other nonradiative processes.

In the adiabatic approximation (2.14) for the atomic polarizations, we have the equations

$$\begin{aligned} \frac{\partial \sigma_{21}}{\partial t} + \sigma_{21}[\gamma_2 + i(\delta_a - \delta_b)] \\ + |\alpha|^2/(\gamma_{32} - i\delta_b) + |\beta|^2/(\gamma_{31} + i\delta_a) \\ = -\alpha\beta^*[n_{23}/(\gamma_{32} - i\delta_b) + n_{13}/(\gamma_{31} + i\delta_a)], \\ \frac{\partial n_{13}}{\partial t} = -\gamma_3(\alpha_1 + w_1 n_{13} + w_2 n_{23}) \\ - 2 \operatorname{Re}[2\alpha(n_{13}\alpha^* + \beta^*\sigma_{21}^*)/(\gamma_{31} - i\delta_a) \\ + \beta(n_{23}\beta^* + \alpha^*\sigma_{21})/(\gamma_{32} - i\delta_b)], \quad (3.2) \end{aligned}$$

$$\begin{aligned} \frac{\partial n_{23}}{\partial t} = -\gamma_3(\alpha_2 + w_2 n_{23} + w_1 n_{13}) \\ - 2 \operatorname{Re}[2\beta(n_{23}\beta^* + \alpha^*\sigma_{21})/(\gamma_{32} - i\delta_b) \\ + \alpha(n_{13}\alpha^* + \beta^*\sigma_{21}^*)/(\gamma_{31} - i\delta_a)]. \end{aligned}$$

Let us consider the simplified symmetric case

$$\gamma_{31} = \gamma_{32} \equiv \gamma, \quad |\mu_{31}| = |\mu_{32}| \equiv \mu, \quad h_a = h_b \equiv h, \\ \omega_a = \omega_{31}, \quad \omega_b = \omega_{32}, \quad \kappa_a = \kappa_b = 0, \quad \rho_{11}^{(0)} = \rho_{22}^{(0)}.$$

The last condition is a reasonable approximation in thermal equilibrium if the splitting frequency is not too large: $\omega_{21} \ll kT/\hbar$. In this simplified symmetric case, we have

$$\begin{aligned} \alpha_1 = \alpha_2 = -n_0 = \rho_{33}^{(0)} - \rho_{11}^{(0)}, \\ w_1^1 = w_2^2 = \rho_{33}^{(0)} + \rho_{11}^{(0)}(1 + \gamma_1/\gamma_3), \\ w_2^1 = w_1^2 = \rho_{11}^{(0)}(1 - \gamma_1/\gamma_3). \end{aligned}$$

If the intensities of the two input fields are equal $|\alpha(0)|^2 = |\beta(0)|^2$, one easily verifies that a stationary regime of propagation can be reached in which $n_{13} = n_{23} \equiv n$ and $|\alpha|^2 = |\beta|^2$. Let us introduce the polar decompositions

$$\sigma_{21} = \sigma e^{i\theta}, \quad \alpha = |\alpha| e^{i\varphi_\alpha}, \quad \beta = |\alpha| e^{i\varphi_\beta}.$$

Then, we have from (2.15) and (3.2) the following equations for the steady-state amplitudes:

$$\frac{d|\alpha|}{dz} = -h|\alpha|(n - \sigma), \quad (3.3a)$$

$$\sigma = 2|\alpha|^2 n / (\gamma\gamma_2 + 2|\alpha|^2), \quad (3.3b)$$

$$n = n_0 - 6|\alpha|^2(n - \sigma)/\gamma_3\gamma, \quad (3.3c)$$

where we have used the steady phase relation $\Psi \equiv \theta - (\varphi_\alpha - \varphi_\beta) = \Psi_0 = \pi \pmod{2\pi}$ and Ψ_0 is the input value of the phase Ψ . Let us substitute (3.3b) into (3.3c):

$$n = n_0(1 + I/I_c) / [1 + I(1/I_c + 3/I_s)]. \quad (3.4)$$

Using (3.3b) and (3.4), we derive from (3.3a) the following equation:

$$\frac{dI}{dz} = -(2hn_0) / [1 + I(3/I_s + 1/I_c)], \quad (3.5)$$

where the intensity is defined by $I = c|E|^2/8\pi$, $I_s = c\hbar^2\gamma_3\gamma/4\pi\mu^2$ is the saturation intensity, and $I_c = c\hbar^2\gamma_2\gamma/4\pi\mu^2$ is the coherence intensity. In terms of the rescaled intensity,

$$x = I(3/I_s + 1/I_c) \quad \text{and} \quad x_0 = I_0(3/I_s + 1/I_c), \quad (3.6)$$

where I_0 is the input intensity, the integration of (3.5) gives the law of intensity transfer,

$$2 \int_0^z hn_0 dz = x_0 - x - \ln(x/x_0). \quad (3.7)$$

Let us show that this result describes a reduction of the field absorption, i.e., bleaching of the medium by the input fields. We define as usual the extinction length l_c as the propagation length after which the field is attenuated by a factor e . In the case $\gamma_3 \gg \gamma_2$ (which corresponds to $I_c \ll I_s$) and at high input intensity defined by the condition $I_0 \gg I_c$, the radiation extinction length l_c is increased by a factor I_0/I_c compared to the linear regime where $l_c \sim 1/(2hn_0)$.

The mechanism of such coherent bleaching is different from saturation bleaching. Indeed, it occurs even if the field intensity is smaller than the saturation intensity since the inequality $I_0 \ll I_s$ is compatible with the conditions $I_0 \gg I_c$ and $I_s \gg I_c$. This effect was investigated recently^{6,17} for the restricted case $\rho_{33}^{(0)} = 0$. Furthermore, we see from (3.4) that $n = n_{13} = n_{23}$ does not tend to vanish (as would be the case for the saturation bleaching of the optical transitions), even in the high intensity limit. In that limit we have from (3.3b) that $\sigma \rightarrow n$ and, from (3.3a), that the absorption coefficient $(n - \sigma)h$ decreases to zero. At the same time, we can see from (3.3b) that the instability condition $\sigma > n$ [see Eq. (2.13)] cannot be realized at any intensity. Therefore, amplification is impossible, in agreement with the law (3.7). This means that, even when there is energy in a medium stored by the atoms in the upper levels ($\rho_{33}^{(0)} \neq 0$), it cannot be extracted by means of such a simple Λ scheme.

IV. THE DOUBLE- Λ SCHEME

In this and the following sections we will show that the amplification condition (2.13) can be fulfilled if another bichromatic field with partial amplitudes E'_a and E'_b resonant to adjacent optical transitions 1-4 and 2-4 (see Fig. 2) is used to create the LF coherence. We stress that the level 4 can be above or below the level 3.

