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Gain without inversion: why and how?

PART II

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Chapter 1

Two-level atoms

In this chapter, we review some properties of an electric field E interacting with a collection of N identical two-level atoms in the semiclassical formulation. The emphasis will be on properties which are significantly modified when atomic interference arise.

1.1 Formulation

1.1.1 Electric field equation

Our starting point is Maxwell's equation for the electric field

$$\left(v^2\partial^2_{xx} - \partial^2_{tt}\right) E_{tot} = (4\pi/\varepsilon_0)\partial^2_{tt} P_{tot} \tag{1.1}$$

where P_{tot} is the total atomic polarization induced by the field E_{tot} and v is the velocity of light in the medium. It is assumed that v is constant. We introduce the decompositions

$$E_{tot} = \frac{1}{2}\left[Ee^{i(k_a x - \omega_a t)} + c.c.\right], \qquad P_{tot} = N\left[Pe^{i(k_a x - \omega_a t)} + c.c.\right] \tag{1.2}$$

where the optical frequency ω_a and the wave number k_a are related by the dispersion relation $\omega_a = vk_a$. In the absence of light-matter interaction, a solution of Maxwell's equation is given by an electric field amplitude E which is constant in space and time. This is no longer true, in general, in the presence of light-matter interaction. Therefore, we seek solutions $E(x,t)$ which vary slowly in space and time, compared with the optical space and time variation

$$\omega_a |P| \gg |\partial P/\partial t|, \qquad \omega_a |E| \gg |\partial E/\partial t|, \qquad k_a |E| \gg |\partial E/\partial x| \tag{1.3}$$

This is usually justified by the fact that the residual variation of the complex field amplitude $E(x,t)$ is related to the atomic temporal variations, which occur in a much longer time scale. With these elements, it is easy to derive a propagation equation for the complex field amplitude

$$(v\partial_x + \partial_t)\, E = (4\pi i N\omega_a/\varepsilon_0)P \tag{1.4}$$

The final step in this derivation is to relate the polarization per atom, P, with the microscopic properties of the medium. This is will be done in the next section.

1.1.2 Material equations

The last equation we have obtained for the electric field is still an open equation since we need a closure relation of the form $P = P(E)$. For this, we have to introduce a model of the atomic system in order to express the reaction of the medium to the applied field. The medium is described by the von Neumann equation

$$i\hbar\frac{\partial\rho}{\partial t} = H\rho - \rho H \tag{1.5}$$

where the density operator ρ is constrained by the trace condition $\mathrm{Tr}\rho = 1$. Since we consider two-level atoms, the density operator and the hamiltonian have a 2x2 representation. For the hamiltonian, we use the representation $H = H_0 + V$ where H_0 is the unperturbed hamiltonian of the two-level atoms ($H_0\psi_j = \hbar\omega_j\psi_j$ and we neglect the motion of the center-of-mass) while the interaction energy V is given by $-erE_{tot}$ where $-er$ is the projection of the electric dipole in the direction of the electric field polarization. Let the upper atomic state be labelled 2 and the lower atomic state be labelled 1. The von Neumann equation (1.5) leads to the evolution equation for the density matrix elements

$$\begin{aligned} \frac{\partial\rho_{21}}{\partial t} &= -i\omega_{21}\rho_{21} + \frac{i\mu}{2\hbar}E_{tot}n & (1.6)\\ \frac{\partial n}{\partial t} &= 2\,\mathrm{Im}\left(\frac{\mu}{\hbar}E_{tot}\rho_{21}^*\right) & (1.7) \end{aligned}$$

where we have used the notation $\mu \equiv \mu_{21}$ and $n \equiv \rho_{11} - \rho_{22}$. Note that $\rho_{12} = \rho_{21}^*$. The closure relation we are looking for is the relation $P = \mu_{12}\rho_{21}e^{i\omega_a t} \equiv \mu^*\sigma$ between the atomic polarization and the off-diagonal elements of the density matrix. Neglecting fast oscillating terms, we obtain the coupled equations

$$\begin{aligned} \left(\partial_x + v^{-1}\partial_t\right) E &= (4\pi i N\mu^*\omega_a/v\varepsilon_0)\sigma & (1.8)\\ \frac{\partial\sigma}{\partial t} &= -i\delta\sigma + \frac{i\mu}{2\hbar}En & (1.9)\\ \frac{\partial n}{\partial t} &= 2\,\mathrm{Im}\left(\frac{\mu}{\hbar}E\sigma^*\right) & (1.10) \end{aligned}$$

with $\delta \equiv \omega_{21} - \omega_a$.

1.1.3 Pumping and decay

The description obtained sofar is incomplete. Equations (1.8)-(1.10) correctly describe the interaction process between *stable* atoms and a medium which is *lossless* in the absence of light-matter interaction. However, the atomic levels are not stable. In addition, the material medium accounts only for the quasi-resonant interaction with the field. Apart from that, there are nonresonant interactions (involving the other atomic levels) which yield a linear, i.e., field-independent loss. The usual procedure is to add phenomenological constants to the evolution equations (1.8)-(1.10) in the following way. For the field equation, we add to the right hand side a term κE

$$\left(\partial_x + v^{-1}\partial_t + \kappa\right) E = (4\pi i N\mu^*\omega_a/v\varepsilon_0)\sigma \tag{1.11}$$

In the absence of interaction with the two-level medium, this leads to a solution which decays in time like $\exp(-\kappa t)$. For the atomic polarization, we also add a linear damping term $-\gamma_\perp\sigma$ while the population difference we add the damping $-\gamma_\| n$. However, we have to take into account the incoherent pumping which populates the upper atomic level. Let n^0 be the population difference reached in steady state in the absence of interaction with the coherent field E_{tot}. Then we add a source term to the population inversion evolution equation and arrive at

$$\frac{\partial\sigma}{\partial t} = -(\gamma_\perp + i\delta)\sigma + \frac{i\mu}{2\hbar}En \tag{1.12}$$

$$\frac{\partial n}{\partial t} = \gamma_\|(n^0 - n) + 2\,\mathrm{Im}\left(\frac{\mu}{\hbar}E\sigma^*\right) \tag{1.13}$$

Equations (1.11)-(1.13) form the basis of our study of two-level atoms interacting with a monochromatic electric field.

