NONLINEAR AND ANISOTROPIC EFFECTS IN MAGNETICALLY TUNED LASER AMPLIFIERS

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ABSTRACT

NONLINEAR AND ANISOTROPIC EFFECTS IN MAGNETICALLY TUNED LASER AMPLIFIERS

Using semiclassical radiation theory and a density matrix formalism we analyze the nonlinear characteristics of a gas laser amplifier operating with two optical frequency signals of arbitrary polarization and having an axial magnetic field. Both perturbational solutions, valid for relatively weak intensities and solutions valid for arbitrarily strong fields are obtained for two nonlinear effects: the saturation interaction of the electromagnetic waves, and the generation of combination tones. An arbitrary amount of Doppler broadening is considered throughout.

The detailed treatment of $J = 1$ to $J = 0$ model yields the frequency, magnetic field and polarization dependence of the non-linear effects. The results are presented analytically and graphically and are discussed using physical arguments. It is found that only saturation but no combination tone generation occurs for two opposite circularly polarized input signals while both are, in general, present for two arbitrary linearly or elliptically polarized fields. For two opposite circular waves the interaction is found to comprise three parts, each with a different behavior: self saturation, common level mutual saturation and a coherent double quantum interaction. The total interaction (coupling) between the two fields is always weak. The limiting case of a single linearly polarized field is considered separately, the zero magnetic field "dip" and the nonlinear behavior

of the Faraday rotation is discussed.

For two linearly (or elliptically) polarized waves the three nonlinear processes listed above take place between opposite circular components. In addition a modulation of the population inversion densities occurs due to the presence of two different frequencies with the same circular polarization. This results in the generation of new frequencies and also contributes to the coupling between the input fields. The coupling depends on the magnetic field, and on the frequency separation and the polarization states of the signals. The limiting case of zero magnetic field is examined. It is found that the medium is made effectively anisotropic by the nonlinear interactions. The polarization vectors of two linearly polarized fields rotate apart unless the angle between them is zero or 90 degrees.

The results are extended to the general J_a to J_b transition. In zero magnetic field the nonlinear effects are found to depend on ΔJ, while for nonzero magnetic field resonances in the interaction occur whenever the frequency difference between two opposite circularly polarized transitions that have common level equals the frequency separation of the input fields. Combination tone generation takes place for all but two opposite circularly polarized signals.

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CHAPTER ONE

INTRODUCTION

It is a well-known fact that in a medium which is population inverted with respect to two of its atomic levels, an electromagnetic field resonant with the transition frequency between these two levels experiences negative absorption or gain (1). This amplification is brought about by induced transitions of the atoms between the laser levels. For very weak intensities the number of induced transitions is so small that the populations of the levels remain essentially unchanged. This is the linear case and, under such conditions, there exists no interaction between various spectral components of the field. As the intensities grow the number of stimulated emissions increases to measurably influence the level populations. Under these conditions the electromagnetic field couples nonlinearly to the medium, as a result of which there exist various nonlinear interactions between the spectral components of the field. In the following chapters we shall study these nonlinear interactions when the electromagnetic field consists of travelling waves containing several nearly monochromatic components with various polarization properties. We are primarily interested in effects that take place within a fairly narrow bandwidth of the original optical frequency inputs. That is to say we are not concerned with harmonic generation and coherent mixing, but with saturation, competition between two frequencies in the input, and side-band or combination tone generation. We are also interested in investigating the effects of an applied D.C. magnetic field and of the

Doppler broadening of the transition. The basis of our calculations is semiclassical radiation theory, as formulated by Kramers (2). The induced polarization of the medium is calculated by finding the dipole moment of the atoms constituting the medium. The induced polarization then is used in Maxwell's equations to study the behavior of the fields.

A number of authors have studied nonlinear effects of optical frequency electromagnetic waves recently. Armstrong, Bloembergen, Ducuing and Pershan (3) treated the interaction of travelling waves in nonlinear dielectrics including both crystals and gaseous media. Damping processes and Doppler broadening were not considered and no polarization effects were studied in any detail. Bloembergen and Shen (4) included the effects of atomic dissipation. Only stationary atoms were examined and no polarization effects were considered.

Lamb has treated nonlinear effects in laser oscillators in considerable detail (5). He used scalar cavity waves and did not attempt a treatment of polarization effects. Most of his results are applicable for the case of strong Doppler broadening only, although the case of stationary atoms was also considered briefly. The induced polarization was calculated to the third order in the fields and the properties of the laser oscillator were studied extensively.

Close (6,7) has used a similar approach for travelling waves. He has studied the effects of intermediate Doppler broadening and those of strong saturation. The generation of combination tones was examined in some detail. In addition, an attempt was made to treat polarization effects by using vector fields and a method of averaging over possible atomic dipole orientations. Haken and Sauermann (8,9) considered similar effects using similar techniques but did not treat combination tone generation or consider strong saturation.

Strong saturation in gas laser amplifiers with a single, scalar input field was studied by Gordon, White and Rigden (10) using a method based on rate equations. Only the case when the input field frequency is at line center is treated although the extension to arbitrary tuning is straightforward and has been done by Hotz (11).

Using still another approach based on the Kramers-Kronig relations, Bennett (12) has studied saturation in Doppler broadened gas lasers. The powerful concept of "hole burning" developed in his work will be made use of at times in our work, in a somewhat extended form, for a physical understanding of some results.

In all of the above references a single atomic transition between two nondegenerate levels was considered. In order to properly account for the effects of arbitrary polarization properties of the fields it is essential to consider the degeneracy of the levels. Since the degeneracy is removed by the application of a DC magnetic field to the medium, such a treatment leads naturally to the study of magnetic fields effects as well. This is what is done in our work. In order to place this aspect too in a proper perspective we list the works published on magnetic field effects as well.

Fork (13) has calculated the dielectric susceptibility tensor for a gaseous medium with an inverted population subjected to a magnetic field, using Kramers-Kronig relations. A $J = 1$ to $J = 0$ transition was treated in detail and formulae for generalizing the results given. Only the linear case was considered. Culshaw and Kannelaud in their early work on magnetic field effects in laser oscillators (14, 15) used a simplified quantum mechanical approach to study mode pulling and coherence effects. Faraday rotation in a population inverted medium has been examined by Tobias and Wallace (16) who have calculated the Verdet constant for a general J_a to J_b transition using a linear theory only.

Tang and Statz (17) have used a density matrix approach to study nonlinear pulling effects in a laser oscillator with Zeeman splitting of the levels by calculating nonlinear susceptibilities for a $J = 1$ to $J = 0$ transition. Only the time dependence of the fields was included and the effects of oppositely travelling waves constituting cavity modes therefore could not be included. Likewise, they did not consider different relaxation rates of the upper and lower laser levels. In the same work they also treated some polarization effects for linearly polarized waves and two nondegenerate levels by a method similar to that of Haken and Sauermann (8) and of Close (6).

To study the nonlinear properties of Zeeman laser oscillators Lamb's theory has been extended recently, to vector electromagnetic fields interacting with levels that are eigenstates of the angular momentum, by several authors. Fork and Sargent (18) have considered competition and beat frequencies between two opposite circularly polarized modes within a single cavity resonance for a $J = 1$ to $J = 0$

transition and an axial magnetic field. They did not, however consider other polarization states.

Culshaw and Kannelaud have considered the effects of cavity anisotropy as well as the behavior of the beat frequencies for a simple $J = 1/2$ to $J = 1/2$ model and axial magnetic field (19). More recently they have extended their results to cover the $J = 1$ to $J = 0$ transition and transverse magnetic field (20). Corney (21) has obtained some linear and nonlinear results for both oscillators and amplifiers by considering only the "classical" Zeeman effect for an axial magnetic field.

Heer and Graft (22) have considered an arbitrary J_a to J_b transition, arbitrary direction of the magnetic field and both cavity and travelling waves. Although the fundamental approach is very general actual results were obtained only for the case of a single linearly, circularly or elliptically polarized wave. The case of a single mode laser oscillator operating on a transition between levels of arbitrary J values was also examined by de Lang and Bouwhuis (23) and by Polder and Van Haeringen (24) who have shown that the polarization state of the mode depends on ΔJ. The effects of intermediate Doppler broadening or combination tone generation were not treated in these works or in references 17 through 22.

More recently there have appeared two other works that are related to ours. Doyle and White (25) have examined a laser oscillator with a general J_a to J_b transition in which the magnetic sublevels are completely degenerate, i.e., zero magnetic field. Two modes with arbitrary elliptical polarizations were considered, covering all possible combinations of polarization states for two frequency operations. Strong Doppler broadening was assumed and combination tone generation was also considered briefly.

Schlossberg and Javan (26) have studied a $J = 1$ to $J = 0$ transition with extension to more complex level structures. Particular emphasis was put on the resonant behavior of the nonlinear polarization and the effects of double quantum transitions on this. Both travelling and standing waves were included and intermediate Doppler broadening was also examined. The electromagnetic field was taken to be a scalar field and consequently polarization effects were not included.

There are various other references that deal with related ideas, such as other nonlinear effects we are not considering and with the Stimulated Raman Effect. Some of these, when appropriate, will be mentioned during the discussion of our results. Others will be listed in the concluding chapter where we discuss the relationship to other nonlinear effects.

Experimental work on the subject has closely followed the development of the theoretical knowledge. Gordon, White and Rigden (9), and Bennett(11) have included experimental results with their work. Several aspects of Lamb's theory (5) have been experimentally investigated (27, 28, 29). Close has reported observations of combination tones in a saturated amplifier (30). On the effects of magnetic fields, Fork and Patel (31) have reported first observation of Faraday rotation in a population inverted medium. Measurement of the beats within a single cavity resonance as well as of the variation of output power with magnetic field and observation of coherence effects were reported by Culshaw and Kannelaud (13, 14, 20, 32) and by Paananen, Tang and Statz (33). Independent measurement of the variation of low frequency beats with magnetic field has been done by Bolwijn (34). Doyle and White have reported observation of high order combination tones within a single resonance of a laser oscillator (35) and have recently verified some of their analytical results (25) for lasers oscillating on transitions between higher J values (36). De Lang and Bouwhuis have carried out experiments on the polarization state of a single mode laser oscillator in a magnetic field (23, 27). The work of Fork, Tomlison and Heilos (38) should also be mentioned in which they reported hysteresis effects that are not explainable by present theories. Very recently Schlossberg and Javan reported measurements of the hyperfine structure of Xenon (29) utilizing their theory (26) of the saturation behavior of magnetically tuned complex level structures.

In the work that follows we use an approach that is generally similar to that taken by Lamb (5), Close (6,7) Heer and Graft (22), and other authors on magnetic field effects. A simple $J = 1$ to $J = 0$ model is presented in chapter two and the equations of motion for the atoms and the vector electromagnetic fields are derived. For the four chapters that follow this simple model is considered. An integral equation "solution" is developed in chapter three, and in chapter four the nonlinear effects expected for various polarization states of the input fields are discussed qualitatively using physical arguments and the results of chapter three. In chapter five detailed results for the case of two opposite circularly polarized fields are derived. Both first and third order iterative solutions, valid for relatively weak fields and some solutions valid for arbitrarily high intensities are found. The important limiting case of a single linearly polarized field is also treated in some detail. In chapter six the case of two linearly or elliptically polarized fields is examined. Most of the nonlinear results are third order, valid for relatively weak fields only, but some semiquantitative expressions for arbitrary intensities are also obtained. Featured are the frequency and polarization dependence of the interactions, the effects of the magnetic field, induced anisotropies of the medium and combination tone generation. In chapter seven the results are extended to an arbitrary J_a to J_b transition and additional results different from those for the simple model are obtained. Finally in chapter eight we summarize the results and discuss applications and extensions of the theory. Throughout the work an arbitrary amount of Doppler broadening is considered although we also obtain results for the limiting cases of very strong Doppler broadening and natural broadening. We do not however, treat the effects of collisions or of boundary conditions. Likewise, the noise properties of the laser amplifier are ignored.

CHAPTER TWO

THE MODEL FOR THE ATOMIC SYSTEM AND THE EQUATIONS OF MOTION

2.1 Introduction

The purpose of this chapter is to introduce a suitable model for calculating the interaction of optical frequency electromagnetic waves with a population inverted medium. For that purpose we first find the equations of motion for a system consisting of a single atom and an electromagnetic field. The description of the atom will be quantum mechanical, i.e., described by the Schroedinger wave equations, while the electromagnetic field is treated classically. A simplified model of a single atom is described in section 2.2 and the electromagnetic field is treated in section 2.3. The interaction of these two and the resulting equations of motion are covered in section 2.4. Only the dipole interaction between the field and the atomic system is considered. In the subsequent chapters the single atom equations will be solved and macroscopic solutions will be obtained by summing over the atoms constituting the laser medium.

2.2 The Model for the Atomic System

Any given atom of the laser medium is fully described by its time independent eigenstates which are solutions of the Schroedinger equation

$$H_o|\varphi\rangle = E|\varphi\rangle \quad , \tag{2.1}$$

where $H_o = P^2/2m + V$ and E is the energy eigenvalue. $\overline{P}$ is the momentum operator and V is the potential energy of the electrons. The energy levels are eigenstates of the total angular momentum J . In the absence of an external magnetic field these eigenstates are degenerate in the magnetic quantum number. An applied D.C. magnetic field is a stationary perturbation that removes the degeneracy and splits a given level into 2J+1 sublevels according to the well known Zeeman formula

$$\Delta E = \frac{1}{2}\frac{e\hbar B}{m} gM_J \quad , \tag{2.2}$$

where ΔE is the change in energy of the M_J sublevel and g is the g factor of the level (40).

For our purposes we shall consider only two of the eigenstates, those which are population inverted and have a transition frequency resonant or nearly resonant with the electromagnetic field. In order to keep the calculations from becoming too complex the model used in the first part of this work will be the simplest possible that approximates an actual laser system. This is one in which the upper level has total angular momentum quantum number $J = 1$ and is thus threefold degenerate while the lower level is nondegenerate with $J = 0$. There are several neutral gas laser systems capable of CW oscillations between $J = 1$ and $J = 0$ levels, among others the 2.65μ line of Xe and the 1.52μ line of Ne are well known (41). For these systems this model is an exact description. It is evident that with trivial modifications the model is also correct for the $J = 0 \rightarrow J = 1$ transition of which the 3.99μ Xe line is an example (41). The theory will be extended to transitions between levels with arbitrary J values in Chapter Seven where additional results that are in some cases different from those obtained for the simple model will be derived. The effects of the nuclear spin and isotopes will also be considered in that chapter. The magnetic field is taken to be such magnitude that the Paschen-Back region is not reached.

The other eigensolutions of equation 2.1 enter into our calculations only by establishing the decay rates γ_a and γ_b for the two levels. In reality the decay rates are also influenced by the atoms' surroundings, radiation trapping etc., but no attempt will be made here to treat these effects. Instead the decay rates will be introduced phenomenologically into the atomic equations of motion.

2.3 The Electromagnetic Field and Its Equations of Motion

We assume the electromagnetic field as a sum of nearly monochromatic travelling waves propagating in the z direction. In view of the extremely narrow spectral width of laser signals this is very reasonable. The waves are also assumed to be transverse and nearly plane, that is to say transverse derivatives in the region of interest are assumed negligible. This second assumption is also reasonably well satisfied in practice. The laser medium is usually a discharge tube and the input signals can usually fill up the tube with only small variation of intensity across the diameter.

It will be found convenient to assume circularly polarized fields, and to define the vector base system as

$$\overline{E} = \sum_{m=0,\pm} (-1)^m E_m \hat{e}_{-m} \tag{2.3}$$

where

$$\hat{e}_{\pm} = \mp \frac{1}{\sqrt{2}} (\hat{e}_x \pm i\hat{e}_y) \text{ and } \hat{e}_o = \hat{e}_z$$

and

$$E_m = \hat{e}_m \cdot \overline{E} \quad .$$

For a magnetic field in the axial direction on the laser amplifier, i.e., in the direction of propagation of the field, the coordinate system of the atoms and of the field coincide. For other directions of the magnetic field a rotation matrix is necessary to relate the two coordinate systems. We shall consider only axial magnetic fields. The electromagnetic field is

$$\overline{E}(t,z) = \text{Re} \sum_{\nu} \sum_{m=\pm} \hat{e}_{-m} E_{\nu m}(z,t) \, e^{i(k_m(\nu)z-\nu_m t+\varphi_m)} , \tag{2.4}$$

where $E_{\nu m}(z,t)$ and φ_m are slowly varying functions of position and time in this approximate expansion. Arbitrary linearly, circularly or elliptically polarized waves result by choosing the magnitude and initial phase of the left and right circular components appropriately.