First of all, as a guideline for the following calculations, we consider the linear stage of amplification when the amplified fields E_a and E_b are small enough and do not influence the state of the medium. In this domain the double- Λ scheme can be approximated under suitable conditions by two simple Λ schemes, one involving the transitions 1-3 and 2-3 and the second involving the transitions 1-4 and 2-4. An example of conditions under which this approximation holds is the combination of the limit (5.10) and the condition $\gamma_2 \ll \gamma_4$ discussed in Sec. V. Furthermore, we restrict this calculation to the simple case of exact resonance and complete symmetry

between the levels 1 and 2, which implies the equalities

$$\omega_a = \omega_{31}, \quad \omega_b = \omega_{32}, \quad \omega'_a = \omega_{41}, \quad \omega'_b = \omega_{42},$$

$$\rho_{11}^{(0)} = \rho_{22}^{(0)}, \quad \mu_{31} = \mu_{32}, \quad \mu_{41} = \mu_{42},$$

$$\gamma_{31} = \gamma_{32}, \quad \gamma_{41} = \gamma_{42},$$

where the parameters with the variable 4 have the same meaning as the corresponding parameters with the variable 3. We can estimate the maximum value of the LF coherence which is excited by the intense pump fields E'_a

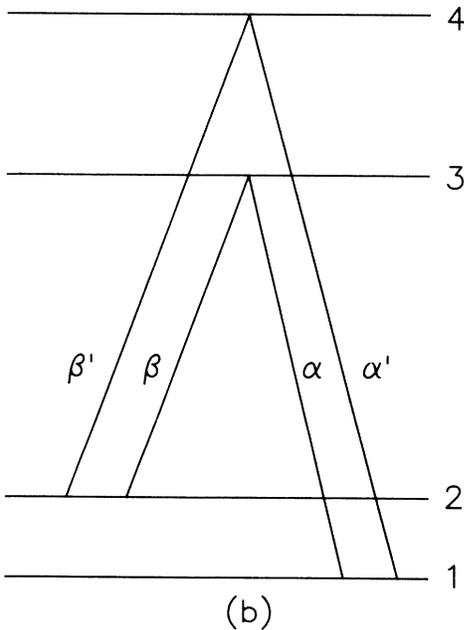
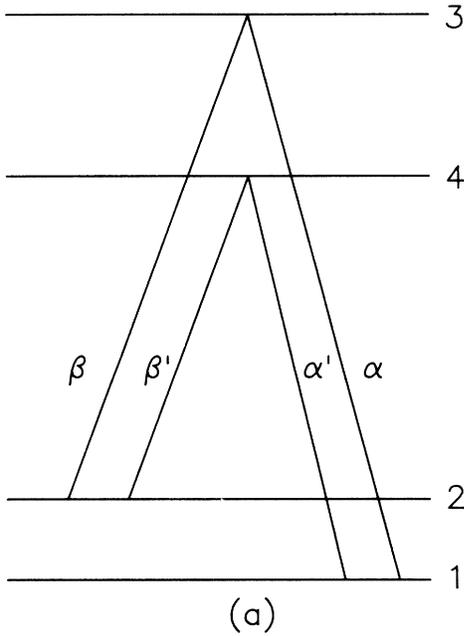


FIG. 2. The two possible double- Λ schemes.

and E'_b on the basis of the results derived at the end of Sec. III. The LF coherence and the population difference are given by (3.3b) and (3.4) with n and n_0 referring to the population difference between levels 1 or 2 and level 4. The upper bound for the LF coherence is $\sigma^{\max} = n'_0 = n_{14} = n_{24}$. Let us substitute this into the inequality (2.13), taking into account $n_{13} = n_{23} = n \approx n_0$. Then the instability condition (2.13) becomes $n'_0 > n_0$, which can also be written $n_{34}^{(0)} = \rho_{33}^{(0)} - \rho_{44}^{(0)} > 0$. It means that the level 3 must be more densely occupied than the level 4. Hence, under thermodynamic equilibrium population distribution, amplification is possible only if level 3 lies below level 4: The frequencies of the fields E_a and E_b are smaller than the frequencies of the input fields (down-conversion process). On the contrary, to obtain amplification with up-conversion, we must create a population inversion between levels 3 and 4. The linear gain $G = |\text{Im}[k(\omega=0)]|$ of such a process is derived from the solution (2.11) of the dispersion equation and is

$$G = \frac{1}{2} \left\{ -n_0(h_a + h_b) + [n_0^2(h_a + h_b)^2 + 4h_a h_b n_{34}^{(0)}(n_0 + n'_0)]^{1/2} \right\}. \quad (4.1)$$

When $n_0 \gg n_{34}^{(0)}$, which is realized in thermodynamic equilibrium, we have

$$G = \frac{h_a h_b n_{34}^{(0)}(n_0 + n'_0)}{n_0(h_a + h_b)}. \quad (4.2)$$

Specifically, for $h_a = h_b = h$ and $\rho_{11}^{(0)} + \rho_{22}^{(0)} \approx 1$, we have

$$G = h n_{34}^{(0)}. \quad (4.3)$$

Thus the gain is the same as for a two-level medium, except that it depends on the population difference $n_{34}^{(0)}$ of the transition 3–4 instead of population difference $n_{13} = n_{23}$ at the optical transition 1–3. It should be emphasized that there is no population inversion for the optical transitions.

Clearly this is only a qualitative analysis and we shall now begin a more rigorous study of the double- Λ scheme. The complete set of equations describing our system contains four wave equations for the bichromatic pump field α' and β' defined by

$$\alpha' = \mu_{41} E'_a / 2\hbar, \quad \beta' = \mu_{42} E'_b / 2\hbar,$$

and the bichromatic amplified field α and β defined by (2.2):

$$\frac{\partial \alpha}{\partial z} + c_a^{-1} \frac{\partial \alpha}{\partial t} + \kappa_a \alpha = 2\pi i \omega_a N |\mu_{13}|^2 \sigma_{31} / c_a \epsilon_a \hbar, \quad (4.4a)$$

$$\frac{\partial \beta}{\partial z} + c_b^{-1} \frac{\partial \beta}{\partial t} + \kappa_b \beta = 2\pi i \omega_b N |\mu_{23}|^2 \sigma_{32} / c_b \epsilon_b \hbar, \quad (4.4b)$$

$$\frac{\partial \alpha'}{\partial z} + (c'_a)^{-1} \frac{\partial \alpha'}{\partial t} + \kappa'_a \alpha' = 2\pi i \omega'_a N |\mu_{14}|^2 \sigma_{41} / c'_a \epsilon'_a \hbar, \quad (4.4c)$$

$$\frac{\partial \beta'}{\partial z} + (c'_b)^{-1} \frac{\partial \beta'}{\partial t} + \kappa'_b \beta' = 2\pi i \omega'_b N |\mu_{24}|^2 \sigma_{42} / c'_b \epsilon'_b \hbar, \quad (4.4d)$$

and the density-matrix equations of the four-level system driven by the pair of bichromatic fields:

$$\begin{aligned} \frac{\partial \sigma_{31}}{\partial t} + \sigma_{31}(\gamma_{31} + i\delta_a) &= i(\alpha n_{13} + \beta \sigma_{21} - \alpha' \sigma_{34}), \\ \frac{\partial \sigma_{32}}{\partial t} + \sigma_{32}(\gamma_{32} + i\delta_b) &= i(\beta n_{23} + \alpha \sigma_{21}^* - \beta' \sigma_{34}), \\ \frac{\partial \sigma_{41}}{\partial t} + \sigma_{41}(\gamma_{41} + i\delta'_a) &= i(\alpha' n_{14} + \beta' \sigma_{21} - \alpha \sigma_{34}^*), \\ \frac{\partial \sigma_{42}}{\partial t} + \sigma_{42}(\gamma_{42} + i\delta'_b) &= i(\beta' n_{24} + \alpha' \sigma_{21}^* - \beta \sigma_{34}^*), \\ \frac{\partial \sigma_{21}}{\partial t} + \sigma_{21}[\gamma_2 + i(\delta_a - \delta_b)] &= i[\beta^* \sigma_{31} - \alpha \sigma_{32}^* + (\beta')^* \sigma_{41} - \alpha' \sigma_{42}^*], \\ \frac{\partial \sigma_{34}}{\partial t} + \sigma_{34}[\gamma_{34} + i(\delta'_a - \delta'_b)] &= i[\alpha \sigma_{41}^* + \beta \sigma_{42}^* - (\alpha')^* \sigma_{31} + (\beta')^* \sigma_{32}], \\ \frac{\partial \rho_{11}}{\partial t} - R_1 &= i[\alpha^* \sigma_{31} + (\alpha')^* \sigma_{41} - \text{c.c.}], \\ \frac{\partial \rho_{22}}{\partial t} - R_2 &= i[\beta^* \sigma_{32} + (\beta')^* \sigma_{42} - \text{c.c.}], \\ \frac{\partial \rho_{33}}{\partial t} - R_3 &= i(\alpha \sigma_{13} + \beta \sigma_{23} - \text{c.c.}), \\ \frac{\partial \rho_{44}}{\partial t} - R_4 &= i(\alpha' \sigma_{14} + \beta' \sigma_{24} - \text{c.c.}), \\ \rho_{11} + \rho_{22} + \rho_{33} + \rho_{44} &= 1, \end{aligned} \quad (4.5)$$

with the definitions

$$\delta'_a = \omega_{41} - \omega'_a, \quad \delta'_b = \omega_{42} - \omega'_b.$$

We consider the case when frequency differences between harmonics of the pump and the amplified fields are equal:

$$\omega_a - \omega_b = \omega'_a - \omega'_b.$$

Here, R_1 , R_2 , R_3 , and R_4 describe the population relaxation processes:

$$\begin{aligned} R_1 &= \gamma_4(\rho_{44}\rho_{11}^{(0)} - \rho_{44}^{(0)}\rho_{11}) + \gamma_3(\rho_{33}\rho_{11}^{(0)} - \rho_{33}^{(0)}\rho_{11}) \\ &\quad + \gamma_1(\rho_{22}\rho_{11}^{(0)} - \rho_{22}^{(0)}\rho_{11}), \\ R_2 &= \gamma_4(\rho_{44}\rho_{22}^{(0)} - \rho_{44}^{(0)}\rho_{22}) + \gamma_3(\rho_{33}\rho_{22}^{(0)} - \rho_{33}^{(0)}\rho_{22}) \\ &\quad + \gamma_1(\rho_{11}\rho_{22}^{(0)} - \rho_{11}^{(0)}\rho_{22}), \\ R_3 &= \gamma_{33}(\rho_{44}\rho_{33}^{(0)} - \rho_{44}^{(0)}\rho_{33}) \\ &\quad + \gamma_3[\rho_{33}^{(0)}(\rho_{11} + \rho_{22}) - \rho_{33}(\rho_{11}^{(0)} + \rho_{22}^{(0)})], \\ R_4 &= \gamma_{33}(\rho_{33}\rho_{44}^{(0)} - \rho_{33}^{(0)}\rho_{44}) \\ &\quad + \gamma_4[\rho_{44}^{(0)}(\rho_{11} + \rho_{22}) - \rho_{44}(\rho_{11}^{(0)} + \rho_{22}^{(0)})]. \end{aligned}$$

In these expressions, γ_4 is the relaxation rate of the population differences between the optical transitions 1–4 and 2–4; γ_{33} and γ_{34} are, respectively, the longitudinal and transverse relaxation rates for the transition 3–4. The transitions 1–2 and 3–4 are considered as forbidden.

V. AMPLIFICATION WITHOUT INVERSION: LINEAR REGIME

We first study the linear regime of amplification using the field equations (4.4). For the atomic variables in the linear approximation, we may set the amplified fields α and β equal to zero which leads from Eq. (4.5) to the set of equations

$$\frac{\partial \sigma_{41}}{\partial t} + \sigma_{41}(\gamma_{41} + i\delta'_a) = i(\alpha' n_{14} + \beta' \sigma_{21}), \quad (5.1a)$$

$$\frac{\partial \sigma_{42}}{\partial t} + \sigma_{42}(\gamma_{42} + i\delta'_b) = i(\beta' n_{24} + \alpha' \sigma_{21}^*), \quad (5.1b)$$

$$\frac{\partial \sigma_{21}}{\partial t} + \sigma_{21}[\gamma_2 + i(\delta_a - \delta_b)] = i[(\beta')^* \sigma_{41} - \alpha' \sigma_{42}^*], \quad (5.1c)$$

$$\frac{\partial \rho_{11}}{\partial t} - R_1 = i[(\alpha')^* \sigma_{41} - \text{c.c.}], \quad (5.1d)$$

$$\frac{\partial \rho_{22}}{\partial t} - R_2 = i[(\beta')^* \sigma_{42} - \text{c.c.}], \quad (5.1e)$$

$$\frac{\partial \rho_{33}}{\partial t} - R_3 = 0, \quad (5.1f)$$

$$\frac{\partial \rho_{44}}{\partial t} - R_4 = i[(\alpha') \sigma_{41}^* + \beta' \sigma_{42}^* - \text{c.c.}], \quad (5.1g)$$

$$\rho_{11} + \rho_{22} + \rho_{33} + \rho_{44} = 1. \quad (5.1h)$$

We shall take for simplicity $\rho_{11}^{(0)} = \rho_{22}^{(0)}$, $\gamma_{41} = \gamma_{42} \equiv \gamma'$, equal pump amplitudes $|\alpha'| = |\beta'|$ which will be resonant with the optical transitions $\delta'_a = \omega_{41} - \omega'_a = \delta'_b = \omega_{42} - \omega'_b = 0$. As a consequence, we have $\rho_{11} = \rho_{22}$ and the material equations Eqs. (5.1d)–(5.1h) for level populations can be reduced to a pair of equations for $n = n_{13} = n_{23}$ and $n' = n_{14} = n_{24}$. Furthermore, we also eliminate adiabatically the atomic polarizations at the optical transitions

$$\begin{aligned} \sigma_{41} &= i(\alpha' n_{14} + \beta' \sigma_{21}) / \gamma', \\ \sigma_{42} &= i(\beta' n_{24} + \alpha' \sigma_{21}^*) / \gamma'. \end{aligned} \quad (5.2)$$