1.2 Linear propagation regime

In the linear regime of propagation, the problem is reduced to the pair of linear coupled equations

$$\left(\partial_x + v^{-1}\partial_t + \kappa\right) E = (4\pi i N\mu^*\omega_a/v\varepsilon_0)\sigma \tag{1.14}$$

$$\frac{\partial\sigma}{\partial t} = -(\gamma_\perp + i\delta)\sigma + \frac{i\mu}{2\hbar}En \tag{1.15}$$

$$\frac{\partial n}{\partial t} = \gamma_\|(n^0 - n) \tag{1.16}$$

Indeed, retaining the E-dependent term in (1.13) leads to nonlinear corrections to the relation $\sigma \propto E$ which derives from (1.14)-(1.15).

We seek plane wave solutions $E = \mathcal{E}e^{i(kx-\omega t)}$, $\sigma = se^{i(kx-\omega t)}$, and $n = n^0$. This leads to

$$\begin{aligned} \mathcal{E}(ik - i\omega/v + \kappa) &= \frac{4\pi i N\omega_a\mu^*}{\varepsilon_0 v}s & (1.17) \\ -i\omega s &= -(\gamma_\perp + i\delta)s + \frac{i\mu}{2\hbar}\mathcal{E}n^0 & (1.18) \end{aligned}$$

The compatibility condition of this pair of homogeneous equations is that the determinant of the coefficients vanishes

$$(k-\omega/v)(\omega-\delta) + i\gamma_\perp(k-\omega/v) - i\kappa(\omega-\delta) + \kappa\gamma_\perp + \frac{2\pi N\omega_a|\mu|^2 n^0}{\hbar\varepsilon_0 v} = 0 \quad (1.19)$$

This is the dispersion relation $k = k(\omega)$ we were looking for. Its solution is

$$\begin{aligned} k &= \frac{\omega}{v} + \frac{(\omega-\delta)/\gamma_\perp}{1+(\omega-\delta)^2/\gamma_\perp^2}\alpha + i\kappa(1-A) & (1.20) \\ \alpha &= -\frac{2\pi N\omega_a|\mu|^2 n^0}{\hbar\varepsilon_0 v\gamma_\perp\left[1+(\omega-\delta)^2/\gamma_\perp^2\right]} & (1.21) \end{aligned}$$

We have introduced the parameter $A = \alpha/\kappa$ where $\alpha \propto -n^0 = \rho_{22} - \rho_{11}$ is the linear (or small signal) gain or loss, depending on whether it is positive or negative, respectively.

The stability properties of the plane wave are easily derived from (1.20), taking ω real:

- $A < 1$: In this case, $\operatorname{Im} k > 0$ and the plane wave is attenuated in the linear regime.
- $A > 1$: In this case, $\operatorname{Im} k < 0$ and the plane wave is amplified in the linear regime.

Hence the plane wave solution is stable below the threshold $A = 1$ and unstable above the threshold. The threshold condition $A = 1$ is the usual laser threshold condition. The important property is that the gain condition $A > 1$ can be written as a condition for the population inversion as $\rho_{22}^0 - \rho_{11}^0 > (\rho_{22}^0 - \rho_{11}^0)_{threshold} > 0$. In other terms, an initial population inversion is necessary, though not sufficient, to ensure gain in the two-level medium.

We can deduce from (1.18) the linearized eigensolution in the form of the ratio $s/\mathcal{E}$, from which we obtain the expression

$$\frac{P}{E} = \frac{\mu^* s}{\mathcal{E}} = \frac{|\mu|^2 n^0}{2\hbar}\left[\frac{\delta-\omega}{\gamma_\perp^2 + (\delta-\omega)^2} + \frac{i\gamma_\perp}{\gamma_\perp^2+(\delta-\omega)^2}\right] \equiv \chi_0(\omega) \quad (1.22)$$

where we have introduced the linear susceptibility $\chi_0 = \chi_0' + i\chi_0''$. We easily derive the well-known relation

$$\chi_0'(\omega) = \frac{\delta - \omega}{\gamma_\perp} \chi_0''(\omega) \tag{1.23}$$

The imaginary part of the susceptibility rules the absorptive properties of the medium. As a function of $\delta - \omega$, it is lorentzian peaked at $\omega = \delta$. The real part of χ_0 rules the dispersive properties of the medium and vanishes where χ_0' is maximum.

1.3 Nonlinear regime

1.3.1 Steady state

The nonlinear equations (1.11)-(1.13) admit steady state solutions of the form

$$\begin{aligned} dE/dx + \kappa E &= (4\pi i N \mu^* \omega_a / v\varepsilon_0)\sigma & (1.24) \\ \sigma &= \frac{i\mu}{2\hbar} \frac{En}{\gamma_\perp + i\delta} & (1.25) \\ n &= n^0 + 2\,\mathrm{Im}\left(\frac{\mu}{\hbar\gamma_\|} E\sigma^*\right) & (1.26) \end{aligned}$$

From the last two equations we obtain an expression of the material variables in terms of the field intensity

$$\begin{aligned} n &= n^0 \left[1 + \frac{|\mu E|^2}{\hbar^2 \gamma_\perp \gamma_\| [1 + (\delta/\gamma_\perp)^2]}\right]^{-1} & (1.27) \\ \sigma &= E \frac{\mu n^0}{2\hbar\gamma_\perp} \frac{i + \delta/\gamma_\perp}{1 + (\delta/\gamma_\perp)^2} \left[1 + \frac{|\mu E|^2}{\hbar^2 \gamma_\perp \gamma_\| [1 + (\delta/\gamma_\perp)^2]}\right]^{-1} & (1.28) \end{aligned}$$

Note that in the strong field limit, $|E| \to \infty$, we have $n \to 0$ and $\sigma \to 0$. Hence we obtain

$$|E| \to \infty: \qquad \rho_{11} \to 1/2, \qquad \rho_{22} \to 1/2, \qquad \rho_{12} \to 0 \tag{1.29}$$

In other terms, a strong field bleaches the atomic system : $n = \rho_{11} - \rho_{22} \to 0$ (which is independent of n^0) but destroys the atomic coherence : $\rho_{12} \to 0$.