The field is governed by Maxwell's wave equations which for a nonmagnetic medium with no free charge can be written in MKS units as

$$\nabla \times \bar{E} = \frac{\partial \bar{B}}{\partial t} \quad , \tag{2.5a}$$

$$c^2 \nabla \times \bar{B} = \frac{\partial \bar{E}}{\partial t} + \frac{1}{\epsilon_o} \frac{\partial \bar{P}}{\partial t} \quad , \tag{2.5b}$$

$$\nabla \cdot \bar{E} = - \frac{1}{\epsilon_o} \nabla \cdot \bar{P} \quad , \tag{2.5c}$$

$$\nabla \cdot \bar{B} = 0 \ , \tag{2.5d}$$

where $\bar{P}$ is the macroscopic polarization of the medium. These can be combined to give

$$\nabla \times \nabla \times \bar{E} = - \frac{1}{c^2} \frac{\partial^2 \bar{E}}{\partial t^2} - \frac{1}{\epsilon_o c^2} \frac{\partial^2 \bar{P}}{\partial t^2} \quad . \tag{2.6}$$

After using the vector identity

$$\nabla \times \nabla \times \bar{E} = -\nabla^2 \bar{E} + \nabla(\nabla \cdot \bar{E}) \ , \tag{2.7}$$

and 2.4c, equation 2.5 can be rewritten as

$$-\nabla^2 \bar{E} + \frac{1}{c^2}\frac{\partial^2 \bar{E}}{\partial t^2} = \frac{1}{\epsilon_o}\nabla(\nabla\cdot\bar{P}) - \frac{1}{\epsilon_o c^2}\frac{\partial^2 P}{\partial t^2} \quad . \tag{2.8}$$

Since we are considering a system near resonance, the polarization of the medium has both real and imaginary parts. It is well-known that the imaginary part is responsible for the loss or gain in the medium while the real part causes a phase shift or index of refraction. Accordingly we assume the following form for the induced polarization $\bar{P}$:

$$\bar{P}(z,t) = \text{Re}\sum_{\nu}\sum_{m}(P_{\nu cm} - iP_{\nu sm})e^{i(k_m z - \nu_m t + \varphi_m)} \quad , \tag{2.9}$$

where the summation runs over the same set as for the electromagnetic field. Since the phase φ_m was assumed to be space and time dependent, the complete nonlinear phase shift suffered by the wave as it travels through the medium could be taken care by it and k_m could be assumed equal to the free space propagation constant ν_m/c . Since, however, we shall separately, albeit briefly, consider the linear results it will be convenient to define a real index of refraction by

$$k_m(\nu) = n_m(\nu)\frac{\nu_m}{c} \quad . \tag{2.10}$$

The index of refraction is convenient to use for linear results. For calculations that include nonlinear effects, however, the accumulated phase φ_m is more suitable. It is of course possible to reserve φ_m for the nonlinear corrections only and keep n_m for the linear part. We have considerable freedom of choice in the matter, and both will be included in the equations. Note also that we neglected the effect of the other, non-resonant transitions. These contribute a small index of refraction that is essentially constant through the small range of frequencies (the width of the resonance) considered, and can thus be taken into account by adding a small constant term to n_m .

We can now substitute the polarization of the medium into Maxwell's equation. Transverse derivatives are set equal to zero and the equation for one harmonic component is thus

$$\left[\left(k+\frac{\partial\varphi}{\partial z}\right)^2 E-2i\left(k+\frac{\partial\varphi}{\partial z}\right)\frac{\partial E}{\partial z}-\frac{\partial^2 E}{\partial z^2}-i\frac{\partial^2\varphi}{\partial z^2}E-\frac{1}{c^2}\left(-\nu+\frac{\partial\varphi}{\partial t}\right)^2 E+\frac{2i}{c^2}\left(-\nu+\frac{\partial\varphi}{\partial t}\right)\frac{\partial E}{\partial t}\right.$$

$$\left.+\frac{1}{c^2}\frac{\partial^2 E}{\partial t^2}+\frac{i}{c^2}\frac{\partial^2\varphi}{\partial t^2}E\right]e^{i(kz-\nu t+\varphi)}=\frac{1}{\epsilon_o c^2}\nu^2(P_c-iP_s)\,e^{i(kz-\nu t+\varphi)}, \quad (2.11)$$

where the subscripts νm have been omitted for convenience. The second derivative of P has been set equal to $-\nu^2 P$ since we know that P is nearly monochromatic at an optical frequency ν . The assumption of slow spatial and time variation will now be used to neglect the terms.

$$\frac{\partial^2\varphi}{\partial z^2}E,\ \frac{\partial\varphi}{\partial z}\frac{\partial E}{\partial z},\ \frac{\partial^2 E}{\partial z^2},\ \frac{1}{c^2}\frac{\partial^2\varphi}{\partial t^2}E,\ \frac{1}{c^2}\frac{\partial E}{\partial t}\frac{\partial\varphi}{\partial t},\ \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} \quad .$$

To justify this more quantitatively we note that we must have, for example,

$$\frac{\partial^2 E}{\partial z^2} \ll k \frac{\partial E}{\partial z} \quad , \text{ and} \tag{2.12a}$$

$$\frac{\partial^2 E}{\partial t^2} \ll \nu \frac{\partial E}{\partial t} \quad . \tag{2.12b}$$

If we assume linear amplification of the fields, then, the spatial variation of E can be expressed roughly as $E = E_o e^{\alpha z}$. Using $k = \nu c$ 2.12a becomes for optical frequencies

$$\alpha \ll \frac{\nu}{c} \approx 10^6/m \ , \tag{2.13a}$$

which is very well satisfied since the highest known α in lasers is of the order of 5. For a rough estimate on the validity of 2.12b let us assume that the time variation of E is $E = E \cos \Delta\nu t$, where $\Delta\nu$ is the total bandwidth over which the input signals are distributed. Then 2.12b becomes

$$\frac{\Delta\nu}{\nu} \ll 1 \quad , \tag{2.13b}$$

which is again very strongly satisfied, typical values of $\Delta\nu/\nu$ being less than 10^{-5} .

Replacing k by ν/c and $k^2 - \nu^2/c^2 = (k+\nu/c)(k-\nu/c)$ by $2\,\frac{\nu}{c}(k - \frac{\nu}{c}) = 2\,\frac{\nu^2}{c^2}\,(n-1)$, and equating real and imaginary parts we obtain the two equations.

$$\left[\frac{\partial \varphi_{\nu m}}{\partial z} + \frac{1}{c}\frac{\partial \varphi_{\nu m}}{\partial t} + \frac{\nu}{c}(n_m - 1)\right] E_{\nu m} = \frac{\nu}{2\epsilon_o c} P_{\nu c m} \tag{2.14a}$$

$$\frac{\partial E_{\nu m}}{\partial z} + \frac{1}{c}\frac{\partial E_{\nu m}}{\partial t} = \frac{\nu}{2\epsilon_o c} P_{\nu sm} \quad , \tag{2.14b}$$

where the subscripts νm have been restored where they are significant. These are the electromagnetic field equations in a form suitable for calculations of nonlinear interactions in the laser medium. The value of P will be calculated by considering the interaction of the field with the atoms of the medium.

2.4 Interaction of the Atoms and the Electromagnetic Field

The interaction Hamiltonian between the atoms and the fields is taken to be $H' = -\overline{p}\cdot\overline{E}$ where $\overline{p}$ is the dipole moment operator and $\overline{E}$ is the electric field. Magnetic dipole, electric quadrupole etc. interactions are neglected. Figure 1 shows schematically our model of an atom interacting with a classical electromagnetic field.

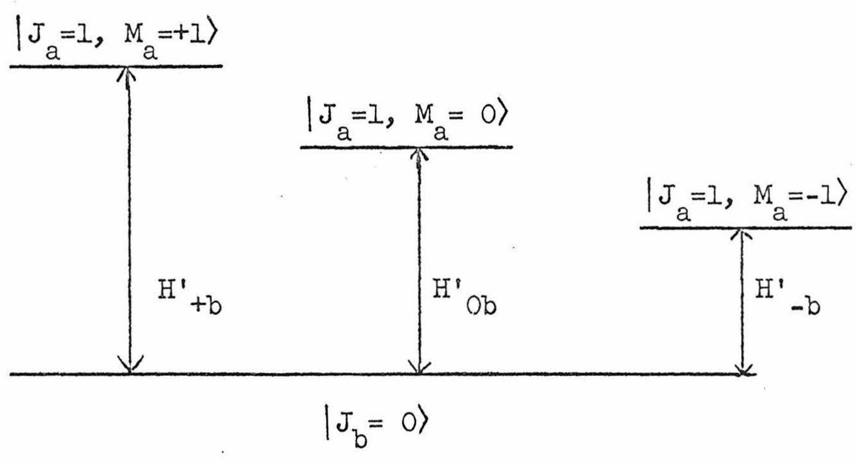


Figure 1

In the subsequent sections the subscript a and b will refer to the three upper levels and the one lower level respectively. The subscripts +, 0, -, are used to indicate the $M = +1, 0, -1$, sublevels of the upper level a. When the latter are used it is understood that we mean the upper levels, and the subscript a is left out.

We need the solutions of the time dependent Schroedinger equation $H|\psi\rangle = i\hbar \frac{\partial\psi}{\partial t}$. (2.15)

It will be convenient to use the density matrix formalism. The perturbed wave function is expanded as

$$|\psi\rangle = \sum_k c_k(t)\, |\varphi_k\rangle \quad , \tag{2.16}$$

where the $|\varphi_k\rangle$'s are the stationary unperturbed states. The density matrix is defined by the operator $|\psi\rangle\langle\psi|$, i.e.

$$\rho_{ij} = \langle\varphi_i|\psi\rangle\langle\psi|\varphi_j\rangle = c_i c_j^* \quad . \tag{2.17}$$

The equations of motion for the density matrix can be derived by

$$\frac{\hbar}{i}\frac{\partial}{\partial t}\Big(|\psi\rangle\Big)\langle\psi| \quad + \quad |\psi\rangle\, \frac{\hbar}{i}\frac{\partial}{\partial t}\Big(\langle\psi|\Big)$$

$$= -(H\rho - \rho H) \quad . \tag{2.18}$$

Since the $|\varphi_k\rangle$'s are the time independent wave functions H is the total Hamiltonian $H_o + H'$. Equation 2.18 is valid only if the complete set of wave functions are included. For our purposes we have limited the atomic model to two energy states, J_a + J_b stationary wave functions we must truncate the density matrix to include only these. The other wave functions are taken into account by introducing the phenomenological decay rates γ_a , γ_b . In the equations for the density matrix this has the effect of adding the anticommutator

$$i\hbar \frac{1}{2}(\Gamma\rho + \rho\Gamma) ,$$

to the RHS of equation (2.18) (42). The equation of motion for the density submatrix is

$$i\hbar\rho = (H_o - i\hbar\Gamma/2)\rho - \rho(H_o + i\hbar\Gamma/2) + H'\rho + \rho H' . \qquad (2.19)$$

That Γ is always diagonal in the energy representation and is given by $\delta_{ij}\gamma_j$ has been shown by Lowen (43). For the $J_a = 1$, $J_b = 1$ model used in the first part of this work the matrix elements of equation 2.19 are given by

$$\rho = \begin{bmatrix} \rho_{--} & \rho_{-o} & \rho_{-+} & \rho_{-b} \\ \rho_{o-} & \rho_{oo} & \rho_{o+} & \rho_{ob} \\ \rho_{+-} & \rho_{+o} & \rho_{++} & \rho_{+b} \\ \rho_{b-} & \rho_{bo} & \rho_{b+} & \rho_{bb} \end{bmatrix}$$

$$H_o = \begin{bmatrix} E_- & 0 & 0 & 0 \\ 0 & E_o & 0 & 0 \\ 0 & 0 & E_+ & 0 \\ 0 & 0 & 0 & E_b \end{bmatrix}$$

$$\Gamma = \begin{bmatrix} \gamma_a & 0 & 0 & 0 \\ 0 & \gamma_a & 0 & 0 \\ 0 & 0 & \gamma_a & 0 \\ 0 & 0 & 0 & \gamma_b \end{bmatrix}$$

and finally H' is the interaction matrix

$$H' = \begin{bmatrix} 0 & 0 & 0 & H'_{-b} \\ 0 & 0 & 0 & H'_{ob} \\ 0 & 0 & 0 & H'_{+b} \\ H'_{b-} & H'_{bo} & H'_{b+} & 0 \end{bmatrix}$$

Since these matrices are all Hermitian, $\rho_{ij} = \rho^*_{ji}$ and $H'_{ij} = H'^*_{ji}$. The polarization of an atom, i.e., the expectation of the dipole moment from the definition of ρ is

$$\overline{P}_{atom} = \text{Trace}\ [\rho\overline{p}] \quad . \tag{2.20}$$

To obtain the macroscopic polarization we must sum over all the atoms

$$\overline{P} = \sum_{\text{all atoms}} \text{Trace}\ [\rho\overline{p}]$$

$$= \text{Trace}\ [\rho_m\overline{p}] \quad , \tag{2.21}$$

where $\rho_m = \sum_{\text{all atoms}} \rho$. In the following chapter we derive closed form expressions for the elements of the macroscopic matrix ρ_m .

CHAPTER THREE

SOLUTIONS OF THE ATOMIC EQUATIONS OF MOTION

3.1 Introduction

In this chapter we obtain formal solutions for the atomic equations of motion. These "solutions" are actually a conversion of the differential equations into a set of integral equations from which perturbational and other approximate solutions can be easily obtained. The treatment is similar to that of Lamb (5) and of Close (6,7) for two non-degenerate atomic levels. We discuss the initial conditions necessary for the integration of the differential equations in section 3.2 and the integrated equations for one atom with a given initial condition are obtained in 3.3. Formal solutions for the macroscopic density matrix are derived in section 3.4. The basic assumption in this treatment is that the velocity of the atom remains constant during the time required for the atom to decay. Another assumption is that the field amplitudes change slowly compared to the decay rates of the levels γ_a, γ_b. Actually, the validity of the first assumption is somewhat marginal and under certain conditions collision effects can be quite important. This topic will be more fully discussed in section 3.3.1.

3.2 Initial Conditions and Excitation

The simplest initial condition that can be assumed for a given atom is that at some time $t = t_o$ it is in one of the two stationary states that we are considering, i.e. either in one of the magnetic substates of the upper J=1 level or in the lower J=0 state. Mathematically this is described by an initial value of the density matrix in which all but one of the diagonal elements is zero and the value of the latter is unity. The differential equations for the density matrix elements can then be integrated. To obtain the macroscopic polarization of the medium it will be necessary to sum over all the atoms that are involved. To account for all the relevant atoms we assume an excitation (or pumping) process whereby a certain number of atoms get excited per unit volume of the medium and unit time interval to each of the four sublevels. In reality, of course, the atoms are excited to various mixtures of the eigenstates. The assumption that the excitation is to one level at a time is equivalent to an assumption of randomness in the possible superposition of the eigenstates at $t = t_o$. We must of course take into account the motion of the atoms. This is done by assuming that any given atom has a velocity $\bar{v}$ at the time of excitation, $t = t_o$, and assuming a velocity distribution for the ensemble of atoms. Thus the number of atoms excited to the levels of interest per unit time and unit volume is given by the quantities

$$\lambda_+ W(\bar{v}), \quad \lambda_o W(\bar{v}), \quad \lambda_- W(\bar{v}), \quad \text{and} \quad \lambda_b W(\bar{v}) \quad .$$

We have taken the velocity distribution to be the same for all levels. In all subsequent calculations we shall assume either $W(\bar{v}) = \left(\frac{1}{\pi u^2}\right)^{3/2} e^{-v^2/u^2}$, a Maxwellian distribution, or $W(\bar{v}) = \delta(\bar{v})$, i.e., stationary or very slow moving atoms. These initial conditions describe many laser systems well, particularly the neutral atom lasers. In most ion lasers, because of the D. C. current carried by the lasering atoms, the velocity distribution is non Maxwellian, which must be taken into account. Other than the simple initial conditions assumed here are also possible. For example, the atom is initially in a specific coherent superposition of the upper states. We might encounter such behavior in a laser excited by absorption of strongly polarized light. No attempt will be made here to treat these various special cases.

3.3 Formal Solutions for One Atom

3.3.1 Simplifying Assumptions and Their Validity

Let us consider an atom that gets excited with velocity $\bar{v}$ to one of the four pertinent levels at time $t = t_o$ and at the position $\bar{r} = \bar{r}_o$. The interaction time is roughly the time it takes the atom to decay, i.e. $1/\gamma_a$ or $1/\gamma_b$ for levels a and b respectively. This being of the order of 10^{-7} sec. we can make three important simplifying assumptions viz. that during the interaction time

1.) the velocity $\bar{v}$ of the atom does not change

2.) the states are not perturbed by collisions, and

3.) the amplitude of the electromagnetic field remains constant.