All this leads to

$$\frac{\partial \sigma_{21}}{\partial t} + \sigma_{21}(\gamma_2 + 2|\alpha'|^2 / \gamma') = -(2/\gamma') n' |\alpha'|^2 e^{i(\varphi_{\alpha'} - \varphi_{\beta'})}, \quad (5.3a)$$

$$\frac{\partial n}{\partial t} = \alpha n + b' n' + e - 2(n' + \sigma \cos \Psi) |\alpha'|^2 / \gamma', \quad (5.3b)$$

$$\frac{\partial n'}{\partial t} = a' n' + b n + e' - 6(n' + \sigma \cos \Psi) |\alpha'|^2 / \gamma', \quad (5.3c)$$

where

$$\begin{aligned}
\sigma_{21} &= \sigma e^{i\theta}, \\
\alpha' &= |\alpha'| e^{i\varphi_{\alpha'}}, \\
\beta' &= |\alpha'| e^{i\varphi_{\beta'}}, \\
\Psi &= \theta - (\varphi_{\alpha'} - \varphi_{\beta'}).
\end{aligned} \tag{5.4}$$

Using the equalities

$$\begin{aligned}
\rho_{11} &= \rho_{22} = (1 + n + n')/4, \\
\rho_{33} &= (1 - 3n + n')/4, \\
\rho_{44} &= (1 - 3n' + n)/4,
\end{aligned} \tag{5.5}$$

the coefficients a, b, e, a', b' , and e' can be written as

$$\begin{aligned}
4a &= -3\gamma_3(1 + n_0 + n_{34}^{(0)}) + \gamma_4(n_0 + n_{34}^{(0)}) \\
&\quad - \gamma_{33}(1 - 2n_0 - 2n_{34}^{(0)}), \\
4b &= -\gamma_3(1 + n_0 + n_{34}^{(0)}) + 3\gamma_4(n_0 + n_{34}^{(0)}) \\
&\quad + \gamma_{33}(1 - 2n_0 - 2n_{34}^{(0)}), \\
4e &= 3\gamma_3 n_0 + \gamma_4(n_0 + n_{34}^{(0)}) - \gamma_{33} n_{34}^{(0)}, \\
4a' &= \gamma_3 n_0 - 3\gamma_4(1 + n_0) - \gamma_{33}(1 - 2n_0), \\
4b' &= 3\gamma_3 n_0 - \gamma_4(1 + n_0) + \gamma_{33}(1 - 2n_0), \\
4e' &= \gamma_3 n_0 + 3\gamma_4(n_0 + n_{34}^{(0)}) + \gamma_{33} n_{34}^{(0)}.
\end{aligned} \tag{5.6}$$

In steady state, we have from (5.3a) the relations

$$\sigma = n'x', \quad \cos\Psi = -1, \tag{5.7}$$

where

$$\begin{aligned}
x' &= 2|\alpha'|^2 / (\gamma_2\gamma' + 2|\alpha'|^2) = I' / (I' + I'_c), \\
I' &= c|E'|^2 / 8\pi, \\
I'_c &= c\hbar^2\gamma_2\gamma' / 4\pi(\mu')^2.
\end{aligned}$$

Substituting (5.7) into Eqs. (5.3b) and (5.3c), we obtain in steady state

$$\begin{aligned}
na + n'(b' - d') &= -e, \\
nb + n'(a' - 3d') &= -e',
\end{aligned} \tag{5.8}$$

where $d' = x'\gamma_2$. The solution of this system is

$$\begin{aligned}
n' &= n'_0 / (1 + Ax'), \\
n &= (n_0 + Bx') / (1 + Ax'),
\end{aligned} \tag{5.9}$$

where

$$\begin{aligned}
A &= \gamma_2 T [2(1 + n'_0)\gamma_3 + (1 - 2n'_0)\gamma_{33}], \\
B &= \gamma_2 T (2\gamma_3 n_0 - \gamma_{33} n_{34}^{(0)}), \\
1/T &= 2\gamma_3\gamma_4\rho_{11}^{(0)} + \gamma_3\gamma_{33}\rho_{33}^{(0)} + \gamma_4\gamma_{33}\rho_{44}^{(0)}.
\end{aligned}$$

We note that if the conditions $A \ll 1$ and $B \ll 1$ are fulfilled, we have $n' \simeq n'_0$ and $n \simeq n_0$ for any intensity. One possible realization of these conditions occurs if the upper levels 3 and 4 are nearly empty, or more precisely, if

$$\rho_{33}^{(0)} \text{ and } \rho_{44}^{(0)} \ll \min[1, \gamma_3/\gamma_{33}, \gamma_4/\gamma_{33}]. \tag{5.10}$$

In that case we have $T \simeq 1/\gamma_3\gamma_4$, $A \simeq \gamma_2/3\gamma_4$, $B \simeq \gamma_2/\gamma_4$, and the conditions $A \ll 1$ and $B \ll 1$ become $\gamma_2 \ll \gamma_4$. This is precisely the usual condition of population trapping in a three-level system, with levels 1, 2, and 4 involved in the Λ configuration. In fact, we always have $n' \simeq n'_0$, $n \simeq n_0$ if the LF coherence relaxation rate γ_2 is sufficiently small. As a result, there is coherent population trapping in the double- Λ scheme and both upper levels 3 and 4 can even remain empty despite the strong field action.

Using the explicit expressions (5.7) for the LF coherence and (5.9) for the population differences, we can study the instability condition (2.13) more rigorously than in Sec. IV. The instability condition becomes now

$$(I'/I'_c)[n_{34}^{(0)} - T\gamma_2(2n_0\gamma_3 - n_{34}^{(0)}\gamma_{33})] > n_0. \tag{5.11}$$

A necessary condition for the instability to occur is that the left-hand side of (5.11) be positive, i.e.,

$$n_{34}^{(0)} > n_c \equiv 2n_0\gamma_3\gamma_2 T / (1 + \gamma_2\gamma_{33}T). \tag{5.12}$$

The condition $n_{34}^{(0)} > 0$, which was obtained in our qualitative analysis in Sec. IV, is included in (5.12). However, we now have a stronger condition since (5.12) implies that $n_{34}^{(0)}$ must exceed a lower nonzero bound which depends on the decay rates and the populations in the absence of the fields. In the limit defined by (5.10) and $\gamma_2 \ll \gamma_4$, the result (5.12) takes the form $n_{34}^{(0)} > \gamma_2/\gamma_4$, which is indeed easily fulfilled. Thus the instability condition also implies population trapping ($n' \simeq n'_0$ and $n \simeq n_0$). If $n_{34}^{(0)}$ largely exceeds n_c , the instability condition (5.11) becomes

$$p(z) > 1, \quad p(z) \equiv I'n_{34}^{(0)} / I'_c n_0. \tag{5.13}$$

Now let us find the laws of pump and amplification field propagation taking into account the depletion of the pump due to the resonant absorption. Using the adiabatic approximations (2.14) and (5.2) for the atomic polarizations, we obtain from the field equations (4.4)