From the definition $P \equiv \chi_s E$, we obtain $\chi_s = \mu^* \sigma / P$ and therefore

$$\chi_s = \chi_s' + i\chi_s'' = \frac{i + \delta/\gamma_\perp}{1 + (\delta/\gamma_\perp)^2} \frac{|\mu|^2 n^0}{2\hbar\gamma_\perp} \left[1 + \frac{|\mu E|^2}{\hbar^2 \gamma_\perp \gamma_\| [1 + (\delta/\gamma_\perp)^2]}\right]^{-1} \tag{1.30}$$

We have the obvious relation between the real and imaginary parts of the susceptibility: $\chi' = (\delta/\gamma_\perp)\chi''$. The connection between this result and the susceptibility given in section 1.2 is quite simple: χ_0 is ω-dependent but field-independent. On the contrary, χ_s is the static or ω-independent (since it is derived in steady state) component of the susceptibility but it is field-dependent. Both expressions represent different approximations of a more general expression which depends on ω and on the field.

1.3.2 Nonlinear propagation

From (1.24) it follows that the complex field amplitude in steady state is given by

$$\frac{dE}{dx} = -\kappa E - \overline{\alpha} E \frac{n}{n^0}(1 - i\delta/\gamma_\perp) \tag{1.31}$$

$$\overline{\alpha} \equiv \overline{\alpha}(x,\delta) = \frac{2\pi N \omega_a |\mu|^2 n^0(x)}{\hbar\gamma_\perp \varepsilon_0 v \left[1 + (\delta/\gamma_\perp)^2\right]} \tag{1.32}$$

We introduce the polar decomposition of the field $E = \overline{E}e^{i\phi}$ in terms of which the electric field real amplitude is the solution of

$$d\overline{E}/dx = -\kappa\overline{E} - \overline{\alpha}\overline{E}n/n^0 \tag{1.33}$$

Let us define an intensity by the relation $I = |\mu E|^2 \left\{\hbar^2\gamma_\perp\gamma_\parallel[1 + (\delta/\gamma_\perp)^2]\right\}^{-1}$. It verifies the equation

$$dI/dx = [-2\kappa - 2\overline{\alpha}/(1 + I)]I \tag{1.34}$$

The difficulty with this equation is the space-dependence of $\overline{\alpha}$. Let us therefore consider the limit of negligible linear loss: $\kappa \ll \overline{\alpha}$. In that limit, we obtain $dI/dx = -2\overline{\alpha}I/(1 + I)$ which can be solved to give the implicit equation

$$I - I_0 + \ln(I/I_0) = -2\int_0^L \overline{\alpha}(x)dx \tag{1.35}$$

Let us define an extinction length l_e by the condition $I(l_e) \equiv I_0/e$. If the initial population difference n^0 is space-independent, we find

$$l_e = [I_0(1 - 1/e) + 1]/(2\overline{\alpha}) \tag{1.36}$$

In the high intensity limit $I_0 \gg 1$, we obtain $l_e \propto I_0/2\overline{\alpha}$.

Chapter 2

Principles of atomic interference

2.1 Physical mechanism

The simplest situation in which atomic interference occurs is the interaction of a three-level medium with a bichromatic field. Let the field be given by

$$E_{tot} = E_a \cos(\omega_a t) + E_b \cos(\omega_b t) \tag{2.1}$$

This electric field interacts with a three-level medium characterized by three atomic levels. The three atomic wave functions are ϕ_j and the corresponding energies are $\hbar\omega_j$. We consider the situation of a single upper level and two closely space low lying states: $\omega_3 \gg \omega_2 \gtrsim \omega_1$. The interaction hamiltonian is $-erE$. The field E_a couples levels 3 and 1 while the field E_b couples levels 3 and 2.

The main point is that an atom in state 3 may decay **either** through the channel 3-1 **or** through the channel 3-2. The transition probability between two states is of the form $P(j \to k) = |C_{jk}|^2$ where the complex coefficient C_{jk} is related to the expansion of the total wave function on the basis of the unperturbed atomic wave functions. When there are two decay channels available to the atom, C_{jk} is the sum of two terms, each coming from one of the decay channels. Hence the total transition probability will not be the sum of the partial transition probabilities. In addition, there will be interference terms which need not be positive. We shall see that these interference terms may compensate, partially or completely, the partial transition probabilities, therefore reducing the total transition probability. This interference process is the basis of most of the work on gain without inversion.

As a practical example, let us consider the three-level system described above, interacting with the bichromatic field (2.1). We still have to specify the selection rules. Let them be given by

$$\langle \phi_j \left| erE_a \right| \phi_3 \rangle = \mu E \delta_{j1} \qquad \langle \phi_j \left| erE_a \right| \phi_3 \rangle = \mu E \delta_{j2} \tag{2.2}$$

all other matrix elements being zero. To simplify the algebra of this example, we have assumed that the matrix elements are identical when they do not vanish.

This simplification is without influence on the physical results we shall describe in this chapter. The probability of transition from state 3 to an arbitrary superposition state $\Phi = C_1\phi_1 + C_2\phi_2$ is given by

$$\begin{aligned} W_1 &= \left|\left\langle C_1\phi_1 e^{-i\omega_1 t} + C_2\phi_2 e^{-i\omega_2 t}\left|-erE\right| e^{-i\omega_3 t}\phi_3\right\rangle\right|^2 \\ &= \frac{|\mu E|^2}{4}\left|C_1 e^{i(\omega_{31}-\omega_a)t} + C_2 e^{i(\omega_{32}-\omega_b)t}\right|^2 \end{aligned} \tag{2.3}$$

where $\omega_{jk} \equiv \omega_j - \omega_k$. The case $\omega_{31} - \omega_a = \omega_{32} - \omega_b$ is of special interest. If this condition is verified, that is, if the detuning between the field E_a and the atomic energy difference ω_{31} equals the detuning between the field E_b and the atomic difference ω_{32}, then the transition probability W_1 becomes a constant in time

$$W_1 = \frac{1}{4}\left|\mu E(C_1 + C_2)\right|^2 \tag{2.4}$$

To appreciate more fully this result, let us consider another possibility of selection rules. Let us assume that each field E_j couples equally the upper level 3 to the two lower states

$$\langle\phi_j\left|erE_a\right|\phi_3\rangle = \langle\phi_j\left|erE_a\right|\phi_3\rangle = \mu E, \qquad j = 1, 2 \tag{2.5}$$