The validity of the first two of these assumptions depends on the decay rates γ_a, γ_b being larger than the collision frequency, which condition is not always satisfied. Typically in a He-Ne laser the collision frequency varies from about 10^{-7} sec. to about 10^{-6} sec., while the decay times $1/\gamma_a$, $1/\gamma_b$ are of the order of 10^{-7} sec. There is thus a region of overlap and for certain operating conditions collision effects cannot be ignored. For lasers without magnetic field and restricted to one linear polarization (e.g. by Brewster angle windows) Pollack and Fork (29) have shown that collision effects can be well accounted for by a simple modification of Lamb's theory. In their work the simple damped resonant atomic response $[\gamma_{ab}+i(\omega-\nu)]^{-1}$ is replaced by the form $e^{ic}/[\gamma'_{ab}+i(\omega_s-\nu)]^{-1}$ where c, γ'_{ab} and $\omega_s-\omega$ are linear functions of pressure. On the other hand Szöke and Javan (28) were able to fit their data for the single mode central tuning dip by considering only the velocity shift effects of collisions. Some attempts have been made to include both effects (44) but these lead to very involved calculations. For our more complicated model (basically a three level instead of a two level system) consideration of pressure effects is even more difficult. In addition to causing velocity shifts and modifying the atomic response curves, collisions can cause decay from one magnetic sublevel to another. Collision induced coupling between substates have been tentatively identified as the explanation for the strong interaction (hysteresis) effects observed in a J=1 → J=0 Xe laser, for certain values of pressure, by Fork, Tomlison and Heilos (38). In this work we will ignore collision effects entirely and concentrate instead on a full description of the nonlinear effects resulting from the presence of strong electromagnetic fields alone.

Finally the validity of the last assumption depends on the gain of the laser not being excessively large. The distance traveled by an atom before it decays is roughly $(1/\gamma_{a,b})\ v_{thermal} \sim 10^{-4}$ m. For a small-signal gain of 80 db/meter this corresponds to about .1% increase in the field intensity which can indeed be neglected. For most laser systems the gain is actually much less. We shall not, in fact, make use of the last assumption in this chapter but only in chapter five where specific forms of the fields are substituted into the interaction matrix.

3.3.2 The Single Atom Solutions. Axial Magnetic Field.

The ten equations implied by 2.19, simplify considerably for axial magnetic field on the laser. For this case the coordinate systems of the atoms and of the optical frequency field coincide. Since the electromagnetic field is considered transverse and the expectation of the dipole moment operator between states of equal magnetic quantum numbers is polarized in the z direction, the perturbation connecting such states is indentically zero. Then, the pertinent equations are:

$$\dot{\rho}_{--} = -\gamma_a \rho_{--} + i(V^*_{-b}\rho_{-b} - V_{-b}\rho_{b-}) \qquad (3.1a)$$

$$\dot{\rho}_{++} = -\gamma_a \rho_{++} + i(V^*_{+b}\rho_{+b} - V_{+b}\rho_{b+}) \qquad (3.1b)$$

$$\dot{\rho}_{bb} = -\gamma_b \rho_{bb} - i(V^*_{-b}\rho_{-b} - V_{-b}\rho_{b-}) - i(V^*_{+b}\rho_{+b} - V_{+b}\rho_{b+}) \qquad (3.1c)$$

$$\dot{\rho}_{+-} = -(\gamma_a + i\omega_{+-})\ \rho_{+-} + i\ V^*_{-b}\rho_{+b} - i\ V_{+b}\rho_{b-} \qquad (3.1d)$$

$$\dot{\rho}_{-b} = -(\gamma_{ab}+i\omega_{-b})\ \rho_{-b}+i\ V_{-b}(\rho_{--} - \rho_{bb})+i\ V_{+b}\rho_{-+} \tag{3.1e}$$

$$\dot{\rho}_{+b} = -(\gamma_{ab}+i\omega_{+b})\ \rho_{+b}+i\ V_{+b}(\rho_{++} - \rho_{bb})+i\ V_{-b}\rho_{+-} \quad , \tag{3.1f}$$

where $\gamma_{ab} = \frac{1}{2}(\gamma_a+\gamma_b)$, the natural linewidth, $\omega_{\pm b} = (E_a(M = \pm 1)-E_b)/\hbar$, the atomic transition frequencies, $\omega_{+-} = \omega_{+b}-\omega_{-b}$, and $V_{ij} = H'_{ij}/\hbar$. The perturbation V_{ij} is, naturally, calculated by using the value of the electric field at the location of the given atom. Thus if the atom's position at time t_o is $\bar{r}_o$, then at time t it is located at $\bar{r} = \bar{r}_o + \bar{v}(t-t_o)$. Thus

$$V_{ij} = V_{ij}(\bar{r}_o + \bar{v}(t-t_o),\ t) \quad . \tag{3.2}$$

Equations 3.1a-f can be integrated. We denote the density matrix element ρ_{ij} of an atom excited to the level q , at time $t = t_o$, and position $\bar{r} = \bar{r}_o$ with velocity $\bar{v}$, by $\rho_{ij}^{(q)}\ (\bar{r}_o,\ t_o,\ \bar{v},\ t)$ which will at time for simplicity be abbreviated as $\rho_{ij}^{(q)}(t,\ t_o)$. Since $\rho_{ij}^{(q)} = 1$ if $i = j = q$ and zero otherwise we get for a typical member of the density matrix

$$\rho_{--}^{(-)}(t,\ t_o) = e^{-\gamma_a(t-t_o)} + i \int_{t_o}^{t} dt'\ e^{\gamma_a(t'-t)}$$

$$\times\Big[\rho_{-b}(t',t_o)V^*_{-b}(\bar{r}+\bar{v}(t'-t_o),\ t') - \rho_{b-}(t',t_o)V_{-b}(\bar{r}_o+\bar{v}(t'-t_o),t')\Big] \ . \tag{3.3}$$

It will be found convenient to express the V's in a form such that they are functions of t, t', $\bar{r}$ and $\bar{v}$ but not of $\bar{r}_o$, t_o. Since $\bar{r}_o = \bar{r}-v(t-t_o)$

$V_{ij}(t', \bar{r}_o + \bar{v}(t'-t_o)) = V_{ij}(t', \bar{r} - \bar{v}(t-t'))$. Then after subtracting ρ_{bb} from ρ_{--} and substituting the expression for $\rho_{\pm b}$ and ρ_{+-} into the resulting equations we obtain the following set of integral equations

$$\rho_{--}^{(-)}(t,t_o)-\rho_{bb}^{(-)}(t,t_o) = e^{-\gamma_a(t-t_o)} - \int_{t_o}^{t} dt' \int_{t_o}^{t'} dt'' \left[e^{\gamma_a(t'-t)} + e^{\gamma_b(t'-t)} \right]$$

$$\times e^{(\gamma_{ab}+i\omega_{-b})(t''-t')} V_{b-}(t', \bar{r} - \bar{v}(t-t'))\, V_{-b}(t'', \bar{r} - \bar{v}(t-t''))$$

$$\times \left[\rho_{--}^{(-)}(t'',t_o) - \rho_{bb}^{(-)}(t'',t_o) \right] - \int_{t_o}^{t} dt' \int_{t_o}^{t'} dt''\, e^{\gamma_b(t'-t)} e^{(\gamma_{ab}+i\omega_{+b})(t''-t')}$$

$$\times V_{b+}(t', \bar{r} - \bar{v}(t-t'))V_{+b}(t'', \bar{r} - \bar{v}(t-t''))\left[\rho_{++}^{(-)}(t',t_o)-\rho_{bb}^{(-)}(t'',t_o) \right]$$

$$- \int_{t_o}^{t} dt' \int_{t}^{t'} dt'' \left[e^{\gamma_a(t'-t)} + e^{\gamma_b(t'-t)} \right] e^{(\gamma_{ab}+i\omega_{-b})(t''-t')} V_{b-}(t', \bar{r} - \bar{v}(t-t'))$$

$$\times V_{+b}(t'',\bar{r}-\bar{v}(t-t''))\rho_{-+}^{(-)}(t'',t_o) - \int_{t_o}^{t} dt' \int_{t_o}^{t'} dt''\, e^{\gamma_b(t'-t)} e^{(\gamma_{ab}+i\omega_{+b})(t''-t')}$$

$$\times V_{b+}(', \bar{r} - \bar{v}(t-t'))\, V_{-b}(t'', \bar{r} - \bar{v}(t-t''))\rho_{+-}^{(-)}(t'',t_o) + \text{complex conj.}$$

(3.4)

$$\rho_{++}^{(-)}(t,t_o) - \rho_{bb}^{(-)}(t,t_o) = -\int_{t_o}^{t} dt' \int_{t_o}^{t'} dt'' \left[e^{\gamma_a(t'-t)} + e^{\gamma_b(t'-t)} \right]$$

$$\times e^{(\gamma_{ab}+i\omega_{+b})(t''-t')} V_{b+}(t', \bar{r} - \bar{v}(t-t')V_{+b}(t'', \bar{r} - \bar{v}(t-t''))$$

$$\times \left[\rho_{++}^{(-)}(t'', t_o) - \rho_{bb}^{(-)}(t'',t_o)\right] - \int_{t_o}^{t} dt' \int_{t_o}^{t'} dt''\, e^{\gamma_b(t'-t)} e^{(\gamma_{ab}+i\omega_{-b})(t''-t')}$$

$$\times V_{b-}(t', \bar{r} - \bar{v}(t-t')\; V_{-b}(t'', \bar{r} - \bar{v}(t-t'')\left[\rho_{--}^{(-)}(t'',t_o) - \rho_{bb}^{(-)}(t'',t_o)\right]$$

$$- \int_{t_o}^{t} dt \int_{t_o}^{t'} dt'' \left[e^{\gamma_a(t'-t)} + e^{\gamma_b(t'-t)} \right] e^{(\gamma_{ab}+i\omega_{+b})(t''-t')} V_{b+}(t', \bar{r}-\bar{v}(t-t')$$

$$\times V_{-b}(t'', \bar{r}-\bar{v}(t-t'')\rho_{+-}^{(-)}(t'',t_o) - \int_{t_o}^{t} dt' \int_{t_o}^{t'} dt'' e^{\gamma_b(t'-t)} e^{(\gamma_{ab}+i\omega_{-b})(t''-t')}$$

$$\times V_{b-}(t', \bar{r}-\bar{v}(t-t')\; V_{+b}(t'', \bar{r}-\bar{v}(t-t''))\rho_{-+}^{(-)}(t'',t_o) + \text{complex conj.}$$

$$(3.5)$$

$$\rho_{+-}^{(-)}(t,t_o) = -\int_{t_o}^{t} dt' \int_{t_o}^{t'} dt'' \, e^{(\gamma_a+i\omega_{+-})(t'-t)} e^{(\gamma_{ab}+i\omega_{+b})(t''-t')}$$

$$\times V_{b-}(t',\bar{r}-\bar{v}(t-t')) \, V_{+b}(t'',\bar{r}-\bar{v}(t-t'')) \left[\rho_{++}^{(-)}(t'',t_o) - \rho_{bb}^{(-)}(t'',t_o)\right]$$

$$-\int_{t_o}^{t} dt' \int_{t_o}^{t'} dt'' \, e^{(\gamma_a+i\omega_{+-})(t'-t)} e^{(\gamma_{ab}-i\omega_{-b})(t''-t')} V_{+b}(t',\bar{r}-\bar{v}(t-t'))$$

$$\times V_{b-}(t'',\bar{r}-\bar{v}(t-t''))\left[\rho_{--}^{(-)}(t'',t_o)-\rho_{bb}^{(-)}(t'',t_o)\right]-\int_{t_o}^{t} dt' \int_{t_o}^{t'} dt'' e^{(\gamma_a+i\omega_{+-})(t'-t)}$$

$$\times\left[e^{(\gamma_{ab}+i\omega_{+b})(t'-t)} V_{b-}(t'',\bar{r}-\bar{v}(t-t'))V_{-b}(t'',\bar{r}-\bar{v}(t-t''))+e^{(\gamma_{ab}-i\omega_{-b})(t'-t)}\right.$$

$$\left.\times V_{+b}(t',\bar{r}-\bar{v}(t-t'))V_{b+}(t'',\bar{r}-\bar{v}(t-t''))\right] \rho_{+-}^{(-)}(t'',t_o) \,, \qquad (3.6)$$

$$\rho_{-b}^{(-)}(t,t_o) = i\int_{t_o}^{t} dt' e^{(\gamma_{ab}+i\omega_{-b})(t'-t)} V_{-b}(t',\bar{r}-\bar{v}(t-t'))\left[\rho_{--}^{(-)}(t',t_o)\right.$$

$$\left.-\rho_{bb}^{(-)}(t',t_o)\right]+i\int_{t_o}^{t} dt' \, e^{(\gamma_{ab}+i\omega_{-b})(t'-t)} V_{+b}(t',\bar{r}-\bar{v}(t-t'))\rho_{-+}^{(-)}(t',t_o)$$

$$(3.7)$$

$$\rho_{+b}^{(-)}(t,t_o) = i\int_{t_o}^{t} dt' \, e^{(\gamma_{ab}+i\omega_{+b})(t'-t)} V_{+b}(t',\bar{r}-\bar{v}(t-t')) \Big[\rho_{++}^{(-)}(t',t_o)$$

$$- \rho_{bb}^{(-)}(t',t_o)\Big] + i\int_{t_o}^{t} dt' \, e^{(\gamma_{ab}+i\omega_{+b})(t'-t)} V_{-b}(t',\bar{r}-\bar{v}(t-t')) \, \rho_{+-}^{(-)}(t',t_o), \tag{3.8}$$

and similar sets of equations for atoms initially in the M = +1 upper and the J = 0 lower state. The only difference is that in the first case the $e^{-\gamma_a(t-t_o)}$ term appears in front of the integrals in the equation for $\rho_{++}^{(+)} - \rho_{bb}^{(+)}$, while in the second case $e^{-\gamma_b(t-t_o)}$ appears in the equations for both $\rho_{++}^{(b)} - \rho_{bb}^{(b)}$ and $\rho_{--}^{(b)} - \rho_{bb}^{(b)}$.

3.4 Formal Solutions for the Medium

To obtain the macroscopic solutions we must sum up over all the relevant atoms, that is over all atoms which at some time are excited to any one of the levels we are considering. The density matrix for the medium is thus given by

$$\rho(\bar{r},t) = \sum_q \int \rho^{(q)} (\bar{r}_o,\, t_o,\, \bar{v},\, t) \, \lambda(r_o,t_o) \, W(\bar{v}) \, d^3r_o dt_o \quad . \tag{3.9}$$

The t_o integral is from t_o to ∞, the space integral over the volume of the medium, and the $\bar{v}$ integral over all velocity space. We also define

$$\rho^{(q)}(\bar{r},t,\bar{v}) = \int \rho^{(q)} (\bar{r}_o,\, t_o,\, \bar{v},\, t), \, \lambda_q(\bar{r}_o,t_o) \, W(\bar{v}) \, d^3r_o dt_o, \tag{3.10}$$

and $\rho(\bar{r}, t, v) = \sum_q \rho^{(q)}(\bar{r}, t, \bar{v})$. (3.11)

In the description of the electromagnetic field we have already specified variation in the z direction (along the length of the tube) only. Provided that λ_i is independent of x and y, that is to say the excitation is uniform across the tube, we can eliminate the x,y part of both the space and velocity integrals. We simply replace $\bar{r}$ and $\bar{r}_o$ with z and z_o respectively and use $v_z = v$ in place of $\bar{v}$.