$$\begin{aligned}
\frac{\partial\alpha'}{\partial z} + (c'_a)^{-1} \frac{\partial\alpha'}{\partial t} &= -h'_a(\beta'\sigma_{21} + \alpha'n_{14}), \\
\frac{\partial\beta'}{\partial z} + (c'_b)^{-1} \frac{\partial\beta'}{\partial t} &= -h'_b(\alpha'\sigma_{21}^* + \beta'n_{24}), \\
\frac{\partial\alpha}{\partial z} + c_a^{-1} \frac{\partial\alpha}{\partial t} &= -h_a(\beta\sigma_{21} + \alpha n_{13}), \\
\frac{\partial\beta}{\partial z} + c_b^{-1} \frac{\partial\beta}{\partial t} &= -h_b(\alpha\sigma_{21}^* + \beta n_{23}),
\end{aligned} \tag{5.14}$$

in a lossless medium $\kappa_a = \kappa_b = \kappa'_a = \kappa'_b = 0$ and with the definitions

$$\begin{aligned}
h'_a &= 2\pi\omega'_a |\mu_{41}|^2 N / [\epsilon'_a c'_a \hbar (\gamma_{41} + i\delta'_a)], \\
h'_b &= 2\pi\omega'_b |\mu_{42}|^2 N / [\epsilon'_b c'_b \hbar (\gamma_{42} + i\delta'_b)].
\end{aligned}$$

Let us introduce $\alpha' = |\alpha'| e^{i\varphi_{\alpha'}}$, $\beta' = |\beta'| e^{i\varphi_{\beta'}}$, and consider the symmetrical case

$$h_a = h_b = h, \quad h'_a = h'_b = h', \\ c_a = c_b = c, \quad c'_a = c'_b = c'.$$

Furthermore, we shall solve (5.14) with the following conditions on the medium boundary where the input fields are injected:

$$|\alpha(0)| = |\beta(0)|, \quad |\alpha'(0)| = |\beta'(0)|, \\ \varphi_\alpha(0) - \varphi_\beta(0) = \varphi'_\alpha(0) - \varphi'_\beta(0).$$

Then it is easy to verify that Eqs. (5.14) preserve these properties and that there is a solution such that for any space-time point (x, t) one has

$$|\alpha| = |\beta|, \quad |\alpha'| = |\beta'|, \quad \varphi_\alpha - \varphi_\beta = \varphi'_\alpha - \varphi'_\beta.$$

With this property, we obtain from (5.14) the following equations for the intensities of the pump and amplified fields:

$$\frac{\partial I'}{\partial z} + (c')^{-1} \frac{\partial I'}{\partial t} = -2h'I'(n' - \sigma), \\ \frac{\partial I}{\partial z} + c^{-1} \frac{\partial I}{\partial t} = -2hI(n - \sigma). \quad (5.15)$$

For the stationary regime of amplification after substituting σ , n , and n' from (5.7) and (5.9), we have

$$\frac{dI'}{dz} = -2h'I'n'_0/[1 + (I'/I'_c)(1 + A)], \quad (5.16a) \\ \frac{dI}{dz} = \frac{-2hIn_0[1 + (I'/I'_c)(1 + B/n_0 - n'_0/n_0)]}{[1 + (I'/I'_c)(1 + A)]}. \quad (5.16b)$$

The integration of Eq. (5.16a) gives the law of bichromatic pump depletion in a four-level medium. It is the same result as (3.6), if we replace I_s by $3I'_c/A$. So this is a similar law as in a three-level medium except that the saturation intensity is modified due to the presence of the fourth level.

The amplification ($dI/dz > 0$) takes place when the expression inside the square brackets of the numerator on the right side of (5.16b) is negative. It is not difficult to see that this condition gives the inequality (5.11).

We have shown in the discussion of the solutions for n and n' derived from (5.9) that in the case of slow relaxation of LF coherence (i.e., small γ_2), we can neglect the dependence of n and n' on the pump intensity. In this approximation, the set of eqs. (5.16) is simplified:

$$\frac{dI'}{dz} = -2h'n'I'_c/(I' + I'_c), \quad (5.17a)$$

$$\frac{dI}{dz} = 2hn_{34}I - 2h'n'I'_cI/(I' + I'_c). \quad (5.17b)$$

The amplification condition takes the form (5.13). So we must have at least $p(0) > 1$ while amplification takes place as long as $p(z) > 1$. If the length of the amplifying layer is large enough, the amplification will be changed by the absorption, because the pump is depleted due to the resonant absorption, and the condition (5.13) becomes invalid.

The law of pump propagation (5.17a) leads to the result

$$2 \int_0^z h'n'dz = x'_0 - x' - \ln(x'/x'_0), \quad (5.18)$$

where $x' = I'/I'_c$ and $x'_0 = I'_0/I'_c$. Using Eq. (5.17a), we can write Eq. (5.17b) in the form

$$\frac{d}{dz} \ln(I/I'_c) = 2hn_{34} + (h/h') \frac{d}{dz} \ln(I'/I'_c). \quad (5.19)$$

The integration of this equation gives the law of amplified field propagation:

$$I/I(0) = (x'/x'_0)^{hn/h'n'} \exp[\beta(x'_0 - x')], \quad (5.20) \\ \beta = hn_{34}/(h'n').$$

The length l_0 of the layer for which the amplification is maximum is given by the equality $p(l_0) = 1$. For $z > l_0$ the pump depletion becomes significant and the intensity is reduced. In particular, for $p(0) \gg 1$, we have $l_0 \simeq l_c$. This means that the amplification takes place at the length of coherent bleaching.

VI. STEADY STATE OF THE DOUBLE- Λ SYSTEM

When the fields E_a and E_b become large enough as a result of their amplification, they will influence the state of the medium. Then it is no longer possible to consider the medium as defined only by the pump fields. We must use the complete set of equations (4.5) to describe the behavior of our double- Λ system driven by a pair of bichromatic fields.

To simplify somewhat the calculation, we adiabatically eliminate the atomic polarizations. Furthermore, we restrict our analysis to the resonant symmetric case defined by

$$\omega_a = \omega_{31}, \quad \omega_b = \omega_{32}, \quad \omega'_a = \omega_{41}, \quad \omega'_b = \omega_{42}, \\ \gamma_{31} = \gamma_{32} = \gamma, \quad \gamma_{41} = \gamma_{42} = \gamma', \\ \rho_{11}^{(0)} = \rho_{22}^{(0)}, \quad |\alpha| = |\beta|, \quad |\alpha'| = |\beta'|, \\ \varphi_\alpha - \varphi_\beta = \varphi_{\alpha'} - \varphi_{\beta'}.$$

In this case $n_{12}(t) = 0$ for all times and $n_{13} = n_{23} = n$, $n_{14} = n_{24} = n'$, where

$$\frac{d\sigma_{21}}{dt} + \sigma_{21}(\gamma_2 + 2|\alpha|^2/\gamma + 2|\alpha'|^2/\gamma') \\ = -2\alpha\beta^*n/\gamma - 2\alpha'(\beta')^*n'/\gamma', \quad (6.1a)$$

$$\frac{dn}{dt} = an + b'n' + e - 2 \operatorname{Re}[2\alpha(n\alpha^* + \sigma_{21}^*\beta^*)/\gamma \\ + \beta(n\beta^* + \sigma_{21}\alpha^*)/\gamma + (\alpha')^*(n'\alpha' + \sigma_{21}\beta')/\gamma'], \quad (6.1b)$$

$$\frac{dn'}{dt} = a'n' + bn + e' \\ - 2 \operatorname{Re}\{2\alpha'[n'(\alpha')^* + \sigma_{21}^*(\beta')^*]/\gamma' \\ + \beta'[n'(\beta')^* + \sigma_{21}(\alpha')^*]/\gamma' \\ + \alpha^*(n\alpha + \sigma_{21}\beta)/\gamma\}, \quad (6.1c)$$

using the definitions (5.6).