In this case, the probability of transition between the upper state and the state Φ becomes

$$W_2 = \frac{|\mu E|^2}{4}\left|C_1(1 + e^{i\omega_{12}t}) + C_2(1 + e^{-i\omega_{12}t})\right|^2 \tag{2.6}$$

There is no way in which this function can become time-independent: it is and remains a periodic function of time. This shows how critical the selection rules become in the three-level case.

Let us consider again the first case which yields the transition probability W_1. An interesting situation occurs if the superposition state is $\Phi_- = C_1(\phi_1 - \phi_2)$, i.e., $C_1 = -C_2$. In this case $W_1 = 0$ as a result of maximum destructive interference. Indeed, the vanishing of W_1 results from the relations $|C_1|^2 = |C_2|^2 = -C_1C_2^* = -C_1^*C_2$. Physically, this means that an atom which is initially in the upper state will be unable to decay in the state Φ_- via a dipole-mediated transiiton. Conversely, an atom which is initially in the state Φ_- will remain in that state forever. Such states are called trap states. However, one can show that there is another state, $\Phi_+ = C_1(\phi_1 + \phi_2)$, with which the upper state can interact.

At this point, we can already grasp the principles which underlie the process of gain without population inversion. Let us consider an initial situation in which there are $N_j \neq 0$ atoms in level j. If we are able to prepare atoms in states Φ_- and Φ_+ with numbers of atoms N_- and N_+, respectively, then gain only requires that $N_3 > N_+$. What makes this result fascinating is that this condition, which is the ordinary two-level gain condition, may be compatible with the condition of no population inversion $N_1 > N_3$ and $N_1 > N_3$. Hence the expression "gain without population inversion".

If the upper state is empty, then $C_1 = 1/\sqrt{2}$ and the trap and interacting states are the well-known combinations $\Phi_\pm = \frac{1}{\sqrt{2}}(\phi_1 \pm \phi_2)$. The presence of atoms in the upper state does not change the discussion. The general wave function of the system is then $\Psi = C_1\phi_1 + C_2\phi_2 + C_3\phi_3$ with the normalization constraint $|C_1|^2 + |C_2|^2 + |C_3|^2 = 1$. The choice $C_1 = -C_2$ implies $2|C_1|^2 + |C_3|^2 = 1$.

2.2 The formal framework

Since we shall be dealing with more than two atomic levels, some caution must be exercised when dealing with the phenomenological constants which are introduced in the evolution of the density matrix elements. For instance, it is well known that for a two-level system, the constraint $2\gamma_\perp \geq \gamma_\parallel$ holds. The question we want to address in this section is what inequality is to be used in the case of a multilevel medium.

Let us consider an isolated atomic system. In the absence of any interaction with any coherent field, we may assume that the coherence decay as

$$\frac{d\rho_{jk}}{dt} = \left[-\gamma_{jk} + i\left(\omega_p - \omega_q\right)\right]\rho_{jk}, \qquad j \neq k \tag{2.7}$$

For the population dynamics, we introduce the transition probabilities $W(j \to k)$ and express the dynamics in terms of a balance between populating and depopulating processes

$$\frac{d\rho_{jj}}{dt} = -\sum_k \rho_{kk} W(j \to k) + \sum_k \rho_{kk} W(k \to j) \tag{2.8}$$

However, the transition probabilities $W(j \to k)$ must be proportional to the population N_j. Hence we write

$$W(j \to k) \equiv N_j \gamma_{\parallel jk} = N_j / T_1^{jk} \tag{2.9}$$

In terms of these transition probabilities, the decay rate of the coherence can be shown to verify the relation

$$\gamma_{\perp jk} \equiv \gamma_{jk} \equiv \frac{1}{T_2^{jk}} = \frac{1}{2}\sum_l \left[W\left(j \to l\right) + W\left(k \to l\right)\right] \tag{2.10}$$

The sum over l is unrestricted and includes the $W\left(l \to l\right)$ transitions which are positive and describe higher order processes in which, for instance, one has the sequence of transitions $l \to j \to l$. Hence we have the inequality

$$\begin{aligned} 2\gamma_{jk} &\geq \sum_{l \neq j} W(j \to l) + \sum_{l \neq k} W(k \to l) \\ &\geq \sum_{l \neq j} N_l \gamma_{\parallel kl} + \sum_{l \neq k} N_l \gamma_{\parallel jl} \end{aligned} \tag{2.11}$$

If there are only two levels with $N_1 + N_2 = 1$, the inequality reduces to the usual relation $2\gamma_{12} \geq \gamma_{\|12}$. However, if there are more than two levels, the interrelations between the various decay rates becomes more complex and introduce constraints on the possible range in which the rates may be chosen. For instance, if all the coherence decay rates are equal ($\gamma_{jk} \equiv \gamma$) and all the population decay rates are equal ($\gamma_{\|jl} \equiv \gamma_{\|}$), it follows from (2.11) that $2\gamma \geq (1 + N_j)\gamma_{\|}$. Summing over j leads to $3\gamma \geq 2\gamma_{\|}$, quite different from the two-level result.