Finally it is necessary to make one more assumption. This is that the quantity $\lambda_q(z_o, t_o)\, W(v)$ the number of atoms excited to the q level varies so slowly compared to the "interaction interval" $1/\gamma_a$, $1/\gamma_b$ and with distance that it can be evaluated at $z_o = z$, $t_o = t$. Interchanging the orders of integration such that

$$\int dz_o \int_{-\infty}^{t} dt_o \int_{t_o}^{t} dt' \int_{t_o}^{t'} dt'' \rightarrow \int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt'' \int_{-\infty}^{t''} dt_o \int dz_o ,$$

we note that neither the V's nor the exponentials in the integrals of equations 3.4-3.8 are functions of z_o, t_o and the integration over these variables is trivial. Since

$$\int_{-\infty}^{t} dt_o \; e^{-\gamma_{a,b}(t-t_o)} = \frac{1}{\gamma_{a,b}} , \qquad (3.12)$$

we get after using 3.9, defining new variables $t_1 = t-t'$ and $t_2 = t'-t''$ and finally performing the summation over q

$$N_{-b}(z,t,v) = N^{o}_{-b}W(v) - \int_{o}^{\infty} dt_1 \int_{o}^{\infty} dt_2 \left[e^{-\gamma_a t_1} + e^{-\gamma_b t_1} \right] e^{-(\gamma_{ab}+i\omega_{-b})t_2}$$

$$\times V_{b-}(t-t_1,\ z-vt_1)\ V_{-b}(t-t_1-t_2,\ z-v(t_1+t_2))N_{-b}(z,t-t_1-t_2,\ v)$$

$$-\int_{o}^{\infty} dt_1 \int_{o}^{\infty} dt_2 e^{-\gamma_b t_1} e^{-(\gamma_{ab}+i\omega_{+b})t_2} V_{b+}(t-t_1,z-vt_1)V_{+b}(t-t_1-t_2,z-v(t_1+t_2)$$

$$\times N_{+b}(z,t-t_1-t_2,v) - \int_{o}^{\infty} dt_1 \int_{o}^{\infty} dt_2 \left[e^{-\gamma_a t_1} + e^{-\gamma_b t_1} \right] e^{-(\gamma_{ab}+i\omega_{-b})t_2}$$

$$\times V_{b-}(t-t_1,\ z-t_1)\ V_{+b}(t-t_1-t_2,\ z-v(t_1+t_2))\rho_{-+}(z,\ t-t_1-t_2,\ v)$$

$$-\int_{o}^{\infty} dt_1 \int_{o}^{\infty} dt_2\ e^{-\gamma_b t_1} e^{-(\gamma_{ab}+i\omega_{+b})t_2} V_{b+}(t-t_1,z-vt_1)V_{-b}(t-t_1-t_2,z-v(t_1+t_2))$$

$$\times \rho_{+-}(z,t-t_1-t_2,v) + \text{complex conj.} \qquad (3.13)$$

$N_{+b}(z,t,v)$ = same as $N_{-b}(z,t,v)$, but with + and - interchanged in all subscripts,

$$\rho_{+-}(z,t,v) = -\int_0^\infty dt_1 \int_0^\infty dt_2\, e^{-(\gamma_a+i\omega_{+-})t_1} V_{b-}(t-t_1,\, z-vt_1)$$

$$\times V_{+b}(t-t_1-t_2, z-v(t_1+t_2))\, e^{-(\gamma_{ab}+i\omega_{+b})t_2} N_{+b}(z,t-t_1-t_2,v) - \int_0^\infty dt_1 \int_0^\infty dt_2$$

$$\times e^{-(\gamma_a+i\omega_{+-})t_1} V_{+b}(t-t_1,z-vt_1)\, V_{b-}(t-t_1-t_2,z-v(t_1+t_2))\, e^{-(\gamma_{ab}-i\omega_{-b})t_2}$$

$$\times N_{-b}(z,t-t_1-t_2,v) - \int_0^\infty dt_1 \int_0^\infty dt_2\, e^{-(\gamma_a+i\omega_{+-})t_1} V_{b-}(t-t_1,z-vt_1)$$

$$\times V_{-b}(t-t_1-t_2,z-vt_1+t_2)\, e^{-(\gamma_{ab}+i\omega_{+b})t_2} \rho_{+-}(z,t-t_1-t_2,v)$$

$$-\int_0^\infty dt_1 \int_0^\infty dt_2\, e^{-(\gamma_a+i\omega_{+-})t_1} V_{+b}(t-t_1,z-vt_1) V_{b+}(t-t_1-t_2,z-v(t_1+t_2))$$

$$\times e^{-(\gamma_{ab}-i\omega_{-b})t_2} \rho_{+-}(z,t-t_1-t_2,\, v)\ , \qquad (3.14)$$

and finally,

$$\rho_{+b}(z,t,v) = i\int_0^\infty dt_3\, e^{-(\gamma_{ab}+i\omega_{+b})t_3} V_{+b}(t-t_3,z-vt_3)\, N_{+b}(z,t-t_3,v)$$

$$+ i\int_0^\infty dt_3\, e^{-(\gamma_{ab}+i\omega_{+b})t_3} V_{-b}(t-t_3,z-vt_3)\, \rho_{+-}(z,t-t_3,v)\ . \qquad (3.15)$$

$\rho_{-b}(z,t,v)$ = same as $\rho_{+b}(z,t,v)$ but with + and - interchanged in all subscripts.

In the above equations $N_{\pm b}(z,t,v) = \rho_{\pm\pm}(z,t,v)-\rho_{bb}(z,t,v)$ and $N^{o}_{\pm b} = \frac{\lambda_{\pm}}{\gamma_a} - \frac{\lambda_b}{\gamma_b}$ are the excitation densities relative to the + and - sublevels respectively. We note that the population inversion densities at any point in the amplifier are proportional to the excitation densities at the same point. This is a consequence of the fact that although we are considering moving atoms, the excited atoms travel only a very short distance during the interaction time γ_a, γ_b. Note also that while the space coordinate of $N_{\pm b}$ and ρ_{+-} on the RHS of equations 3.13-3.15 has been, for convenience, written as z it is actually $z-v(t_1+t_2)$ or $z-vt_3$ same as the space coordinate of the V's appearing alongside.

These are the formal solutions, actually a set of coupled integral equations, for the macroscopic density matrix. In the subsequent chapters we will substitute specific expressions for the perturbation matrix V for various types of electromagnetic fields. Then, after finding approximate solutions and using the formula $\overline{P} = \text{Trace}\,(\rho\overline{p})$, where ρ is now the macroscopic density matrix, and finally performing the v integration we will find the polarization of the medium. First, however, we will examine the integral equations and draw some qualitative conclusions regarding the interaction between two waves of various polarizations in a laser medium which has an axial magnetic field.

CHAPTER FOUR

INTERACTION OF WAVES OF VARIOUS POLARIZATIONS IN A LASER MEDIUM

4.1 Introduction

In this chapter we examine qualitatively the type of nonlinear effects produced by two optical frequency electromagnetic waves in a laser amplifier which may have a nonzero axial magnetic field. In particular we are interested in predicting how the interactions depend on the polarization states of the two signals. The integral equations of the previous chapter, derived for the simple $J = 1 \rightarrow J = 0$ model will be utilized together with physical arguments. In the subsequent chapters quantitative calculations of these effects will be performed and the results checked against the predictions made here.

4.2 Saturation

The most obvious nonlinear effect is saturation of the gain. Population inversion of the medium means the existence of gain for waves resonant with the inverted transition. The inversion is accomplished by some sort of pumping process and in the absence of fields has the value $N^o_{ij} = \lambda_i \gamma_i^{-1} - \lambda_j \gamma_j^{-1}$ as defined in the previous chapter. For very weak fields the population inversions stay essentially constant at this value and the gain is not influenced by the field intensities.

This is the linear case which will be briefly examined in the following chapter. For somewhat stronger field intensities, however, the number of stimulated emissons is large enough to spoil the population inversions densities. On Figure 2 is shown schematically the $J = 1 \rightarrow J = 0$ transition interacting with an electromagnetic field $\overline{E}(z,t)$. The polarization vector of the expectation value of the dipole moment is indicated for each transition. The left and right circular components of the total field interact with different transitions which terminate on a common level. The non-linear saturation effects are several kinds. Each component, by inducing stimulated emissions, empties the upper and fills up the lower level of its own transition thus spoiling the corresponding population inversion. This is shown mathematically by the first integral in each of the equations for N_{+b}, N_{-b}. The two field

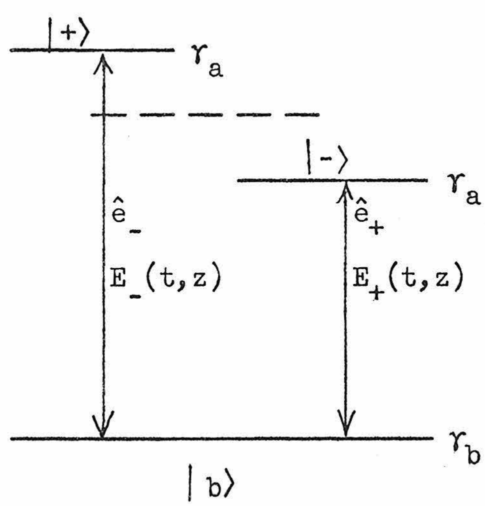


Figure 2.

components also influence each other by filling up the common lower level and thus spoiling each other's population inversion. This may be called a mutual saturation effect and is indicated mathematically by the second integral in each of the equations for N_{+b} , N_{-b} . In addition to these another, somewhat less obvious, process also takes place. An atom initially in the M = +1 state may interact simultaneously with both left and right circularly polarized components and transfer to the M = -1 state via this double quantum interaction. The reverse can of course also happen. This is indicated by the equation for ρ_{+-} . A finite ρ_{+-} signifies coherence of the magnetic sublevels which contributes to the nonlinear polarization or, in other words, the gains of the opposite circular components are influenced by the transfer between the two upper levels. The magnitude and precise nature of the influence is not obvious from the integral equations but will be calculated in detail in the subsequent chapters. Additional terms in the integral equations show higher order effects of the three processes described above. For example, the coherence of the upper levels influences the two population inversion densities and ρ_{+-} itself saturates as is shown by the last two integrals in its equation. These higher order terms will become important at high field intensities. It can be concluded that gain saturation in the laser amplifier will be caused by the self saturation of the circularly polarized components, the mutual saturation between opposite circular components, due to the common level, and finally by coherent double quantum interactions. Regardless of whether the polarization states of the input fields are circular, linear or elliptical these are the physical processes that occur and the fields should be defined so that quantitative calculations put them in evidence.

4.3 Combination Tone Generation

From the form of the electromagnetic field, given in equation 2.4 it is evident that only first, third, fifth etc. powers of the field contain frequencies close to the optical frequency ν around which the input field frequencies are distributed. It is also evident, however, that for more than one (essentially monochromatic) signal these powers contain frequencies that are not in the input. For example, for $E(z,t)$ containing two frequencies ν_1 and ν_2, $[E(z,t)]^3$ contains terms oscillating at $2\nu_2 - \nu_1$ and at $2\nu_1 - \nu_2$, $[E(z,t)]^5$ terms at $3\nu_2 - 2\nu_1$ and $3\nu_1 - 2\nu_2$, etc. There is thus the possibility of combination tone or sideband generation in the laser medium. The integral equations 3.13- 3.15 contain these new frequency components, since the interaction matrix V is the dot product of the dipole moment operator with the total electromagnetic field.

Physically, combination tone generation is caused by a coherent modulation of the population inversion densities. If a given transition is acted on by two fields of frequencies ν_1 and

ν_2 an atom in either the upper or lower state of that transition sees a field with beats at the difference frequency $\nu_1 - \nu_2$. The beats travel through the medium with the propagation constant $\Delta k = k_1 - k_2$. The time between successive field maxima for the atom is

$$\tau = \frac{\text{beat wavelength}}{\text{group velocity}} \simeq \frac{2\pi}{c\Delta k} \simeq \frac{2\pi}{\Delta\nu} \quad . \tag{4.1}$$

If τ is longer than the decay time of the levels $1/\gamma_a$, $1/\gamma_b$ the population inversion will follow the beating of the field. The gain being proportional to the population inversion, it will be modulated at the difference frequency $\Delta\nu$. This results in parametric generation of frequencies at $\nu_1 \pm n\Delta\nu$ and $\nu_2 \pm n\Delta\nu$. The essential condition for this process to take place is two (or more) fields of different frequency acting on a given transition. If different transitions are involved no modulation and therefore no combination tone generation occurs. In the integral equations this is seen by examining the first two integrals in the equation for N_{-b} (or N_{+b}) . If V_{-b} and V_{+b} each contain only a single frequency, even if the frequency of one is different from the other, these integrals are constant in time (except for frequency doubling, at 2ν which however is strongly antiresonant with $\omega_{\pm b}$ and can be neglected to a very high accuracy). On the other hand, if either

V_{+b} or V_{-b} (or both) contains two frequencies ν_1 and ν_2 there will be terms oscillating at $\Delta\nu = \nu_1 - \nu_2$ in both population inversions and combination tone generation results. We are now in the position to examine specific cases. In the subsequent discussion the term "two fields" means a field having two essentially monochromatic components at frequencies ν_1 and ν_2 .

1) Two fields of the same circular polarization. Since only one polarization (say $E_-(z,t)$) is present only one transition is involved (except of course spontaneous emission in the other transition and its subsequent amplification which is neglected here). The model reduces to a two level one (or to several non interacting two level systems for higher J values). The integral equations greatly simplify since $\rho_{+-} = 0$ and $N_{-b} = N^{o}_{-b}$. There is both mutual saturation between the two fields and combination tone generation. This is identical to the two level scalar field problem treated in detail by other authors (5 ,6,7) and will not therefore be considered in this work.

2) Two opposite circularly polarized fields. There is one field acting on each transition. Because of the common level of the two transitions there is mutual saturation interaction between the two fields and the double quantum processes also cause additional gain saturation as discussed in section 4.2. There is, however, no side-band production since no modulation of either of the population inversion densities occurs. While it might seem that the double quantum

interactions could cause combination tone generation since both of the fields are involved, this is not the case. If we substitute ρ_{+-} into the equation for ρ_{+b} interaction matrix combinations of the type $V_{-b}V_{b-}V_{+b}$ result, the same as from the contribution of N_{-b} . The first two cause no modulation and the third contributes the optical frequency variation at the frequency ν_- only. This will be confirmed in the calculations of the next chapter. We can in fact repeat the statement made by physical reasoning that only by modulation of the inversion densities can sideband generation occur and this can take place only when two fields act on a given transition. The same holds for higher J values of the levels and of course for the limiting case of zero magnetic field too, the only condition being that collision induced mixing of the sublevels be negligible. This condition was assumed in the derivation of the integral equations.

3) Two linearly polarized fields. A linearly polarized field can be treated as the superposition of two oppositely circularly polarized fields. For this case each of the transitions is acted on by two fields of different frequency. (Both E_- and E_+ contain two frequency components from the two linearly polarized waves). Both saturation effects and combination tone generation are present. Some exceptions to the latter should be pointed out. For the $J = 1 \rightarrow J = 0$ transition there exists an orthogonal combination

of the $M = +1$ and $M = -1$ upper states that give perpendicular linearly polarized transitions to the common $J = 0$ lower level. Letting

$$\frac{1}{\sqrt{2}}(\langle a, 1, +1| - \langle a, 1, -1|) = \langle a, \hat{x}| \text{ , and} \tag{4.2a}$$

$$\frac{i}{\sqrt{2}}(\langle a, 1, +1| + \langle a, 1, -1|) = \langle a, \hat{y}| \text{ ,} \tag{4.2b}$$

the matrix elements of the dipole operator $\bar{p}$ are

$$\langle a,\hat{x}|\bar{p}|b\rangle = \frac{1}{2}(\hat{e}_x+i\hat{e}_y)\langle a\|p\|b\rangle \frac{1}{\sqrt{3}} - \frac{1}{2}(-\hat{e}_x+i\hat{e}_y)\langle a\|p\|b\rangle\frac{1}{\sqrt{3}}$$

$$= \hat{e}_x \frac{1}{\sqrt{3}}\langle a\|p\|b\rangle \tag{4.3a}$$

$$\langle a,\hat{y}|\bar{p}|b\rangle = \hat{e}_y \frac{1}{\sqrt{3}}\langle a\|p\|b\rangle \text{ .} \tag{4.3b}$$

For zero magnetic field then these are equally good states. Consequently two fields of perpendicular linear polarization can be treated as interacting with two different transitions and no combination tones are generated. The apparent sideband production using the magnetic sublevels as a model <u>must be such as to exactly cancel</u>. The same situation exists for the $J = 0 \rightarrow J = 1$ and for the $J = 1 \rightarrow J = 1$ transitions (the latter can be made up of the superposition of the two former). For higher J values of the levels such combinations of the substates do not exist, p_x , p_y , p_z are not diagonal in any representation of the substates of the two energy levels.

Therefore, for laser transitions with either J_a or J_b larger than unity combination tones are generated even for perpendicular linearly polarized signals in zero magnetic field.

4) Two elliptically polarized fields. This is exactly the same as the previous case except the amplitudes of the circularly polarized components are unequal. There is combination tone generation, except for the three "special" transitions for which no such effect exists in zero magnetic field and two orthogonal elliptically polarized fields.

5) Three circularly polarized fields. Two of the fields have the same circular polarization and therefore combination tones must exist. It is seen from the integral equations that both population inversion densities are modulated. Sidebands separated by $\nu_1 - \nu_2$ are produced not only to the two identically polarized fields but to the one oppositely polarized signal as well. Otherwise the results are qualitatively not very different from the previous two cases and will not be treated here. More complicated combinations of fields likewise will not be considered.

4.3 Other Nonlinear Effects

Harmonic generation which is insignificant in a gas laser unless the harmonics coincide with other population inverted transitions will not be considered in this work. There exists, however, still another nonlinear effect that can under some circumstances be significant. The nonzero value of ρ_{+-} indicates the presence of magnetic dipole and electric quadrupole radiation at the frequencies $\nu_{i-} - \nu_{j+}$. The induced oscillating dipole, for example, is equal to $\rho_{+-}m$ where m is the matrix element of the magnetic dipole moment operator between the substates $M = \pm 1$ (For more complicated transitions the various matrix element between substates with $\Delta M = 2$ must be summed). ρ_{+-} depends on products of field amplitudes and while m is usually small, for strong fields the generation of power at the difference frequencies can become significant. A theory of this optical frequency mixing using a general three level system has been given by Javan and Szőke (45). We shall not include it in the calculations that follow.