In steady state, Eqs. (6.1) give

$$\sigma = -(\bar{x}n + \bar{x}'n')\cos\Psi, \quad \cos\Psi = \pm 1 \quad (6.2)$$

and two equations for the population differences n and n' :

$$\begin{aligned} n[a + 3F(\bar{x} - 1) + F'\bar{x}] \\ + n'[b' + 3F\bar{x}' + F'(\bar{x}' - 1)] &= -e, \\ n[b + F(\bar{x} - 1) + 3F'\bar{x}] \\ + n'[a' + 3F'(\bar{x}' - 1) + F\bar{x}'] &= -e', \end{aligned} \quad (6.3)$$

where

$$\begin{aligned} \bar{x} &= \bar{I}/(1 + \bar{I} + \bar{I}'), \quad \bar{x}' = \bar{I}'/(1 + \bar{I} + \bar{I}'), \\ \bar{I} &= I/I_c, \quad \bar{I}' = I'/I'_c, \quad F = \bar{I}\gamma_2, \quad F' = \bar{I}'\gamma_2. \end{aligned}$$

The solution of Eqs. (6.3) is

$$\begin{aligned} n_{34} &\equiv n' - n = [n_{34}^{(0)} + (f' - f)]/(1 + D) \\ &= \{n_{34}^{(0)} + \gamma_2 T [2n'_0 \gamma_4 \bar{I} - 2n_0 \gamma_3 \bar{I}' + n_{34}^{(0)} \gamma_{33} (\bar{I} + \bar{I}')] / (1 + \bar{I} + \bar{I}')\} / (1 + D), \end{aligned} \quad (6.5)$$

as well as the sum $n' + n$,

$$n' + n = [n_0 + n'_0 + 2T\gamma_2(n_0\gamma_3 + n'_0\gamma_4)(\bar{I} + \bar{I}' + 2\bar{I}\bar{I}') / (1 + \bar{I} + \bar{I}')] / (1 + D). \quad (6.6)$$

Using the normalization condition $\rho_{11} + \rho_{22} + \rho_{33} + \rho_{44} = 1$, the sum of the upper level populations is obtained:

$$\rho_{33} + \rho_{44} = [1 - (n + n')] / 2.$$

When both fields I and I' become intense, the population differences n and n' become equal:

$$\begin{aligned} n \rightarrow n' \rightarrow \Delta N \\ = (n_0\gamma_3 + n'_0\gamma_4) / [4\gamma_2 + (1 + n_{34}^{(0)})\gamma_3 + (1 - n_{34}^{(0)})\gamma_4]. \end{aligned} \quad (6.7)$$

This implies that $n + n' \rightarrow 2\Delta N$ and that the population difference n_{34} tends to zero.

For the case of slow LF coherence relaxation $\gamma_2 \ll \gamma_3, \gamma_4$ and equal longitudinal relaxation times $\gamma_3 = \gamma_4$, we have from (6.6) the invariance relation

$$n + n' = n_0 + n'_0.$$

The same result holds if $\gamma_2 \ll \gamma_3, \gamma_4$ and $n_{34}^{(0)} \ll 1$. This means that the strong field equalizes the populations of the upper levels 3 and 4, but does not equalize the populations at any optical transitions. In particular, if the upper levels were empty before the field action, they will stay almost empty in spite of the strong field action. This means that fluorescence from the medium and field absorption remain approximately zero under the coherent action of a pair of bichromatic fields. These properties will be restored if the synchronization condition

$$\omega_a - \omega_b = \omega'_a - \omega'_b = \omega_{21}, \quad \varphi_a - \varphi_b = \varphi'_a - \varphi'_b$$

is not verified. This phenomenon in the double- Λ scheme

$$n = (n_0 + f)/(1 + D), \quad n' = (n'_0 + f')/(1 + D), \quad (6.4a)$$

$$\begin{aligned} D &= \frac{\gamma_2 T}{1 + \bar{I} + \bar{I}'} \\ &\times \{2\bar{I}\bar{I}'[4\gamma_2 + (1 + n_{34}^{(0)})\gamma_3 + (1 - n_{34}^{(0)})\gamma_4] \\ &+ \bar{I}[2(1 + n_0)\gamma_3 + (1 - 2n_0)\gamma_{33}] \\ &+ \bar{I}'[2(1 + n'_0)\gamma_3 + (1 - 2n'_0)\gamma_{33}]\}, \end{aligned} \quad (6.4b)$$

$$f = T\gamma_2\bar{x}'[2\bar{I}(n_0\gamma_3 + n'_0\gamma_4) + 2n_0\gamma_3 - n_{34}^{(0)}\gamma_{33}], \quad (6.4c)$$

$$f' = T\gamma_2\bar{x}[2\bar{I}'(n_0\gamma_3 + n'_0\gamma_4) + 2n'_0\gamma_4 + n_{34}^{(0)}\gamma_{33}]. \quad (6.4d)$$

When $\alpha = 0$, the results (6.4) are identical to (5.9) derived when there is only one bichromatic field. For the phase angle Ψ the only possible choice is $\Psi = \pi$ since $xn + x'n'$ is positive and σ has to be positive as well.

From (6.4a), we can find the population difference n_{34} ,

is quite similar to the well-known phenomenon of coherent population trapping in the simple Λ scheme.¹⁻⁶

VII. AMPLIFICATION WITHOUT INVERSION: NONLINEAR REGIME

Let us investigate the nonlinear regime of amplification for the symmetric case studied in Sec. V, when there is a steady regime of propagation with $|\alpha| = |\beta|$, $|\alpha'| = |\beta'|$, $\varphi_\alpha - \varphi_\beta = \varphi'_\alpha - \varphi'_\beta$. It is described by the intensity equations (5.15), with the atomic variables σ , n , and n' defined by the solutions (6.2) and (6.4), which characterize the behavior of the double- Λ system under the action of two bichromatic fields.