Chapter 3

The Λ schemes

3.1 Formulation

In this section, we shall analyze more systematically the scheme which was described in chapter 2. The electric field and atomic polarization are decomposed as follows

$$
\begin{aligned}
E_{tot} &= \frac{1}{2}\left[E_a e^{i(k_a x-\omega_a t)} + E_b e^{i(k_b x-\omega_b t)} + c.c.\right] \\
P_{tot} &= \frac{1}{2}\left[P_a e^{i(k_a x-\omega_a t)} + P_b e^{i(k_b x-\omega_b t)} + c.c.\right] \qquad (3.1)
\end{aligned}
$$

The relations between the components P_j of the polarization and the density matrix are

$$
P_a = \mu_{13}\rho_{31}e^{i\omega_a t} \equiv \mu_{13}\sigma_{31}, \qquad P_b = \mu_{23}\rho_{32}e^{i\omega_b t} \equiv \mu_{23}\sigma_{32} \qquad (3.2)
$$

Therefore the two field equations are

$$
\begin{aligned}
\left(\partial_x + v_a^{-1}\partial_t + \kappa_a\right) E_a &= (4\pi i N\mu_{13}\omega_a/v_a\varepsilon_a)\sigma_{31} \qquad (3.3) \\
\left(\partial_x + v_b^{-1}\partial_t + \kappa_b\right) E_b &= (4\pi i N\mu_{23}\omega_b/v_b\varepsilon_b)\sigma_{32} \qquad (3.4)
\end{aligned}
$$

Likewise, the equations which govern the time evolution of the optical coherence are

$$
\begin{aligned}
\frac{\partial\sigma_{31}}{\partial t} &= -(\gamma_{31}+i\delta_a)\sigma_{31} + \frac{i\mu_{31}}{2\hbar}E_a n_{13} + \frac{i\mu_{32}}{2\hbar}E_b\sigma_{21} \qquad (3.5) \\
\frac{\partial\sigma_{32}}{\partial t} &= -(\gamma_{32}+i\delta_b)\sigma_{32} + \frac{i\mu_{32}}{2\hbar}E_b n_{23} + \frac{i\mu_{31}}{2\hbar}E_a\sigma_{12} \qquad (3.6) \\
& \delta_a = \omega_{31}-\omega_a, \qquad \delta_b = \omega_{32}-\omega_b
\end{aligned}
$$

In these equations, we have introduced the important low frequency coherence ρ_{21} and its slowly varying envelope defined by $\rho_{21} = \sigma_{21}\exp\left[i\left(\omega_b-\omega_a\right)t\right]$. To be complete, we should write the evolution equations for the low frequency coherence and for the populations ρ_{jj}. However, these equations are not necessary to determine the nature of the solutions in the linearized regime. Therefore, we shall analyze these additional equations at a later stage of the analysis. For the linearized analysis, it will be sufficient to assume that σ_{21} and the ρ_{jj} are constants, given therefore by their initial value.

3.2 Linear propagation regime

In the linearized regime, we seek plane wave solutions of the form

$$\begin{pmatrix} E_a \\ E_b \\ \sigma_{31} \\ \sigma_{32} \end{pmatrix} = \begin{pmatrix} \mathcal{E}_a \\ \mathcal{E}_b \\ s_{31} \\ s_{32} \end{pmatrix} e^{i(kx-\omega t)} \tag{3.7}$$

Our purpose is to derive the dispersion relation $k = k(\omega)$. Inserting these expressions into equations (3.3)-(3.6) yields a pair of equations for the atomic polarizations

$$s_{31} = \frac{i}{2\hbar}\left(\mu_{32}\mathcal{E}_b\sigma_{21} + \mu_{31}\mathcal{E}_a n_{13}\right) / \left(\gamma_{31} + i\delta_a - i\omega\right) \tag{3.8}$$

$$s_{32} = \frac{i}{2\hbar}\left(\mu_{32}\mathcal{E}_b n_{23} + \mu_{31}\mathcal{E}_a\sigma_{12}\right) / \left(\gamma_{32} + i\delta_b - i\omega\right) \tag{3.9}$$

and a pair of equations for the fields

$$\left[i\left(k - \omega/v_a\right) + \kappa_a + g_a n_{13}\right]\mu_{31}\mathcal{E}_a + g_a\sigma_{21}\mu_{32}\mathcal{E}_b = 0 \tag{3.10}$$

$$g_b\sigma_{12}\mu_{31}\mathcal{E}_a + \left[i\left(k - \omega/v_b\right) + \kappa_b + g_b n_{23}\right]\mu_{32}\mathcal{E}_b = 0 \tag{3.11}$$

where we have introduced the notation

$$g_a = \frac{2\pi N\omega_a \left|\mu_{31}\right|^2}{\gamma_{31}\varepsilon_a v_a\hbar\left[1 + i\left(\delta_a - \omega\right)/\gamma_{31}\right)]} \tag{3.12}$$

$$g_b = \frac{2\pi N\omega_b \left|\mu_{32}\right|^2}{\gamma_{32}\varepsilon_b v_b\hbar\left[1 + i\left(\delta_b - \omega\right)/\gamma_{32}\right)]} \tag{3.13}$$

The final step is to express the compatibility condition for the existence of non trivial solutions $\mathcal{E}_j$. This leads to the dispersion relation

$$\begin{aligned} 0 &= (k-\omega/v_a)(k-\omega/v_b) - i(k-\omega/v_a)(\kappa_b + g_b n_{23}) \\ &\quad -i(k-\omega/v_b)(\kappa_a + g_a n_{13}) + L \end{aligned} \tag{3.14}$$

$$L = g_a g_b \left|\sigma_{12}\right|^2 - (\kappa_a + g_a n_{13})(\kappa_b + g_b n_{23}) \tag{3.15}$$

Note that although we follow the same reasoning as in section 1.2, the structure of this problem is completely different from the corresponding two-level problem. In the two-level problem, the dispersion relation arises from a compatibility condition between the field amplitude and the optical coherence. On the contrary, here the dispersion relation arises from the compatibility condition between the two field amplitudes. Rather than undertake a study of the general dispersion relation, we shall introduce some simplifications in order to reach more transparent physical conclusions. Let us therefore introduce the following assumptions:

- We shall assume that $\delta_a = \delta_b = \omega$ because this corresponds to the condition of maximum linear gains g_j.
- We assume negligible linear losses: $\kappa_j \simeq 0$.
- We assume that the two fields have equal velocity in the absence of the nonlinear medium: $v_a = v_b$.