CHAPTER FIVE

TWO OPPOSITE CIRCULARLY POLARIZED FIELDS

5.1 Introduction

In this chapter we study the nonlinear effects that are present when two opposite circularly polarized traveling waves interact in the laser amplifier. As it was pointed out in the previous chapter there is no combination tone production for this case and the induced polarization of the medium oscillates only at the frequencies of the input waves. This will greatly facilitate the handling of the field equations 2.14a,b. The integral equations 3.13-3.15 are the basis to our calculations. These are developed further by substituting into the interaction matrix V the specific form of the electromagnetic field; this is done in section 5.2. Two types of solutions are obtained from the resultant equations. In section 5.3 we solve the integral equations by iteration and obtain for the polarization the first order, linear, result and the third (lowest) order nonlinear correction to it. This approach is equivalent to Lamb's method of expanding the polarization in a perturbational series. The resulting solutions are straightforward to handle and complete generality in the frequencies and the atomic parameters is possible. Unfortunately, as we shall see later, this approach is only valid for weak fields and the range of applicability of the results is limited. In section

5.4 we develop another method and solve the integral equations to some approximation such that the results are valid for arbitrarily strong fields. To be able to do that we must forgo some of the generality of the iterative solutions. In many ways the two types of solutions will be complementary to each other, both being necessary for a complete description of the nonlinear phenomena.

We will most of the time be considering the Doppler broadened case, i.e., a Maxwellian velocity distribution, but solutions for stationary atoms will also be given for purposes of comparison. The special case when the frequencies and the field amplitudes are equal corresponds to a single linearly polarized signal. This will be treated separately in sections 5.3.4 and 5.4.4. for weak and strong fields respectively.

5.2 The Integral Equations

For the case of two opposite circularly polarized travelling waves in the laser the electromagnetic field is

$$\bar{E}(t,z) = E_- \frac{1}{\sqrt{2}}\left[\hat{e}_x \cos(k_- z-\nu_- t+\varphi_-) - \hat{e}_y \sin(k_- z-\nu_- t+\varphi_-)\right]$$

$$+ E_+ \frac{1}{\sqrt{2}}\left[-\hat{e}_x \cos(k_+ z-\nu_+ t+\varphi_+) - \hat{e}_y \sin(k_+ z-\nu_+ t+\varphi_+)\right] . \qquad (5.1)$$

The field amplitudes E_+, E_- and the frequencies ν_+, ν_- are in general different. Since $-V = \bar{p} \cdot \bar{E}/\hbar$

$$V_{-b} = (\langle a\|p\|b\rangle/\sqrt{3}\hbar)\,\frac{1}{2}\left[E_- e^{+i(k_- z-\nu_- t+\varphi)} - E_+ e^{-i(k_+ z-\nu_+ t+\varphi_+)}\right] \tag{5.2a}$$

$$V_{+b} = (\langle a\|p\|b\rangle/\sqrt{3}\hbar)\,\frac{1}{2}\left[-E_- e^{-i(k_- z-\nu_- t+\varphi_-)} + E_+ e^{+i(k_+ z-\nu_+ t+\varphi_+)}\right] \tag{5.2b}$$

We can now substitute these into the integral equations 3.13-3.15. In doing so we make use of the last assumption discussed in section 3.3.1, namely that the amplitude and phase of the field seen by an atom during the interaction time remains constant. Thus, we freely replace $E_\pm(z-v(t_1+t_2),t-t_1-t_2)$ by $E_\pm(z,t)$ and $\varphi_\pm(z-v(t_1+t_2),t-t_1-t_2)$ by $\varphi_\pm(z,t)$, etc. In all subsequent calculations the slow z,t dependence of these quantitites will be understood and will not be written out explicitly. We get the following set of equations for the population inversion densities $N_{\pm b}(z,t,v) = \rho_{\pm\pm}(z,t,v)-\rho_{bb}(z,t,v)$ and the density matrix element ρ_{+-} :

$$N_{+b}(z,t,v) = N^o_{+b}W(v) - \frac{\gamma_a\gamma_b}{4}\,\frac{E_-^2}{E_o^2}\int_o^\infty dt_1 \int_o^\infty dt_2 \left[e^{-\gamma_a t_1} + e^{-\gamma_b t_1}\right]$$

$$e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_-+kv)]t_2}\, N_{+b}(z,t-t_1-t_2,v) - \frac{\gamma_a\gamma_b}{4}\,\frac{E_+^2}{E_o^2}\int_o^\infty dt_1 \int_o^\infty dt_2 e^{-\gamma_b t_1}$$

$$\times e^{-[\gamma_{ab}+i(\omega_{-b}-\nu_{+}+k_{+}v)]t_2} N_{-b}(z,t-t_1-t_2,v) - \frac{\gamma_a\gamma_b}{4}\frac{E_-E_+}{E_o^2}\int_o^\infty dt_1 \int_o^\infty dt_2$$

$$\times\Big\{\Big[e^{-[\gamma_a+i((\nu_--\nu_+)-(k_--k_+)v)]t_1} + e^{[\gamma_b+i((\nu_--\nu_+)+(k_--k_+)v)t_1}\Big]$$

$$\times e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_{+}+k_{+}v)]t_2} + e^{-[\gamma_b+i((\nu_--\nu_+)+(k_--k_+)v)]t_1}$$

$$\times e^{-[\gamma_{ab}-i(\omega_{-b}-\nu_{-}+k_{-}v)]t_2}\Big\} \rho_{+-}(z,t-t_1-t_2,v)\, e^{-i\Delta} + \text{c.c.}\,, \qquad (5.3)$$

where $\Delta = (k_--k_+)\, z-(\nu_--\nu_+)t+\varphi_--\varphi_+$;

$N_{-b}(z,t,v)$ = same as $N_{+b}(z,t,v)$ but with + and - interchanged in all subscripts ($\therefore\ \Delta \to -\Delta$) ; and

$$\rho_{+-}(z,t,v) = \Big\{-\frac{\gamma_a\gamma_b}{4}\frac{E_-E_+}{E_o^2}\Big[\int_o^\infty dt_1 \int_o^\infty dt_2\, e^{-[\gamma_a+i(\omega_{+-}-(\nu_--\nu_+)+(k_--k_+)v)]t_1}$$

$$\times e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_{-}+k_{-}v)]t_2} N_{+b}(z,t-t_1-t_2,v) + \int_o^\infty dt_1 \int_o^\infty dt_2$$

$$\times e^{-[\gamma_a+i(\omega_{+-}-(\nu_--\nu_+)+(k_--k_+)v)]t_1}\, e^{-[\gamma_{ab}-i(\omega_{-b}-\nu_{+}+k_{+}v)]t_2}$$

$$\times N_{-b}(z,t-t_1-t_2,v)]\Big\} e^{i\Delta} - \frac{\gamma_a\gamma_b}{4}\frac{E_+^2}{E_o^2}\int_o^\infty dt_1 \int_o^\infty dt_2\, e^{-[\gamma_a+i\omega_{+-}]t_2}$$

$$e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_{+}+k_{+}v)]t_2}\ \rho_{+-}(z,t-t_1-t_2,v)\ -\ \frac{\gamma_a\gamma_b}{4}\frac{E_-^2}{E_o^2}\int_o^\infty dt_1\int_o^\infty dt_2$$

$$e^{-[\gamma_a+i\omega_{+-}]t_2}\ e^{-[\gamma_{ab}-i(\omega_{-b}-\nu_{-}+k_{-}v)]t_2}\ \rho_{+-}(z,t-t_1-t_2,v). \qquad (5.4)$$

Finally, in terms of the above quantities, after using 2.9, the equations for the macroscopic polarization are:

$$P_-(z,t,v) = -i\,\frac{|\langle a\|p\|b\rangle|^2}{3\hbar}\left\{E_-\int_o^\infty dt_3\ e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_{-}+k_{-}v)]t_3}N_{+b}(z,t-t_3,v)\right.$$

$$\left. +\ E_+\int_o^\infty dt_3\ e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_{+}+k_{+}v)]}\ \rho_{+-}(z,t-t_3,v)e^{-i\Delta}\right\}\ , \qquad (5.5)$$

and $P_+(z,t,v)$ = same with + and - interchanged in all subscripts.

where $E_o^2 = 3\hbar^2\gamma_a\gamma_b/|\langle a\|p\|b\rangle|^2$, the saturation field intensity, and c. c. indicates the complex conjugate, for every integral.

5.3 The Iterative Solutions

The simplest approximate solutions of the integral equations 5.3-5.5 we get by iteration. It is evident from the form of these expressions that this method gives valid results only as long as $E_\pm^2/E_o^2 \lesssim .5$. With this limitation kept in mind we proceed to calculate the polarization of the medium to third order in the fields. As mentioned in chapter three two kinds of velocity distribution will be considered:

1.) $W(v) = (1/\sqrt{\pi}u) \exp(-v^2/u^2)$, Maxwellian velocity profile, where $u^2 = 2kT/M$, corresponding to normal Doppler broadening;

2.) $W(v) = \delta(v)$, corresponding to stationary or very slow moving atoms. The second case is calculated separately for convenience even though it is actually a limiting case of the first as $v \to 0$.

From equations 5.3 and 5.4 to the zeroeth order:

$$N_{+b}(z,t,v) = N^{o}_{+b}\, W(v) \tag{5.6a}$$

$$N_{-b}(z,t,v) = N^{o}_{-b}\, W(v) \tag{5.6b}$$

$$\rho_{+-}(z,t,v) = 0 \quad . \tag{5.6c}$$

That is, the population inversions retain their zero field value. Then from 5.5 the first order, linear, polarization due to atoms of velocity v is

$$P^{(1)}_{-}(v) = -i \frac{|\langle a\|p\|b\rangle|^2}{3\hbar} N_{+b} E_{-} \frac{W(v)}{\gamma_{ab} + i(\omega_{+b} - \nu_{-} + k_{-}v)} , \tag{5.7a}$$

$$P^{(1)}_{+}(v) = -i \frac{|\langle a\|p\|b\rangle|^2}{3\hbar} N_{-b} E_{+} \frac{W(v)}{\gamma_{ab} + i(\omega_{-b} - \nu_{+} + k_{+}v)} , \tag{5.7b}$$

where $P_{\pm} = P_{c\pm} - iP_{s\pm}$ as defined by equation 2.5.

We arrive at the second order correction to $N_{\pm}(z,t,v)$ and to ρ_{+-} by using the zeroeth order values in equations 5.3, 5.4.

$$N_{+b}^{(2)}(z,t,v) = -\frac{\gamma_a\gamma_b}{4} N_{+b}^{o} \frac{E_-^2}{E_o^2}\left[\frac{1}{\gamma_a}+\frac{1}{\gamma_b}\right]\frac{W(v)}{\gamma_{ab}+i(\omega_{+b}-\nu_-+k_-v)}$$

$$-\frac{\gamma_a\gamma_b}{4} N_{-b}^{o} \frac{E_+^2}{E_o^2}\frac{1}{\gamma_b}\frac{W(v)}{\gamma_{ab}+i(\omega_{-b}-\nu_++k_+v)} + \text{c. c.}$$

$$= -\frac{\gamma_{ab}}{2} N_{+b}^{o} \frac{E_-^2}{E_o^2}\frac{W(v)}{\gamma_{ab}+i(\omega_{+b}-\nu_-+k_+v)} - \frac{\gamma_a}{4} N_{-b}^{o} \frac{E_+^2}{E_o^2}\frac{W(v)}{\gamma_{ab}+i(\omega_{-b}-\nu_++k_+v)} + \text{c.c.} \tag{5.8a}$$

Similarly,

$$N_{-b}^{(2)}(z,t,v) = -\frac{\gamma_{ab}}{2} N_{-b}^{o} \frac{E_+^2}{E_o^2}\frac{W(v)}{\gamma_{ab}+i(\omega_{-b}-\nu_++k_+v)}$$

$$-\frac{\gamma_a}{4} N_{+b}^{o} \frac{E_-^2}{E_o^2}\frac{W(v)}{\gamma_{ab}+i(\omega_{+b}-\nu_-+k_-v)} + \text{c. c.} \tag{5.8b}$$

$$\rho_{+-}^{(2)}(z,t,v) = -\frac{\gamma_a\gamma_b}{4}\frac{E_-E_+}{E_o^2}\frac{1}{\gamma_a+i(\omega_{+-}-(\nu_--\nu_+)+(k_--k_+)v)}$$

$$\times\left[\frac{N_{+b}^{o}\,W(v)}{\gamma_{ab}+i(\omega_{+b}-\nu_-+k_-v)} + \frac{N_{-b}^{o}\,W(v)}{\gamma_{ab}-i(\omega_{-b}-\nu_++k_+v)}\right] e^{i\Delta} \;. \tag{5.8c}$$

And from 5.5 the third order correction to the macroscopic polarization is

$$P_-^{(3)}(v) = i\,\frac{|\langle a\|p\|b\rangle|^2}{3\hbar}\,W(v)\,\Big\{N^o_{+b}\,\frac{E_-^3}{E_o^2}\,\frac{\gamma_a}{4}\Big[\Big(\frac{1}{\gamma_{ab}+i(\omega_{+b}-\nu_-+k_-v)}\Big)^2$$

$$+\frac{1}{\gamma_{ab}+i(\omega_{+b}-\nu_-+k_-v)}\,\frac{1}{\gamma_{ab}-i(\omega_{+b}-\nu_-+k_-v)}\Big] + N^o_{-b}\,\frac{E_+^2\,E_-}{E_o^2}\,\frac{\gamma_a}{4}\,\frac{1}{\gamma_{ab}+i(\omega_{+b}-\nu_-+k_-v)}$$

$$\times\Big[\frac{1}{\gamma_{ab}+i(\omega_{-b}-\nu_++k_+v)}+\frac{1}{\gamma_{ab}-i(\omega_{-b}-\nu_++k_+v)}\Big]$$

$$+\frac{E_+^2\,E_-}{E_o^2}\,\frac{\gamma_a\gamma_b}{4}\,\frac{1}{\gamma_a+i(\omega_{+-}-(\nu_--\nu_+)+(k_--k_+)v)}\Big[\frac{N^o_{+b}}{[\gamma_{ab}+i(\omega_{+b}-\nu_-+k_-v)]^2}$$

$$+\frac{N^o_{-b}}{\gamma_{ab}-i(\omega_{-b}-\nu_++k_+v)}\,\frac{1}{\gamma_{ab}+i(\omega_{+b}-\nu_-+k_-v)}\Big]\Big\}\;, \qquad (5.9)$$

$P_+^{(3)}(z,v)$ = Same with + and - interchanged in all subscripts.
To calculate $P_\pm^{(1)}$ and $P_\pm^{(3)}$ we have to integrate over the velocities. As predicted in chapter four, there are no new frequency components, P_- and P_+ oscillate at ν_- and ν_+ only.

5.3.1 Maxwellian Velocity Distribution

This is the Doppler (or inhomogeneously) broadened case we usually encounter in a gas laser. To perform the integration over v we first make an important simplification. We let $k_- = k_+ = k$ in equations 5.7 and 5.9. This step is justifiable since $k_\pm$ occurs only in the fractions which are insensitive to a very small difference between k_+ and k_-. Since the quantity ku is related to the Doppler width by

$$ku = \frac{\nu}{nc} u \sim \omega \frac{u}{c} = 2\pi(\Delta f)_{Doppler}/2\sqrt{\ell n 2} ,$$

what we have done in effect is to neglect the slight difference in the Doppler width of the left and right circularly polarized transitions (Δku). The approximation is good as long as $\Delta ku \ll \gamma_a$. Since $\Delta k = k[(\Delta\nu/\nu)-(\Delta n/n)] \sim \Delta\nu/c$ this condition can be written as $\Delta\nu(u/c) \ll \gamma_a$. Taking $u \sim 6 \times 10^2$ m/sec gives $\Delta\nu \ll .5 \times 10^6 \gamma_a$. Thus even for γ_a as low as 10^6 the approximation is excellent for magnetic field splitting as high as 10 KMc or higher.

Dividing and multiplying the right hand side of equations 5.7a and 5.9 by ku we get, after defining $\xi = v/u$ ($\therefore dv = ud\xi$),

$$\frac{\nu}{2\epsilon_o c} P_-^{(1)+(3)} = \frac{\nu}{2\epsilon_o c} \int_{-\infty}^{+\infty} \left[P^{(1)}(v) + P^{(3)}(v)\right] dv = -i \frac{\nu |\langle a||p||b\rangle|^2}{6\epsilon_o \hbar cku} \frac{E_-}{\sqrt{\pi}} \int_{-\infty}^{+\infty} e^{-\xi^2}$$

$$\left\{ N^o_{+b} \frac{1}{a+i(x_-+\xi)} + \frac{E_-^2}{2E_o^2} a N^o_{+b} \left[\frac{1}{[a+i(x_-+\xi)]^2} + \frac{1}{a+i(x_-+\xi)} \frac{1}{a-i(x_-+\xi)} \right] \right.$$

$$+ \frac{E_+^2}{2E_o^2} AN^o_{-b} \left[\frac{1}{a+i(x_-+\xi)} \frac{1}{a+i(x_++\xi)} + \frac{1}{a+i(x_-+\xi)} \frac{1}{a-i(x_++\xi)} \right]$$

$$\left. + \frac{E_+^2}{2E_o^2} \frac{AB}{A+i[(\omega_{+-}-\Delta\nu)/2ku]} \left[\frac{N^o_{+b}}{[a+i(x_-+\xi)]^2} + \frac{N^o_{-b}}{a+i(x_-+\xi)} \frac{1}{a-i(x_++\xi)} \right] \right\} d\xi \tag{5.10}$$

where $a = \gamma_{ab}/ku$, $A = \gamma_a/2ku$, $B = \gamma_b/2ku$ ($A+B = a$), and $x_\pm = (\omega_\mp - \nu_\pm)/ku$.