In the general case, the analysis of the pair of equations (5.15) is quite complicated. Let us therefore introduce one additional simplifying assumption. We shall neglect the dependence of the population differences on the intensity ($n \simeq n_0$, $n' \simeq n'_0$, $n_{34} \simeq n_{34}^{(0)}$), keeping only the dependence of σ on I and I' . This is justified when $\gamma_2 \ll \gamma_3, \gamma_4$ (i.e., $I_c \ll I_s$, $I'_c \ll I'_s$) because in that limit coherence bleaching occurs without saturation bleaching, which implies that the nonlinearity connected with the LF coherence creation is much stronger than the nonlinearity connected with the population equalization. Using the steady-state solutions (6.4) for n and n' , and the steady-state solution (6.5) for n_{34} , we can express this approximation more explicitly in the form

$$f \ll n_0, \quad f' \ll n'_0, \quad f' - f \ll n_{34}^{(0)}, \quad D \ll 1. \quad (7.1)$$

After substituting $n = n_0$, $n' = n'_0$, and σ_{21} from Eq. (6.2)

into the intensity equations (5.15), we obtain the non-linear steady-state equations

$$\frac{d\tilde{I}}{dz} = -2\tilde{I}h[n_0 - (n_0\tilde{I} + n'_0\tilde{I}')/(1 + \tilde{I} + \tilde{I}')], \quad (7.2)$$

$$\frac{d\tilde{I}'}{dz} = -2\tilde{I}'h'[n'_0 - (n_0\tilde{I} + n'_0\tilde{I}')/(1 + \tilde{I} + \tilde{I}')].$$

First, we consider the limit $\tilde{I} + \tilde{I}' \gg 1$ or equivalently $I'_c + I'_c \gg I_c I'_c$. This condition will be fulfilled even at low intensities if the relaxation rate of the LF coherence tends to zero ($\gamma_2 \rightarrow 0$), i.e., if both I_c and $I'_c \rightarrow 0$. Then (7.2) takes the form

$$\frac{d\tilde{I}}{dz} = 2hn_{34}^{(0)}\tilde{I}\tilde{I}'/(\tilde{I} + \tilde{I}'), \quad (7.3a)$$

$$\frac{d\tilde{I}'}{dz} = -2h'n_{34}^{(0)}\tilde{I}\tilde{I}'/(\tilde{I} + \tilde{I}'). \quad (7.3b)$$

From these equations we derive the conservation law

$$\tilde{I}/h + \tilde{I}'/h' = \text{const}, \quad (7.4)$$

which can also be written as

$$I/\omega + I'/\omega' = m = \text{const}. \quad (7.5)$$

This last relation expresses the conservation of the photon number m . In particular, if $I(0) \simeq 0$ and in the case of long medium ($z \rightarrow \infty$) with inversion between the two upper levels ($n_{34}^{(0)} > 0$), we have from (7.3b) that $I'(\infty) \rightarrow 0$ and the photon conservation law (7.5) yields $I(\infty) = I'(0)\omega/\omega'$. For the up-conversion process ($\omega/\omega' > 1$), the output radiation intensity $I(\infty)$ is larger than the input intensity $I'(0)$ by a factor ω/ω' . Hence the energy extracted from the medium is $I(\infty) - I'(0) = m(\omega - \omega')$. On the contrary, in the absence of population inversion, only the downward conversion process is possible and a fraction $m(\omega' - \omega)$ of the energy is transferred to the medium. In this sense, there is total analogy with the Raman process, where the transfer of radiation into the anti-Stokes component is possible only in a medium with population inversion at LF transition, while energy transfer into the Stokes component takes place in the absence of the inversion. From the intensity steady-state equations (7.3), we find the law of pump and amplified fields propagation:

$$Y = Y_0 [1 - (Y - Y_0)h'/h]^{h/h'} \exp \left[\int_0^z 2hn_{34}^{(0)} dz \right], \quad (7.6a)$$

$$X = Y_0^{(-h'/h)} [Y_0 - (X - 1)h'/h]^{h'/h} \exp \left[- \int_0^z 2h'n_{34}^{(0)} dz \right], \quad (7.6b)$$

where

$$Y = \tilde{I}/\tilde{I}'(0),$$

$$X = \tilde{I}'/\tilde{I}'(0),$$

$$Y_0 = \tilde{I}(0)/\tilde{I}'(0).$$

In particular, for $h = h'$, we have

$$Y = \frac{Y_0(1 + Y_0) \exp \left[\int_0^z 2hn_{34}^{(0)} dz \right]}{\left[1 + Y_0 \exp \left[\int_0^z 2hn_{34}^{(0)} dz \right] \right]}, \quad (7.7a)$$

$$X = \frac{(1 + Y_0) \exp \left[- \int_0^z 2hn_{34}^{(0)} dz \right]}{\left[Y_0 + \exp \left[- \int_0^z 2hn_{34}^{(0)} dz \right] \right]}. \quad (7.7b)$$

We now perform a similar analysis on the more general steady-state equations for the intensity, Eqs. (7.2), which can be rewritten as

$$\frac{d\tilde{I}}{dz} = 2h\tilde{I}(\tilde{I}'n_{34}^{(0)} - n_0)/(1 + \tilde{I} + \tilde{I}'), \quad (7.8a)$$

$$\frac{d\tilde{I}'}{dz} = -2h'\tilde{I}'(\tilde{I}n_{34}^{(0)} + n'_0)/(1 + \tilde{I} + \tilde{I}'), \quad (7.8b)$$

from which we derive

$$\frac{d}{dz} (\tilde{I}/h + \tilde{I}'/h') = -2(\tilde{I}n_0 + \tilde{I}'n'_0)/(1 + \tilde{I} + \tilde{I}'). \quad (7.8c)$$

According to (7.8c), the fields lose energy during their propagation due to the absorption of radiation by the resonant transitions; this energy is then dissipated into the medium.

Let us divide Eq. (7.8a) by Eq. (7.8b). Then we have

$$\frac{d\tilde{I}}{d\tilde{I}'} = h\tilde{I}(n_0 - n_{34}^{(0)}\tilde{I}')/h'\tilde{I}'(n'_0 + n_{34}^{(0)}\tilde{I}). \quad (7.9)$$

First of all, let us consider the particular case $n_{34}^{(0)} = 0$ so that amplification is impossible. Then the solution of (7.9) is

$$\tilde{I}/\tilde{I}'(0) = [\tilde{I}'/\tilde{I}'(0)]^{h/h'}. \quad (7.10)$$

In particular, when $h = h'$, the law of energy dissipation has the same form for both bichromatic fields: $\tilde{I}/\tilde{I}'(0) = \tilde{I}'/\tilde{I}'(0)$. An implicit equation for $\tilde{I}/\tilde{I}'(0)$ is easily obtained by combining (7.8a) with the equality $\tilde{I}/\tilde{I}'(0) = \tilde{I}'/\tilde{I}'(0)$:

$$2 \int_0^z hn_0 dz = [1 - \tilde{I}/\tilde{I}'(0)][\tilde{I}(0) + \tilde{I}'(0)] - \ln[\tilde{I}/\tilde{I}'(0)]. \quad (7.11)$$

When only one bichromatic field is sent in the medium, the result (7.11) reduces to (3.7). With two bichromatic fields, however, the radiation extinction length is derived from (7.11) and is

$$2hn_0 l_c = 1 + [\tilde{I}(0) + \tilde{I}'(0)](1 - 1/e). \quad (7.12)$$

Under the condition $\tilde{I}(0) + \tilde{I}'(0) \gg 1$, l_c is increased by a factor $\tilde{I}(0) + \tilde{I}'(0)$ compared to the case of linear propagation. It should be emphasized that it is enough to have only one strong bichromatic input field for the coherent bleaching of the medium at both optical transitions.