With these simplification, it is easy to verify that the amplification condition, $\mathrm{Im}(k) < 0$, is equivalent to the condition $L > 0$ which is therefore

$$|\sigma_{12}|^2 > n_{13}n_{23} \tag{3.16}$$

Let us analyze this condition. First, we note that if there is no population in the upper level 3, (3.16) can never be verified. Indeed, if $\rho_{33} = 0$ we have $|\sigma_{12}|^2 \equiv |\rho_{12}|^2 > \rho_{11}\rho_{22}$ which is impossible for a density matrix which is definite positive. Hence, amplification requires atoms in the upper state. Next, we note that the amplification condition involves only functions which are in fact not yet defined: in this chapter, we have assumed up to now that σ_{12} and the ρ_{jj} are given constants. It turns out that they are not independent constants but matrix elements of the same density operator and therefore they cannot be chosen arbitrarily. Finally, the essential result is that the amplification condition cannot be verified spontaneously with the extra condition $n_{13} > 0$ and $n_{23} > 0$ which express the absence of population inversion. That is, a bichromatic field of arbitrary amplitude is unable to create spontaneously enough low frequency coherence σ_{12} to verify selfconsistently the gain condition. Fortunately! Otherwise, we would have proved that using a bichromatic field of arbitrary amplitude, gain can be achieved. To prove this result, an analysis similar to the nonlinear propagation made in section 1.3.2 must be carried out. This is a fairly heavy calculation which will not be carried out here. Hence, we arrive at the conclusion that in order to reach gain, we need to add a mechanism which produces extra low frequency coherence.

Despite the negative conclusion we have reached so far, it is worth considering the nature of the state of the system at threshold, even though the threshold can only be reached asymptotically for arbitrarily intense fields. We note that at threshold, i.e., for $|\sigma_{12}|^2 = n_{13}n_{23}$, it follows from (3.8)-(3.9) that the optical coherences σ_{31} and σ_{32} identically vanish. This is another significant difference with the two-level system. As a consequence, the density matrix has the structure

$$\rho = \begin{pmatrix} \rho_{11} & \rho_{12} & 0 \\ \rho_{21} & \rho_{22} & 0 \\ 0 & 0 & \rho_{33} \end{pmatrix} \tag{3.17}$$

It is quite easy to find the eigenvalues (the atomic populations) and the eigenvectors of this density matrix. The eigenvectors are

$$N_1 = N_3, \qquad N_2 = 1 - 2N_3, \qquad N_3 = \rho_{33} \tag{3.18}$$

while the corresponding eigenvectors are

$$\Psi_1 = \begin{pmatrix} -\sqrt{\rho_{22} - \rho_{11}} \\ \sqrt{\rho_{11} - \rho_{33}} \\ 0 \end{pmatrix}, \qquad \Psi_2 = \begin{pmatrix} \sqrt{\rho_{11} - \rho_{33}} \\ \sqrt{\rho_{22} - \rho_{11}} \\ 0 \end{pmatrix}, \qquad \Psi_3 = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \tag{3.19}$$

In terms of the atomic states

$$\varphi_1 = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, \qquad \varphi_2 = \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}, \qquad \varphi_3 = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \tag{3.20}$$

the eigenvectors of the density matrix are

$$\Psi_1 = E_b\varphi_2 + E_a\varphi_1, \qquad \Psi_2 = E_b\varphi_2 - E_a\varphi_1 \tag{3.21}$$

These eigenvectors (which are not normalized) generalize quite clearly the trap and the interacting states Φ_- and Φ_+ introduced in section 2.1. They also provide a simple physical interpretation of the mechanisms which lead to gain without inversion. Since $|\langle\Psi_2 |erE_{tot}| \Psi_3\rangle|^2 = 0$, only the pair of states (Ψ_1, Ψ_3) contribute to the absorption and emission processes. However, they are equally populated. Hence they are transparent to the radiation which can propagate without attenuation. And since the trap state Ψ_2 is the most populated state if $N_3 < 1/3$, this threshold obviously does not require population inversion between level 3 and the other two levels. This is in complete opposition with the result that amplification requires $A > 1$ if the field propagates in a two-level medium since this threshold condition implies an initial population inversion.

3.3 The driven Λ scheme

3.3.1 Formulation

The next step is to consider a system in which there is an additional element which will induce the necessary low frequency coherence. Various strategies have been imagined to realize this program: as usual, the imagination of physicists has shown itself to be pretty unbounded. For the purpose of this course, we shall analyze the driven Λ scheme because it will provide an opportunity to discuss the question of the two mechanisms on which gain without inversion relies.

Let us therefore consider the Λ system described in the previous section and add a strong driving field

$$\overline{E}_p = \frac{1}{2}\left(E_p e^{i(k_p x - \omega_p t)} + c.c.\right) \tag{3.22}$$

which couples levels 1 and 2. We also assume that this driving field is strong enough that its attenuation due to the light-matter interaction can be neglected. Hence it will be treated, in first approximation, as a constant. The presence of the driving field induces a modification of the density matrix whose optical coherences are now given by

$$\frac{\partial \sigma_{31}}{\partial t} = -(\gamma_{31} + i\delta_a)\sigma_{31} + \frac{i\mu_{31}}{2\hbar}E_a n_{13} + \frac{i\mu_{32}}{2\hbar}E_b\sigma_{21} - \frac{i\mu_{21}}{2\hbar}E_p\sigma_{32} \tag{3.23}$$

$$\frac{\partial \sigma_{32}}{\partial t} = -(\gamma_{32} + i\delta_b)\sigma_{32} + \frac{i\mu_{32}}{2\hbar}E_b n_{23} + \frac{i\mu_{31}}{2\hbar}E_a\sigma_{12} + \frac{i\mu_{12}}{2\hbar}E_p^*\sigma_{31} \tag{3.24}$$