Although we could continue to keep N^o_{+b} and N^o_{-b} different, indicating possible unequal pumping of the magnetic sublevels, for most of this work we shall for convenience assume $N^o_{+b} = N^o_{-b} = N_o$. It will be a very simple matter to restore the generality if necessary.

The integration in equation 5.10 can be carried out; the details of this somewhat tedious computation appear in Appendix I. The results are expressed in terms of the error function of complex argument, defined by $w(z) = \exp(-z^2)\ \mathrm{erfc}(-iz) = (2/\sqrt{\pi})\exp(-z^2) \int_z^\infty \exp(-t^2)dt$ (46). This is essentially the complex conjugate of Lamb's (5) "plasma dispersion function." The results are:

$$\frac{\nu}{2\epsilon_o c}P_-^{(1)+(3)} = i\alpha E_-\left\{w^*(x_o+y-\zeta+ia) - \frac{E_-^2}{2E_o^2}F_1(y,\zeta) - \frac{E_+^2}{2E_o^2}\left[F_2(y,\zeta)+F_3(y,\zeta)\right]\right\}, \tag{5.11}$$

where

$$F_1(y,\zeta) = a\left[\frac{2}{\sqrt{\pi}} - 2\Big(a+i(x_o+y-\zeta)\Big)w^*(x_o+y-\zeta+ia) + \frac{1}{a}\,\mathrm{Re}\ w^*(x_o+y-\zeta+ia)\right], \tag{5.12a}$$

$$F_2(y,\zeta) = A\left[\frac{i}{2(y-\zeta)}\Big(w^*(x_o+y-\zeta+ia) - w^*(x_o-y+\zeta+ia)\Big)\right.$$

$$\left. + \frac{1}{2[a+i(y-\zeta)]}\Big(w^*(x_o+y-\zeta+ia) + w(x_o-y+\zeta+ia)\Big)\right], \tag{5.12b}$$

$$F_3(y,\zeta) = \frac{AB}{A+i(y-\zeta)}\left[\frac{2}{\sqrt{\pi}} - 2\Big(a+i(x_o+y-\zeta)\Big)w^*(x_o+y-\zeta+ia)\right.$$

$$+ \frac{1}{2[a+i(y-\zeta)]} \Big(w^*(x_o+y-\zeta+ia) + w(x_o-y+\zeta+ia) \Big) \Big] \quad . \qquad (5.12c)$$

The following limiting case is also necessary:

$$\lim_{y-\zeta\to 0} \frac{i}{2(y-\zeta)} \Big(w^*(x_o+y-\zeta+ia) - w^*(x_o-y+\zeta+ia) \Big) = \frac{2}{\sqrt{\pi}} - 2(a+ix_o)\, w^*(x_o+ia)$$

(5.12d)

where $\alpha = N_o\sqrt{\pi\nu}\,|\langle a\|p\|b\rangle|^2/6\epsilon_o\hbar cku$, the small signal gain parameter,

$x_o = \big[(\omega_{+b}+\omega_{-b})-(\nu_+ + \nu_-)\big]/2ku$, the deviation of the mean frequency from line center,

$y = (\omega_{+b}-\omega_{-b})/2ku = \omega_{+-}/2ku$, the Zeeman shift or half the line separation,

$\zeta = (\nu_- - \nu_+)/2ku = \Delta\nu/2ku$, half the frequency separation of the input signals, all measured in units of ku. Thus $x_o+y-\zeta = x_- = (\omega_{+b}-\nu_-)/ku$ and $x_o-y+\zeta = x_+ = (\omega_{-b}-\nu_+)/ku$.

Similarly we get

$$\frac{\nu}{2\epsilon_o c} P_+^{(1)+(3)} = -i\alpha E_+\Big\{ w^*(x_o-y+\zeta+ia) - \frac{E_+^2}{E_o^2} F_1(-y,-\zeta)$$

$$- \frac{E_-^2}{E_o^2}\Big[F_2(-y,-\zeta) + F_3(-y,-\zeta)\Big]\Big\} \quad . \qquad (5.13)$$

The first term in both 5.11 and 5.13 is the linear result while the rest of the terms show the nonlinear effects to the lowest order. We shall discuss the physical significance of these results in section 5.3.3, where we substitute 5.13 into the electromagnetic field equations 5.6a,b , but first we derive the corresponding equations for a predominantly naturally broadened transition.

5.3.2 Stationary Atoms

For stationary (or very slow moving) atoms $W(v) = \delta(v)$. For this case the spontaneous emission line shape is Lorentzian and we speak of natural or homogeneous broadening. The integration over v is trivial, the delta function simply sets v = 0 everywhere. Then,

$$\frac{\nu}{2\epsilon_o c} P_-^{(1)+(3)} = i\alpha_o E_- \left\{ \frac{1-i(x_o'+y'-\zeta')}{1+(x_o'+y'-\zeta')^2} - \frac{E_-^2}{E_o^2} G_1(y',\zeta') - \frac{E_+^2}{E_o^2}\left[G_2(y',\zeta')+G_3(y',\zeta')\right]\right\} , \tag{5.14}$$

where $\alpha_o = N_o \, \nu |\langle a\|p\|b\rangle|^2 / 6\epsilon_o \hbar c \gamma_{ab}$ and

$$G_1(y',\zeta') = \frac{1-i(x_o'+y'-\zeta')}{1+(x_o'+y'-\zeta')^2} \, \frac{1}{1+(x_o'+y'-\zeta')^2} , \tag{5.15a}$$

$$G_2(y',\zeta') = \frac{\gamma_a}{2\gamma_{ab}} \, \frac{1-i(x_o'+y'-\zeta')}{1+(x'+y'-\zeta')^2} \, \frac{1}{1+(x_o'-y'+\zeta')^2} , \tag{5.15b}$$

$$G_3(y',\zeta') = \frac{\gamma_a/2\gamma_{ab}}{\gamma_a/2\gamma_{ab}+i(y'-\zeta')}\,\frac{1-i(x_o'+y'-\zeta')}{1+(x_o'+y'-\zeta')^2}$$

$$\frac{\gamma_b}{2\gamma_{ab}}\left[\frac{1-i(x_o'+y'-\zeta')}{1+(x_o'+y'-\zeta')^2}+\frac{1+i(x_o'-y'+\zeta')}{1+(x_o'-y'+\zeta')^2}\right] \quad . \qquad (5.15c)$$

The quantities x_o' , y' , ζ' are the same as the corresponding unprimed ones except measured in units of γ_{ab} (the natural linewidth parameter); e.g., $y' = (\omega_{+b}-\omega_{-b})/2\gamma_{ab}$, etc. In a similar manner,

$$\frac{\nu}{2\epsilon_o c}P_+^{(1)+(3)} = i\alpha_o E_+ \left\{\frac{1-i(x_o'-y'+\zeta')}{1+(x_o'-y'+\zeta')^2} - \frac{E_+^2}{E_o^2}G_1(-y',-\zeta')\right.$$

$$\left. - \frac{E_-^2}{E_o^2}\left[G_2(-y',-\zeta') + G_3(-y',-\zeta')\right]\right\} \quad . \qquad (5.16)$$

Equations 5.14-5.16 could have been derived from 5.11-5.13 by evaluating the limit as $a, A, B \rightarrow \infty$.

5.3.3 Discussion

The real and imaginary parts of the circular components of the polarization can now be substituted into the equations of the electromagnetic field. This is the final step in our calculations and by it we obtain equations describing the nonlinear characteristics of the medium in terms of the input fields. Let us repeat here the electromagnetic field equations.

$$\left[\frac{\partial\varphi_\pm}{\partial z} + \frac{1}{c}\frac{\partial E_\pm}{\partial t} + \frac{\nu}{c}(n_\pm - 1)\right] E_\pm = \frac{\nu}{2\epsilon_o c} P_{c\pm} \tag{5.17a}$$

$$\frac{\partial E_\pm}{\partial z} + \frac{1}{c}\frac{\partial E_\pm}{\partial t} = \frac{\nu}{2\epsilon_o c} P_{s\pm} \quad . \tag{5.17b}$$

Since we are considering only one frequency for each circular polarization, the ν subscripts have now been omitted from the fields and from the polarization components. These equations express the well known fact that the in phase component of the polarization influences the phase shift while the out of phase component governs the gain (or attenuation) of the medium. We have shown earlier that for the case discussed here no combination tones are produced and the circular components of the induced polarization oscillate only at the frequencies of the corresponding fields. (This can be seen even more readily from equations 5.3-5.5.) In view of this fact, for steady state operation we can physically set $\partial E/\partial t = 0$, $\partial\varphi/\partial t = 0$ and change the partial space derivatives to total derivatives. It is also evident from 5.17a that the in phase component of the polarization can be interpreted as contributing either to $(\nu/c)(n-1)$ or to $d\varphi/dz$. It is useful to talk about an index of refraction, however, only for the linear case. When saturation effects are included the phase is no longer a linear function of z and it is more convenient to consider the actual phase rather than some equivalent index of refraction.

After these simplifications, combining 5.11, 5.12a-d and 5.17a, we obtain for the Doppler broadened case

$$\frac{d\varphi_{\mp}}{dz} = \alpha \, \mathrm{Im} \left\{ w^{*}(x_o \pm y \mp \zeta + ia) - \frac{E_{\mp}^2}{2E^2} F_1 \pm y, \pm\zeta) - \frac{E_{\pm}^2}{E_o^2} \left[F_2(\pm y, \pm\zeta) + F_3(\pm y, \pm\zeta) \right] \right\} . \qquad (5.18)$$

The functions $F_{1,2,3}$ have been defined previously (5.12a-d), Im indicates the imaginary part. Similar expression is obtained, using equations 5.14-5.16, for homogeneous broadening

$$\frac{d\varphi_{\mp}}{dz} = \alpha_o \left\{ \frac{-(x_o' \pm y' \mp \zeta')}{1+(x_o' \pm y' \mp \zeta')^2} - \frac{E_{\mp}^2}{E_o^2} \, \mathrm{Im} \, G_1(\pm y', \pm\zeta') - \frac{E_{\pm}^2}{E_o^2} \left[G_2(\pm y', \pm\zeta') + G_3(\pm y', \pm\zeta') \right] \right. . \qquad (5.19)$$

The gain equations we cast into a slightly different form; from 5.17b,

$$dE_-/dz = \alpha_- E_- - \beta_- E_-^3 - \theta_{-+} E_- E_+^2 \tag{5.20a}$$

$$dE_+/dz = \alpha_+ E_+ - \beta_+ E_+^3 - \theta_{+-} E_+ E_-^2 \tag{5.20b}$$

where, for the Doppler broadened case, $\alpha_{\mp} = \alpha \text{Re } w^*(x_o \pm y \mp \zeta + ia)$, $\beta_{\mp} = (\alpha/2E_o^2) \text{ Re } F_1(\pm y, \pm\zeta)$, and $\theta_{\mp\pm} = (\alpha/2E_o^2) \text{ Re}\left[F_2(\pm y, \pm\zeta) + F_3(\pm y, \pm\zeta)\right]$; while for natural broadening $\alpha_{\mp} = \alpha_o\left[1+(x_o' \pm y' \mp \zeta')^2\right]^{-1}$ $\beta_{\mp} = (\alpha_o/E_o^2) \text{ Re } G_1(\pm y', \pm\zeta')$ and $\theta_{\mp\pm} = (\alpha_o/E_o^2) \text{ Re}\left[G_2(\pm y', \pm\zeta') + G_3(\pm y', \pm\zeta)\right]$.

It is instructive to examine the characteristics of the various terms in equations 5.18-5.20, or equivalently, the terms of the induced polarization as given by 5.11 and 5.12.

5.3.3A Linear Amplification

The first term in each of the equations for the induced polarization is the linear result, derived by the lowest order approximation, and valid for very weak fields. If $E^2/E_o^2 \ll 1$ and all nonlinear effects are negligible, for a Doppler broadened line we have

$$dE_\pm/dz = \alpha \text{Re } w^*(x_\pm + ia)\, E_\pm = \alpha_\pm E_\pm \tag{5.21a}$$

$$\left.\begin{matrix} d\varphi_{\pm}/dz \\ \text{or} \\ \frac{\nu}{c}(n_{\pm}-1) \end{matrix}\right\} = \alpha \, \text{Im} \, w^*(x_{\pm}+ia) \quad . \tag{5.21b}$$

And for a naturally broadened line

$$dE_{\pm}/dz = \alpha_o\left[1+(x_{\pm}')^2\right]^{-1} E_{\pm} = \alpha_{\pm}E_{\pm} \tag{5.22a}$$

$$\left.\begin{matrix} d\varphi_{\pm}/dz \\ \text{or} \\ \frac{\nu}{c}(n_{\pm}-1) \end{matrix}\right\} = \alpha_o \frac{-x_{\pm}'}{1+(x_{\pm}')^2} \tag{5.22b}$$

These results are similar to the corresponding linear solutions for Zeeman laser oscillators derived earlier by numerous investigators (18-22). The only difference is that for cavity fields the space Fourier components of the polarization (and time derivatives) are used and consequently the gain parameter α is proportional to $\bar{N}$ (the Fourier projection of the excitation density) rather than to N. Since in the linear theory each circularly polarized wave interacts only with the corresponding transition, the gain and the phase shift are also identical to those for scalar fields and nondegenerate levels, derived either by a density matrix approach (6) or by the Kramers-Kronig relations (12). We include the linear solutions here for the sake of completeness.

The solution of the differential gain equations is the well known exponential amplification formula

$$E_\pm(z) = E(0)\, e^{\alpha_\pm z} \text{ or } E_\pm^2(z) = E^2(0)\, e^{2\alpha_\pm z}, \tag{5.23}$$

while the phase shift increases linearly with z . The dependence of $\alpha_\pm$ and $n_\pm$-1 on the frequencies is shown on Figure 3 for various degrees of Doppler broadening (indicated by the value of the parameter $a = \gamma_{ab}/ku$), and also for the case of stationary atoms. The tabulated function w occurs in the theory of Doppler broadening and is better known in an approximate form, useful when $a \ll 1$. To the lowest order in a (6)

$$\alpha_\pm = \alpha \left[e^{-x_\pm^2} - \frac{2a}{\sqrt{\pi}} \left(1-2x_\pm F(x_\pm)\right) \right] \tag{5.24a}$$

and

$$n_\pm - 1 = \frac{c}{\nu}\, \alpha \left[\frac{2}{\sqrt{\pi}} F(x_\pm) - 2a x_\pm e^{-x_\pm^2} \right] \tag{5.24b}$$