In the general case, i.e., for arbitrary $n_{34}^{(0)}$, the integration of Eq. (7.9) gives

$$Z = -\ln Z + Q(X), \quad (7.13)$$

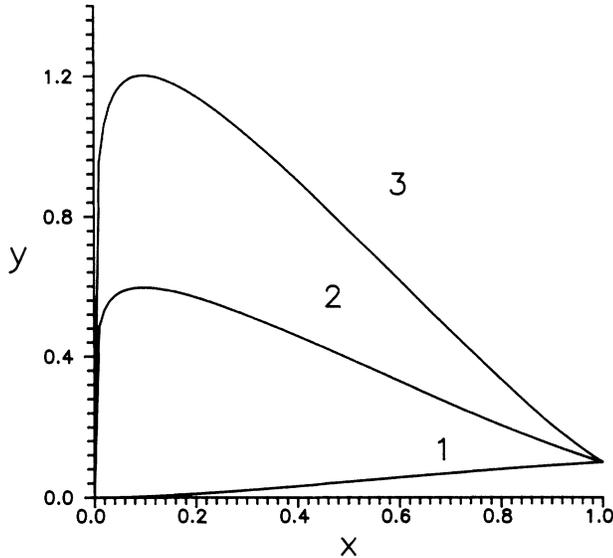


FIG. 3. The scaled intensity of the amplified field Y as a function of the scaled intensity of the pump field X defined by Eq. (7.13). The common parameters are $Y_0=0.1$, $n_{34}^{(0)}/n_0'=0.025$. For curve (1), $p(0)=0.5$, $h/h'=2$; for curve (2), $p(0)=10$, $h/h'=1$; for curve (3), $p(0)=10$, $h/h'=2$.

where

$$Z = Yp(0), \quad p(0) = I'(0)n_{34}^{(0)}/I'_c n_0,$$

$$Q(X) = Y_0 p(0) + \ln[Y_0 p(0)]$$

$$+ [(1-X)p(0) + (\ln X)n_0/n_0']h/h'.$$

From (7.13), we have the obvious limiting situations

$$Z \approx \begin{cases} Q(X) - \ln Q(X) & \text{for } Q(X) \gg 1 \\ \{1 + \exp[-Q(X)]\}^{-1} & \text{for } Q(X) \ll 1. \end{cases} \quad (7.14)$$

The solution of Eq. (7.13) is plotted in Fig. 3 for various values of the parameters $p(0)$ and h/h' . When the amplification condition $p(0) > 1$ is not fulfilled, both fields are absorbed as shown on curve 1 in agreement with Eqs. (7.8). When the condition $p(0) > 1$ is fulfilled, the intensity I increases following (7.8a), until the condition $p(z) > 1$ is no longer satisfied due to pump depletion (curves 2 and 3). It should be emphasized that, for the up-conversion process ($\omega > \omega'$), the maximum value of the amplified field exceeds the input pump intensity (curve 3). This is made possible due to the extraction of the energy from the medium, as already discussed.

VIII. CONCLUSIONS

The main physical results of this paper are the following.

(1) A bichromatic field can be amplified in a three-level medium without population inversion, if the upper level is partially occupied ($\rho_{33}^{(0)} \neq 0$) and a large enough LF coherence is excited by external sources. The physical mechanism of this amplification without inversion is the parametric instability of two optical waves contributing

with opposite signs to the total energy.

(2) In the case of a three-level system in the Λ configuration of Fig. 1, it is impossible to extract energy stored in the upper atomic level using only the self-consistent propagation of bichromatic fields in the absence of population inversion. But the LF coherence excited by this process induces a decrease in the field absorption which leads to coherent bleaching of the medium.

(3) There is a coherent population trapping in the double- Λ configuration under the action of either one or two bichromatic fields when the LF coherence relaxation time is large enough. Specifically, if the upper levels are initially empty ($\rho_{33}^{(0)} = \rho_{44}^{(0)} = 0$), they remain practically empty in spite of the strong field action. Therefore, field absorption and fluorescence are absent. In the general case ($\rho_{33}^{(0)} \neq \rho_{44}^{(0)} \neq 0$), the atomic response under the action of two bichromatic fields is reduced to the upper-level population redistribution only, while the lower level populations remain practically unchanged.

(4) There is a stationary regime of resonant ($\omega_a = \omega_{31}$, $\omega_b = \omega_{32}$) optical bichromatic field amplification during its propagation in a four-level medium with the double- Λ configuration under the action of a bichromatic pump resonant to the adjacent transitions ($\omega'_a = \omega_{41}$, $\omega'_b = \omega_{42}$). It should be emphasized that the process goes without population inversion at any of the optical transitions. It is accompanied by an increase of the frequencies ($\omega_a > \omega'_a$, $\omega_b > \omega'_b$) if there is a population inversion between the two upper levels ($n_{34}^{(0)} > 0$). The intensity of the amplified output radiation can exceed significantly the input pump intensity due to the extraction of the energy from the medium stored in the upper atomic level.

The suggested double Λ scheme can be realized, for example, in sodium vapor. The D_1 (589.6-nm) and D_2 (589-nm) optical lines can be used as operating and adjacent transitions. The pumping can be achieved by a dye laser. The excitation of LF coherence at the hyperfine splitting of the ground state (1.77 GHz) is possible. Let us make an estimation of the available gain without inversion and the pump intensity which is necessary to satisfy the amplification condition. We take for the estimation the following values of the sodium vapor parameters:

$$\mu^2 \sim (\mu')^2 \sim 5 \times 10^{-35} \text{ CGSE}, \quad \gamma_3^{-1} \sim \gamma_4^{-1} \sim 10 \text{ ns},$$

$$\gamma^{-1} \sim (\gamma')^{-1} \sim 1 \text{ ns}, \quad \gamma_{33}^{-1} \sim 1 \text{ ms}, \quad \gamma_2^{-1} \sim 1 \mu\text{s},$$

$$n_0 \sim n_0' \sim \frac{1}{2}, \quad N \sim 10^{11} \text{ cm}^{-3}.$$

In an equilibrium state, the population difference $n_{34}^{(0)}$ is very small:

$$n_{34}^{(0)} \approx (\hbar\omega_{43}/kT)\exp(-\hbar\omega_{31}/kT).$$

It can be increased, for example, by incoherent optical pumping. Let us take for the estimation $n_{34}^{(0)} \sim 10^{-2}$. Then, from (4.3), we have $G \approx 0.03 \text{ cm}^{-1}$. According to this estimation, the gain is not too high because of the small population difference between levels 3 and 4. Apparently, it is easier to obtain inversionless generation

than amplification. From the threshold condition $e^{GL}R > 1$, we can obtain the estimation of the reflection coefficient. At $L \sim 3$ cm, we have $R \geq 0.9$. It is not difficult to see that the amplification condition (5.11) is fulfilled in our case if the input pump intensity exceeds 5×10^{-2} W/cm².

The most obvious application of inversionless amplification, as was stressed earlier,¹⁸⁻²⁰ would be to obtain light generation in those very short-wavelength

quantum transitions where population inversion is difficult to achieve, including the x-ray domain.

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