The rest of the analysis proceeds as in the previous section: we seek plane wave solutions and the compatibility condition between the two equations for the complex field amplitudes E_a and E_b determines the dispersion relation from which we derive the amplification threshold as the instability condition. The general form of the dispersion relation is

$$\begin{aligned} 0 &= (k - \omega/v_a)(k - \omega/v_b) - i(k - \omega/v_a)\Gamma_b \\ &\quad -i(k - \omega/v_b)\Gamma_a + G - \Gamma_a\Gamma_b \end{aligned} \tag{3.25}$$

where we have defined

$$G = g_a g_b \frac{(\sigma_{21} - i\gamma_p n_{23}\nu_b)(\sigma_{12} - i\gamma_p^* n_{13}\nu_a)}{(1 + \nu_a\nu_b|\gamma_p|^2)^2} \tag{3.26}$$

$$\Gamma_a = \kappa_a + g_a\frac{n_{13} - i\gamma_p\sigma_{12}\nu_b}{1 + \nu_a\nu_b|\gamma_p|^2} \tag{3.27}$$

$$\Gamma_b = \kappa_b + g_b\frac{n_{23} - i\gamma_p^*\sigma_{21}\nu_a}{1 + \nu_a\nu_b|\gamma_p|^2} \tag{3.28}$$

and

$$1/\nu_a = \gamma_{\perp 31} + i(\omega_{31} - \omega_a - \omega) \tag{3.29}$$

$$1/\nu_b = \gamma_{\perp 32} + i(\omega_{32} - \omega_b - \omega) \tag{3.30}$$

$$\gamma_p = \mu_{21}E_p/2\hbar \tag{3.31}$$

For simplicity, we again assume $v_a = v_b = v$. There are two branches of solutions to the dispersion relation

$$k_\pm = \omega/v + \frac{i}{2}\left\{\Gamma_a + \Gamma_b \pm \sqrt{(\Gamma_a + \Gamma_b)^2 + 4(G - \Gamma_a\Gamma_b)}\right\} \tag{3.32}$$

3.3.2 The two mechanisms

There are two ways in which this expression may lead to amplification, i.e., $\mathrm{Im}\, k_\pm < 0$: either $\mathrm{Re}(G - \Gamma_a\Gamma_b) > 0$ or $\mathrm{Re}(\Gamma_a + \Gamma_b) < 0$. We shall see that these two possibilities correspond to two mechanisms..

To simplify somewhat the analysis, let us consider the much simpler case of line center amplification with equal detunings ($\omega_{31} - \omega_a = \omega_{32} - \omega_b = \omega$), resonant pumping ($\omega_{21} = \omega_p$), and negligible passive losses ($\kappa_a = \kappa_b = 0$).

- $\mathrm{Re}(G - \Gamma_a\Gamma_b) > 0$. With the simplifications we have just introduced, this condition reduces nicely to the condition (3.16): $|\sigma_{12}|^2 > n_{13}n_{23}$. The difference between the present result and (3.16) is that now the density matrix is a function of the driving field E_p. Therefore, the condition $|\sigma_{12}|^2 > n_{13}n_{23}$ is an implicit equation for the driving filed. The other point which merits attention is the following. It has been shown that the condition $|\sigma_{12}|^2 > n_{13}n_{23}$, which is expressed in the bare state basis, is strictly equivalent to the condition of population inversion in the dressed state basis. Hence, in this case there is a unitary transformation of the density matrix which leads to a representation in which gain occurs in the natural way, namely with population inversion. This situation is referred to as *hidden inversion.* Its is intellectually reassuring since we can still understand gain in terms of a classical picture, without recourse to purely quantum arguments.

- $\mathrm{Re}(\Gamma_a + \Gamma_b) < 0$. Here the situation is more tricky since one of the two transitions may be forbidden. In such a case, there is no more interference between two decay channels and all the arguments we have used up to now fail to explain why there could be gain. Recently, it has been shown that the gain mechanism is still atomic interference.. More precisely, it has been shown that only one Feynmann diagrams contributes to the emission process which retains its usual expression but that absorption displays an interference between two diagrams. This leads once again to an interference process for the absorption coefficient which is a transition probability and therefore the modulus squared of the sum of two complex coefficients. Because of this interference, the symmetry between emission and absorption is broken, paving the way to gain without population inversion.

Let us now analyze more explicitly the two gain conditions. The first condition, $|\sigma_{12}|^2 > n_{13}n_{23}$, requires a knowledge of the populations and the low frequency coherence in steady state. For this, we need to specify the evolution equations for these variables. Using the reservoir theory, it can be shown the three-level generalization of (1.12) for the low frequency coherence and (1.13) for the population differences is

$$\frac{\partial \sigma_{21}}{\partial t} = -\left[\gamma_{21} + i\left(\omega_{21} - \omega\right)\right]\sigma_{21} + i\gamma_p n_{12}$$

$$+i\frac{\mu_{23}E_b^*}{2\hbar}\sigma_{31} - i\frac{\mu_{31}E_a}{2\hbar}\sigma_{23} \tag{3.33}$$

$$\frac{\partial\rho_{11}}{\partial t} = \gamma_{\|21}\left(N_1\rho_{22} - N_2\rho_{11}\right) + \gamma_{\|31}\left(N_1\rho_{33} - N_3\rho_{11}\right) - 2\,\mathrm{Im}\left(\frac{\mu_{13}E_a^*}{2\hbar}\sigma_{31} + \gamma_p^*\sigma_{21}\right) \tag{3.34}$$

$$\frac{\partial\rho_{22}}{\partial t} = \gamma_{\|21}\left(N_2\rho_{11} - N_1\rho_{22}\right) + \gamma_{\|32}\left(N_2\rho_{33} - N_3\rho_{22}\right) - 2\,\mathrm{Im}\left(\frac{\mu_{23}E_b^*}{2\hbar}\sigma_{32} - \gamma_p^*\sigma_{21}\right) \tag{3.35}$$

In the absence of light-matter interaction, the steady state solution for the populations are $\rho_{jj} = N_j$ which represents the net balance between the incoherent pumping and decay processes taking place in the medium. Note, however, that we are considering here a closed system since $N_1 + N_2 + N_3 = 1$. The reason for imposing this constraint is that it accounts in a simple way for the fact that, in practical situations, populating a level means depopulating other levels. Therefore, all the conclusions we shall reach are restricted to these closed systems.