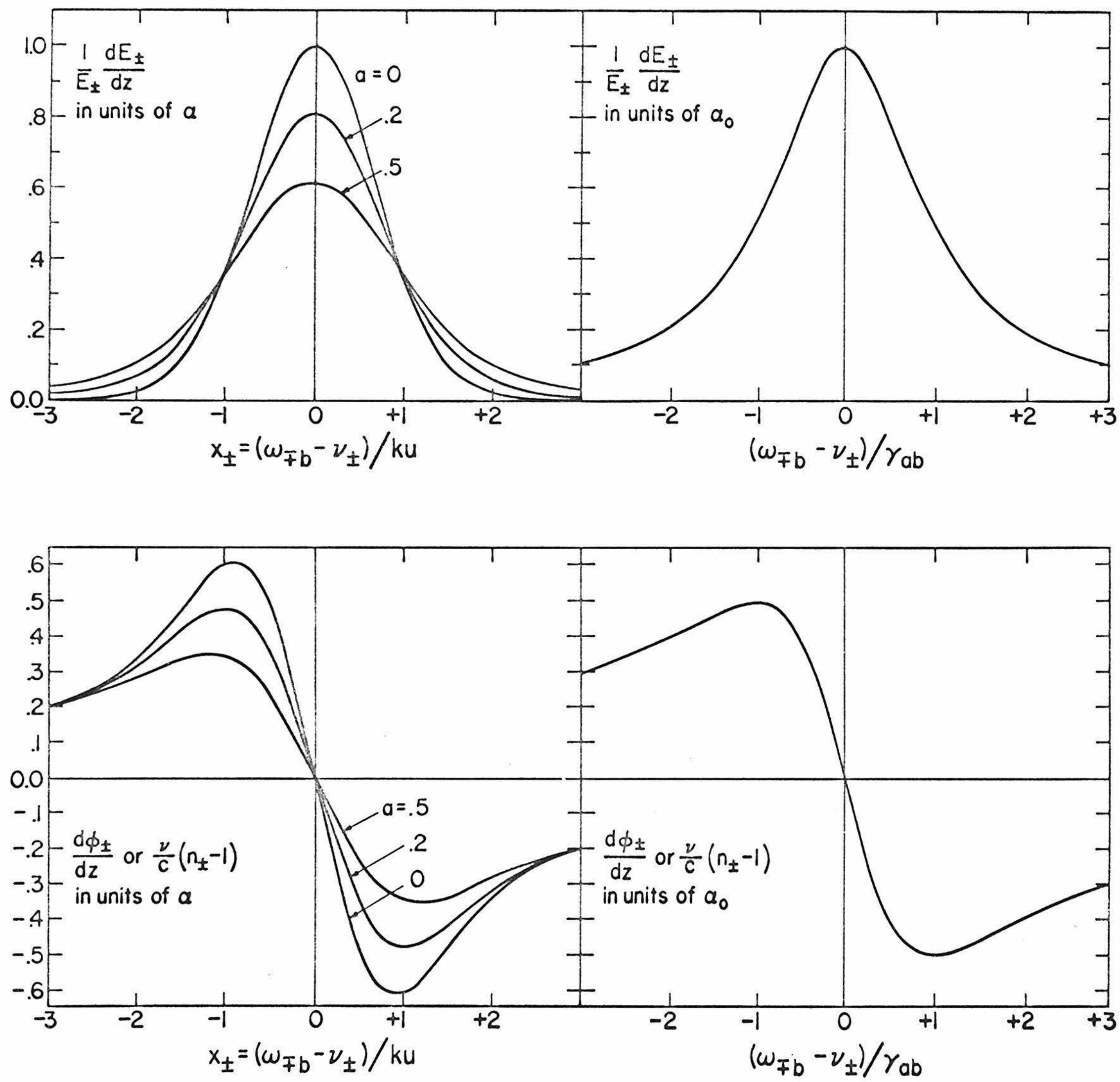
where

$$F(x) = e^{-x^2} \int_0^x e^{t^2}\, dt \,.$$

The equation for $\alpha_\pm$ is the familiar Gaussian with Lorentzian wings. If we let $a \to 0$ we have purely inhomogeneous broadening and

$$\alpha_\pm = \alpha e^{-x_\pm^2} = \alpha e^{-(\omega_{\mp b} - \nu_\pm)^2/(ku)^2} \tag{5.25a}$$

$$n_\pm - 1 = \frac{c}{\nu}\, \alpha \frac{2}{\sqrt{\pi}} F(x_\pm) \tag{5.25b}$$



(a) (b)

Figure 3.

Linear Gain and Phase Shift (or Index of Refraction)

(a) Doppler Broadened Transition (b) Naturally Broadened Transition

5.3.3B Lowest order nonlinear effects

Let us now examine the characteristics of the third order corrections to the polarization (i.e. to the gain and the phase shift). These are determined by the three functions F_1, F_2, F_3 (or G_1, G_2, G_3) each of which describes a different nonlinear effect. The real and imaginary parts of these quantities are shown on Figures 5 - 7 for various atomic parameters. In the following the properties of the three functions, as manifested by equations 5.12a-d (Doppler broadened case), 5.16a-c (stationary atoms), and by the plots of Figures 5 - 7 will be discussed in terms of the physical picture of the nonlinear interactions they represent. First the moving atom solutions ($F_{1,2,3}$) are examined in detail, after which we briefly discuss the corresponding stationary atom solutions ($G_{1,2,3}$), pointing out differences in behavior. To facilitate the discussion we show, on Figure 4 a schematic picture showing the Doppler gain curves of the two

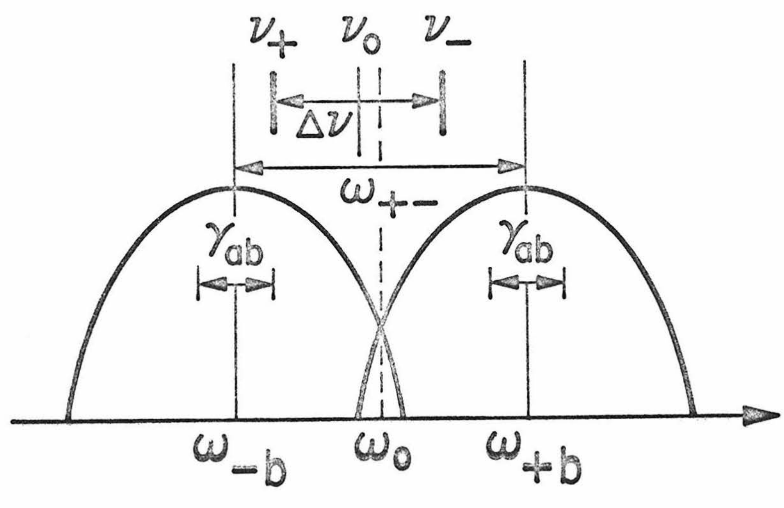


Figure 4.

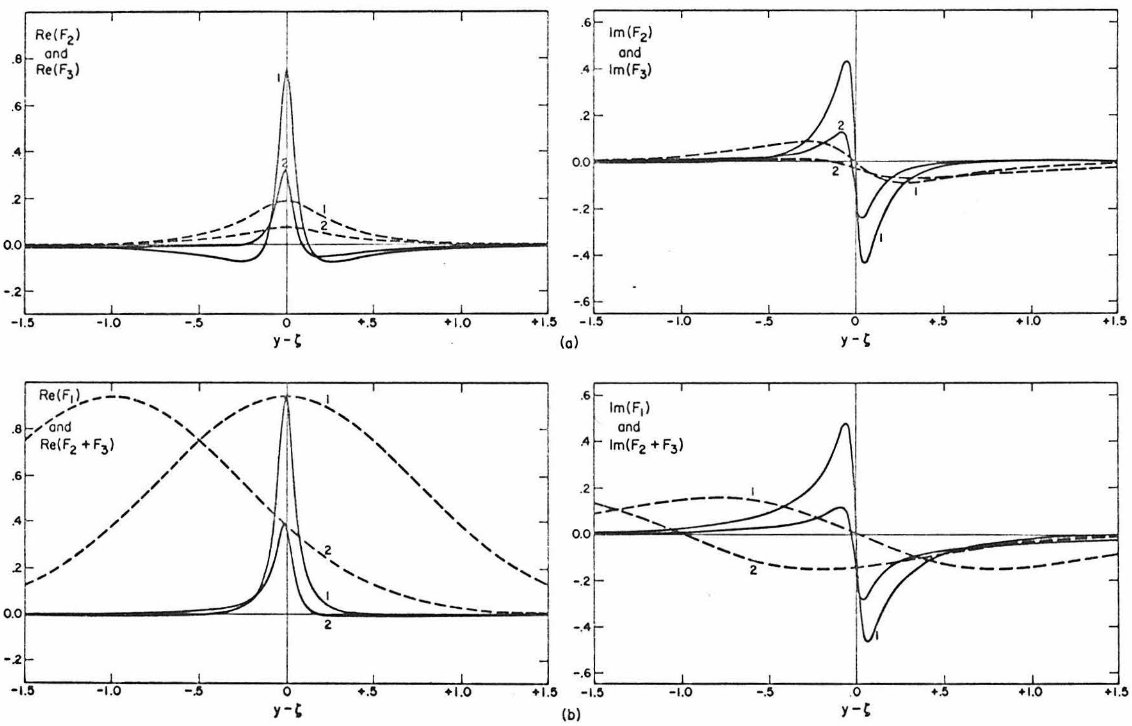


Figure 5.

The Nonlinear Polarization Functions: A = .06, B = .24, a = .3 ;

(a) F_2(dashed) and F_3 (solid) (b) F_1 (dashed) and $F_2 + F_3$ (solid) ;

1. $x_o = 0$, 2. $x_o = 1.0$.

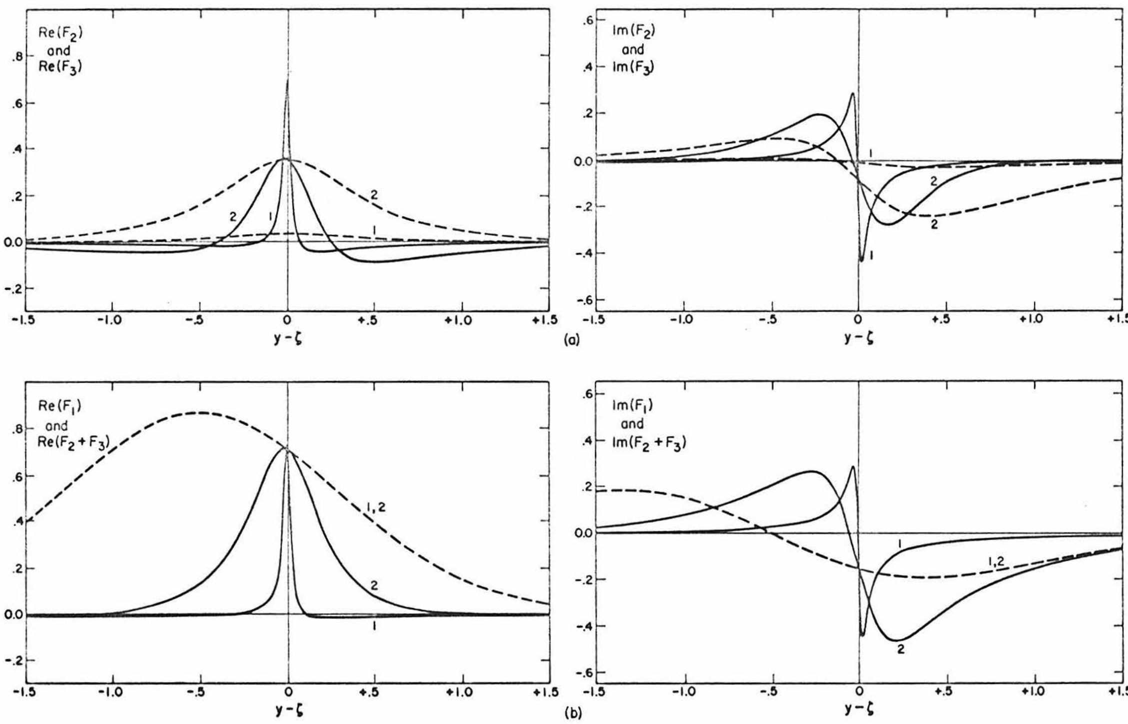


Figure 6.

The Nonlinear Polarization Functions: a = .5, x_o = .5 ;

(a) F_2 (dashed) and F_3(solid) (b) F_1(dashed) and $F_2 + F_3$ (solid) ;

1. A = .025, B = .475 2. A = .25, B = .25.

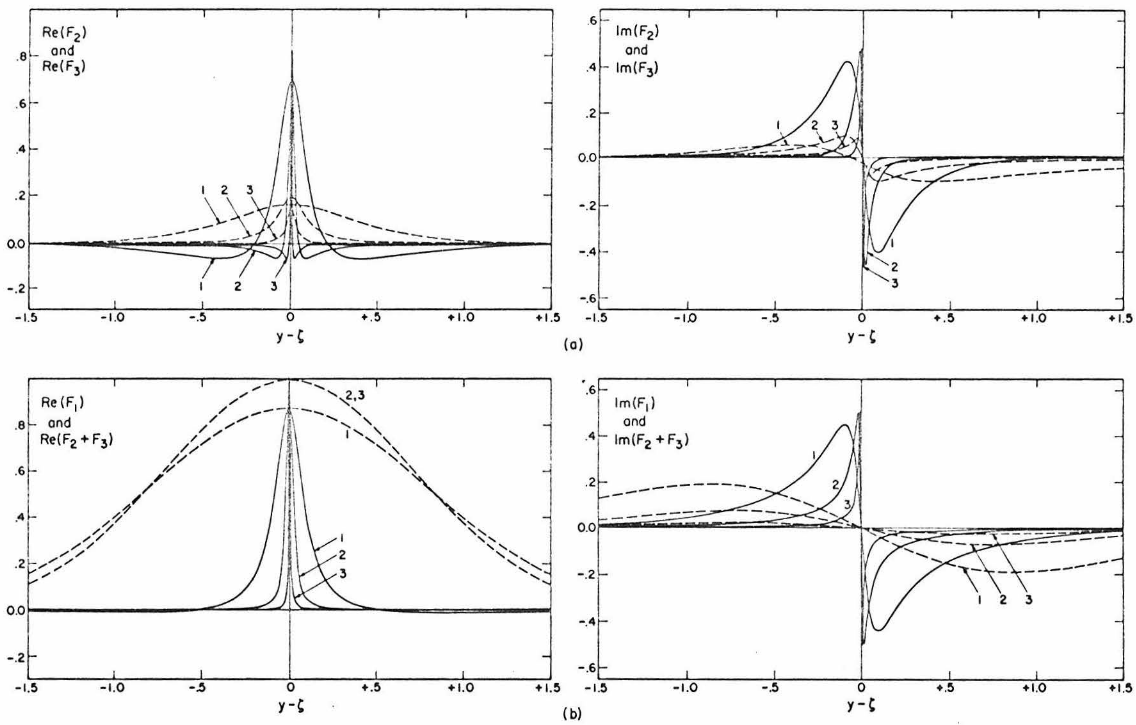


Figure 7.

The Nonlinear Polarization Functions: $x_o = 0$;

(a) F_2 (dashed) and F_3 (solid) (b) F_1 (dashed) and $F_2 + F_3$ (solid) ;

1. a = .5, A = .1, B = .4 2. a = .1, A = .02, B = .08

3. a = .03, A = .005, B = .025 .

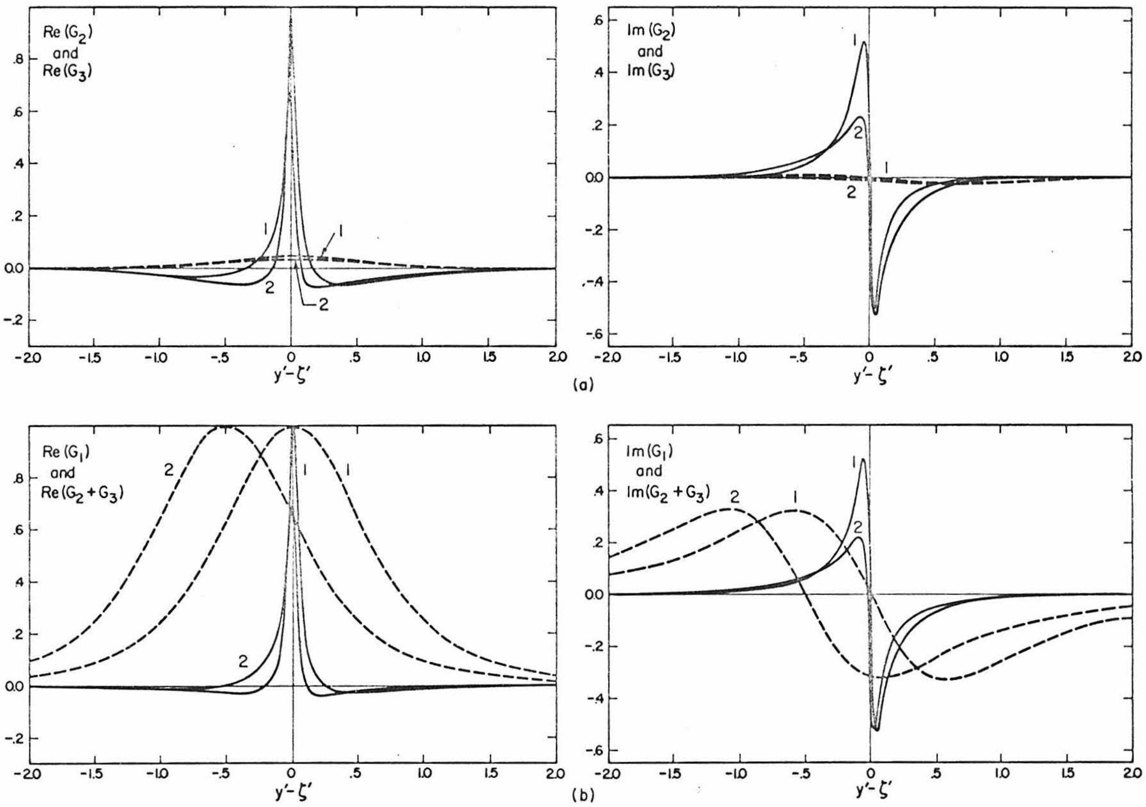


Figure 7A.

The Nonlinear Polarization Functions for Natural Broadening: $\gamma_a/2\gamma_{ab} = 0.05$, $\gamma_b/2\gamma_{ab} = 0.95$; (a) G_2 (dashed) and G_3 (solid) (b) G_1 (dashed) and $G_2 + G_3$ (solid); 1. $x_o' = 0$ 2. $x_o' = 0.5$.

circularly polarized transitions, the locations of the input frequencies and other relevant atomic parameters. For the Doppler broadened case it is found convenient to describe the frequency dependence of the nonlinear processes in terms of the two parameters $x_o = (\omega_o - \nu_o)ku$ and $y-\zeta = (\omega_{+-} - \Delta\nu)/ku$. The plots of $F_{1,2,3}$ are also made vs. these quantities.