Since we are considering the response of the atomic medium driven by a strong pump field E_p and weak probe fields E_a and E_b, it is sufficient that we evaluate the steady state expressions of σ_{21}, ρ_{11}, ρ_{22} and ρ_{33} with zero probe fields. This gives

$$\sigma_{21} = i\gamma_p n_{12}/\left[\gamma_{21} + i\left(\omega_{21} - \omega\right)\right] \equiv i\overline{\gamma}_p n_{12} \tag{3.36}$$

$$n_{13} = \left(N_{13} + 4\left|\overline{\gamma}_p\right|^2\gamma_{21}/\gamma_e\right) / \left(1 + +4\left|\overline{\gamma}_p\right|^2\gamma_{21}/\gamma_f\gamma_e\right) \tag{3.37}$$

$$n_{23} = \left(N_{23} + 4\left|\overline{\gamma}_p\right|^2\gamma_{21}/\gamma_e\right) / \left(1 + +4\left|\overline{\gamma}_p\right|^2\gamma_{21}/\gamma_f\gamma_e\right) \tag{3.38}$$

$$n_{12} = N_{12}/\left(1 + +4\left|\overline{\gamma}_p\right|^2\gamma_{21}/\gamma_f\gamma_e\right) \tag{3.39}$$

with $n_{jk} \equiv \rho_{jj} - \rho_{kk}$, $N_{jk} = N_j - N_k$ and where we have used the notation

$$\gamma_e = \frac{1}{2}\left(N_{13}\gamma_{\|31} + N_{23}\gamma_{\|32}\right)/d \tag{3.40}$$

$$\gamma_f = \left(N_1\gamma_{\|31} + N_2\gamma_{\|32} + \frac{1}{2}N_3\gamma_{\|32} + \frac{1}{2}N_3\gamma_{\|31}\right)/d \tag{3.41}$$

$$d = N_1\gamma_{\|21}\gamma_{\|31} + N_2\gamma_{\|21}\gamma_{\|32} + N_3\gamma_{\|31}\gamma_{\|32} \tag{3.42}$$

The correction to these expressions for σ_{21}, ρ_{11}, ρ_{22} and ρ_{33} is quadratic in the probe fields E_a and E_b. Although these are rather heavy expressions, it turns out that the gain condition, $\left|\sigma_{12}\right|^2 > n_{13}n_{23}$, has eventually a simple expression in terms of the auxiliary variable $x \equiv 4\left|\gamma_p\right|^2/(\gamma_e\gamma_{21})$, namely

$$f(x) = x^2 + x\left[N_{13} + N_{23} - N_{12}^2/(4\gamma_e\gamma_{21})\right] + N_{13}N_{23} > 0 \tag{3.43}$$

The problem is eventually reduced to finding a domain in the parameter space $\{N_1, N_2, N_3, \gamma_{12}, \gamma_{13}, \gamma_{23}, \gamma_{\|12}, \gamma_{\|13}, \gamma_{\|23}\}$ subject to the constraints $N_3 < N_1$, $N_3 < N_2$, $\sum_j N_j = 1$ and the inequality (2.11) where $f(x) > 0$. For instance, the gain condition (3.43) does not have a solution if either $\gamma_{\|31} = \gamma_{\|32}$ or $\gamma_{\|21} \ll \gamma_{\|32}\gamma_{\|31}$. However, if $\gamma_{\|21} \gg \gamma_{\|31} \gg \gamma_{\|32}$, there exists a domain of gain with the restrictions we have imposed. Note also that if we do not impose the assumption of a closed system, and therefore do not impose the inequality (2.11), the domain of parameters becomes significantly larger. The main point, however, is the proof that gain without population inversion may be reached even with all the restrictions we have imposed.

Let us consider next the second mechanism. Since it does not require two probe fields, we shall assume that the transition 3-2 is forbidden. This becomes a variant of the V scheme, although we call it the h scheme to stress the fact that we have in mind schemes in which there is a low frequency driving field and a high-frequency probe field. In this case, the Bloch equations (3.23)-(3.24) and (3.33)-(3.35) imply that there is nevertheless a coherence which is induced between levels 3 and 2. Indeed, the low frequency pump field is coupled to the low frequency population difference to drive the low frequency coherence. This coherence, coupled to the probe field E_a is the source of the coherence σ_{32} despite the restriction $\mu_{32} = 0$.

To analyze the gain condition $\mathrm{Re}(\Gamma_a + \Gamma_b) < 0$, we shall assume resonant driving ($\omega_{31} - \omega_a = \omega$ and $\omega_{21} = \omega_p$) and the symmetric case $\gamma_{\|31} = \gamma_{\|31}$. With these simplifications the gain condition becomes

$$|\gamma_p|^2 > N_{13}/\left[\left(N_{12}/\gamma_{32} - 4\gamma_e\right)/\gamma_{21}\right] \tag{3.44}$$

In the absence of population inversion, $N_{13} > 0$, gain requires $N_{12} > 4\gamma_e\gamma_{32}$ which can be transformed into

$$N_{12}\left[(N_1 + N_2)\gamma_{\|12} + N_3\gamma_{\|13}\right] > (N_{13} + N_{23})\left(\gamma_{\|13} + N_1\gamma_{\|12}\right) \tag{3.45}$$

If $\gamma_{\|12} \leq \gamma_{\|13}$, the inequality (3.45) cannot be fulfilled. However, in the opposite regime $\gamma_{\|12} \gg \gamma_{\|13}$, the inequality $N_{12} > 4\gamma_e\gamma_{32}$ becomes $2N_1(2N_3 - N_2) > N_2^2$ which is verified in the range

$$\frac{1}{2}N_2 < \lambda < N_3 < N_2 < 1/3 \tag{3.46}$$

where λ is the smallest root of $x^2 - x(1 - \frac{1}{2}N_2) + \frac{1}{2}N_2 = 0$. This completes the proof that there is a domain in parameter space where all the constraints we have imposed are verified and gain occurs.

3.4 A short bibliography

A short and incomplete bibliography of the "firsts" on gain without inversion in three-level media.

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