The function F_1 describes the self saturation, or the nonlinear interaction of the field with itself. This effect is independent of the relative sizes of the decay rates γ_a , γ_b and its variation with either x_o or $y-\zeta$ has a width roughly equal to the Doppler width. In fact, just as the linear gain function, this quantity, for each circular polarization, depends only on $x_\pm = (\omega_{\mp b} - \nu_\pm)/ku$ the detuning of the field frequency from the center of its own transition, the peak of the real part of F_1 occurring at $x_\mp = x_o \pm y \mp \zeta = 0$ when the signal is tuned to the center of the corresponding transition. These properties are consistent with the physical description of self saturation, the signals depleting the upper and filling up the lower levels of their respective transitions, and thus burning a hole in their respective population inversions. In addition it is seen from Figure 7 that while the real part is quite insensitive to variations of a, in fact increasing somewhat with decreasing a, the imaginary part has the interesting feature that it decreases roughly proportionally to a and is negligible for a = .01. This property can be explained by the fact that for an inhomogeneously broadened line it is the atoms Doppler shifted away from resonance that contribute primarily to the phase shift, while those on or near resonance determine the gain. Thus the atoms responsible for the gain and therefore subject to saturation do not influence the phase shift. For a homogeneous line, however, where the same atoms cause the gain and the phase shift, the third order correction to each of these quantities is equally important.

$F_2 + F_3$ gives the total interaction between the two opposite circularly polarized waves. F_2 is a saturation term, due to the common lower level of the two transitions. The + field, for example, depletes the population inversion of the - field by filling up the common lower level. By its nature this coupling must be weaker than the self saturation since only the lower level is filled up, the upper level is not influenced. The effect of one field on the population inversion of the other is therefore smaller than that on its own. The strength of this interaction (both real and imaginary parts of F_2) depends on the relative sizes of γ_a, γ_b . For $\gamma_a \ll \gamma_b$ the common level decays much quicker than the other one and F_2 is negligibly small. Equation 5.12b also shows that this part of the nonlinear polarization has a resonance at $y-\zeta = 0$, when the signal frequency separation equals the Zeeman splitting, with a width equal to 2a. When we vary the detuning x_o on the other hand, with $y-\zeta$ kept fixed, the interaction has width 2 about $x_o = 0$. These properties can be explained physically in terms of holeburning. Because the transitions terminate on a common level, each signal burns a hole not only in its own Doppler gain profile, but in that of the other signal, too, at the corresponding point. The width of the holes being the natural line-width γ_{ab}, mutual gain saturation occurs only if the frequency separation of the signals is within γ_{ab} of the distance $\omega_{+-} = \omega_{+b} - \omega_{-b}$ between the two holes made by each signal. This situation is shown on Figure 8 . For the sake of clarity only the holes made by the right helicity (ν_+) signal are shown. The atoms involved are those travelling with velocity v_R, such that $\nu_-(1-v_R/c) = \omega_{+b}$. Evidently, only when $|\Delta_- - \Delta_+| < 2\gamma_{ab}$ is there significant saturation interaction between the left and right circular waves. The above condition can be rewritten as $|(\omega_{+b}-\nu_-)-(\omega_{-b}-\nu_+)| = |\omega_{+-} - \Delta\nu| < 2\gamma_{ab}$ which leads to the behavior discussed above. Variation of $\omega_o - \nu_o$ (i.e. x_o), on the other hand merely introduces a slowly varying Doppler weighting function. We have thus fully explained the behavior of F_2.

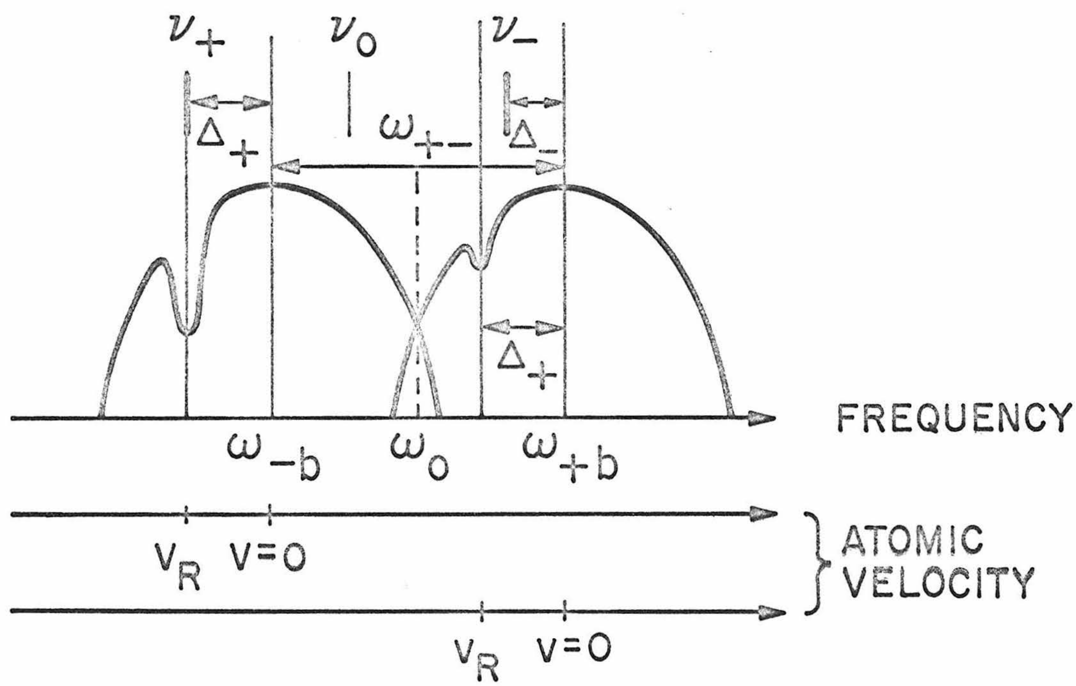


Figure 8.

The coupling described by F_3 is due to a Raman type simultaneous interaction of an atom with both fields. This results in a coherence of the magnetic sublevels, manifested by a nonzero ρ_{+-}. The relative magnitude of this interaction compliments that of the common level interaction F_2, i.e. F_3 is large when $\gamma_a \ll \gamma_b$ and vice versa. This double quantum process has the characteristics of the normal Raman effect, that is, it remains significant only if the frequency separation $\Delta\nu$ is resonant with the Zeeman separation ω_{+-}, the width depending only on γ_a, the width of the magnetic sublevels. (For our case, the common lower level plays the role of the virtual level and its decay rate does not influence the width of the resonance.) In addition, the real part of F_3 shows a peculiar behavior in the vicinity of $\omega_{+-}-\Delta\nu > \gamma_a$ $(y-\zeta > A)$, actually changing sign in this region. This is due to the fact that for its contribution to the induced polarization ρ_{+-} is multiplied by another atomic response function, resulting in the mixing of the real and imaginary parts of ρ_{+-}. It is obvious from the figures, however, that this behavior does not show up in the total interaction $F_2 + F_3$ of the two opposite circular waves because of a cancellation that happens between F_2 and F_3. As a result the variation of $F_2 + F_3$ with $y-\zeta$ is particularly simple, the real part having an essentially Lorentzian shape with a width 2A. Although this behavior is apparently present even for intermediate Doppler broadening it is easiest to show mathematically for a strongly Doppler broadened line. For this case the results take on an especially simple form. Combining F_2 and F_3,

$$F_2 + F_3 = \frac{A[(B+A)+i(y-\zeta)]}{A + i(y-\zeta)} \frac{1}{2[a+i(y-\zeta)]} [w^*(x_o+y-\zeta+ia) + w(x_o-y+\zeta+ia)]$$

$$+ \quad A \frac{i}{2(y-\zeta)} [w^*(x_o+y-\zeta+ia) - w^*(x_o-y-\zeta+ia)] + \frac{AB}{A+i(y-\zeta)}$$

$$[\frac{2}{\sqrt{\pi}} - 2\,(a + i(x_o + y - \zeta))\, w^*(x_o + y - \zeta + ia)\,] \quad . \tag{5.26}$$

For a, A, B $\ll$ 1, expanding to lowest order in these quantities and using our knowledge that $y-\zeta < a$ for any significant contribution, we obtain

$$F_2 + F_3 \simeq \frac{A}{A+i(y-\zeta)}\, e^{-x_o^2} = \frac{\gamma_a}{\gamma_a+i(\omega_{+-}-\Delta\nu)}\, e^{-(\omega_o-\nu_o)^2/(ku)^2} \tag{5.27}$$

which shows the simple behavior discussed above, with a Gaussian weighting envelope. Our reason for keeping F_2 and F_3 in general separate is that they describe two distinct physical processes. An interesting additional feature that can be observed from the plots is that for $x_o \neq 0$ the resonance in the interaction F_3 (or $F_2 + F_3$) occurs not exactly at $y-\zeta = 0$ but at a slightly different value. This indicates a weak frequency pulling effect, which is the peak of the coherent double quantum interaction of the atoms with the fields shifting somewhat from the resonant value $\Delta\nu = \omega_{+-}$.

The functions $G_{1,2,3}$, describing the nonlinear interactions for stationary atoms in general manifest characteristics similar to the Doppler broadened case. The differences are: 1) The cancellation between the common level saturation and the Raman interaction, discussed for moving atoms, is not perfect. As a result, the total interaction between the opposite circularly waves changes sign near $\omega_{+-}-\Delta\nu = \gamma_a$, i.e., the left circular signal actually gives a small positive contribution to the gain of the right circular one and vice versa. 2) The imaginary part of G_1 (self saturation) is substantial, unlike that of F_1 for strong Doppler broadening, since for stationary atoms the same atoms contribute to both the gain and the phase shift. These functions are plotted on Figure 7A vs. the parameters $x_o' = (\omega_o - \nu_o)/\gamma_{ab}$ and $y' - \zeta' = (\omega_{+-} - \Delta\nu)/2\gamma_{ab}$. The similarities in behavior are evident, with the natural width γ_{ab} of course replacing the Doppler width ku everywhere.

Except for the fact that space derivatives dE_m/dz take the place of the time derivatives $\dot{E}_m$, the incremental gain equations 5.20a,b are similar to those used to describe mode interactions in laser oscillators. When α_+ and α_- are equal or nearly equal, by the usual terminology of nonlinear differential equations, the coupling between the two fields is weak if $\beta_+ \beta_- > \theta_{+-} \theta_{-+}$ and strong if $\beta_+ \beta_- < \theta_{-+} \theta_{+-}$. Physically these inequalities mean respectively that the effect of one field on itself is larger or smaller than its effect on the other. In an oscillator, weak coupling results in the simultaneous oscillation of both modes while for strong coupling there exist two stable operating points with one or the other of the modes oscillating depending on the initial conditions. Both modes cannot coexist when there is strong coupling since this results in an unstable situation where if one of the fields becomes slightly weaker its effective gain becomes negative and it is quenched by the other, stronger field.

For our laser amplifier it is evident that since no losses were considered, $dE_{\pm}/dz$ cannot become zero or negative except for fields so strong that equations 5.20a,b are no longer valid. If, on the other hand, we introduce loss terms into the linear gains, i.e. let $\alpha_{\mp} = \alpha \mathrm{Re}\, w(x_o \pm y \mp \zeta + ia) - \kappa$ (or $\alpha_{\mp} = \alpha_o [1 + (\omega_{\pm b} - \nu_{\mp})^2/\gamma_{ab}^2]^{-1} - \kappa$, for homogeneous broadening), we have equations exactly like those for an oscillator. In any case, however, $\beta_+\beta_- > \theta_{+-}\theta_{-+}$, the coupling is weak and both fields are amplified without quenching occurring. Critical coupling, $\beta_+\beta_- = \theta_{+-}\theta_{-+}$, exists for $y - \zeta = 0$ (Zeeman separation of the transitions equal to the frequency difference of the input signals) since at this point $F_1 = F_2 + F_3$ and $G_1 = G_2 + G_3$ as seen from 5.12a-d and 5.15a-c and is also indicated on Figures 5 - 7 . To the extent that it is possible to compare the results for cavity fields and for travelling waves, these results agree with those of Fork and Sargent (18) and of Culshaw and Kannelaud (20) obtained for a J=1 → J=0 laser oscillator with an axial magnetic field, and strong Doppler broadening. For a laser oscillator with completely degenerate magnetic sublevels (zero magnetic field) Doyle and White (25) have shown that coherence of the magnetic sublevels can result in strong coupling for various polarizations of the fields, depending on the J values of the levels. For a $J = 1 \rightarrow J = 0$ transition and opposite circularly polarized modes, however, their perturbational analysis predicts weak coupling in agreement with our results for an amplifier of the same type.

So far in this chapter the input field frequencies and the relative magnitudes of the amplitudes were completely arbitrary, the only restriction being that the intensities remain small compared to the saturation parameter E_o^2. In the following section we will examine a special case separately, namely when the two opposite circular waves have the same frequency and amplitude. This is identical to the case of a single, linearly polarized input field.

5.3.4 Single, Linearly Polarized Input Field

When the two frequencies ν_+ and ν_- and the two field amplitudes E_+ and E_- are equal, we have the case of a single linearly polarized input signal. It is, however, meaningful to treat this case separately only if the gains for the opposite circularly polarized components are equal, so that the wave remains linearly polarized. This happens if $\nu_- = \nu_+ = \nu = (\omega_{+b} - \omega_{-b})/2$, i.e., if the input signal is tuned to the zero magnetic field line center. Then, since $E_+ = E_- = E/\sqrt{2}$,

$$\frac{dE}{dz} = \sqrt{2}\,\frac{\nu}{2\epsilon_o c}\,P_{s+} = \sqrt{2}\,\frac{\nu}{2\epsilon_o c}\,P_{s-} = \alpha E \,\mathrm{Re}\left\{w^*(y+ia) - \frac{E^2}{4E_o^2}[F_1(y,0) + F_2(y,0) + F_3(y,0)]\right\}, \tag{5.28a}$$

for the Doppler broadened case and

$$\frac{dE}{dz} = \alpha_o E \left\{ \frac{1}{1+y'^2} - \frac{E^2}{2E_o^2} \operatorname{Re}\left[G_1(y',0) + G_2(y',0) + G_3(y',0) \right] \right\} , \qquad (5.28b)$$

for the naturally broadened case.

The other quantity we wish to calculate for this case is the Faraday rotation. With $\varphi_- = -\pi/2 + \Phi + \varphi$ and $\varphi_+ = -\pi/2 - \Phi + \varphi$

$$\bar{E} = \operatorname{Re} \left\{ E\left[\hat{e}_x \sin \Phi + \hat{e}_y \cos \Phi \right] e^{i(kz-\nu t+\varphi)} \right\} \quad . \qquad (5.29)$$

The Faraday rotation angle is thus given by $\Phi = (\varphi_- - \varphi_+)/2$, and its differential equation is

$$\frac{1}{2}\left[\frac{d\varphi_-}{dz} - \frac{d\varphi_+}{dz} \right] \frac{E}{\sqrt{2}} = \frac{1}{2} \frac{\nu}{2\epsilon_o c} \left[P_{c-} - P_{c+} \right] \quad . \qquad (5.30)$$

Since for the special case considered here $P_{c+} = -P_{c-}$, the incremental Faraday rotation angle Φ is given by

$$\frac{d\Phi}{dz} = \alpha \operatorname{Im} \left\{ w^*(y+ia) - \frac{E^2}{4E_o^2} \left[F_1(y,0) + F_2(y,0) + F_3(y,0) \right] \right\} , \text{ and} \qquad (5.31a)$$

$$\frac{d\Phi}{dz} = \alpha_o \left\{ \frac{-y'}{1+y'^2} - \frac{E^2}{2E_o^2} \operatorname{Im}\left[G_1(y',0) + G_2(y',0) + G_3(y',0) \right] \right\} \cdot \qquad (5.31b)$$

We note that the incremental Faraday rotation is a constant only for weak fields. The effect of saturation is to make the rotation a function of the electromagnetic field intensity which in turn is a function of z . Thus we can no longer speak of a Faraday rotation factor or Verdet constant, but must evaluate Φ by integrating 5.31b and combining it with equation 5.28b.

The characteristics of the individual saturation functions $F_{1,2,3}$ and $G_{1,2,3}$ were discussed in section 5.3.3. The nonlinear characteristics of our laser amplifier for the single linearly polarized signal are described by $F_1 + F_2 + F_3$, and $G_1 + G_2 + G_3$, the real and imaginary parts of which are shown on Figure 9 for various atomic parameters. Interesting behavior is indicated for a transition where the decay rate γ_a of the J = 1 level is very small compared to the natural linewidth (γ_{ab}) or to the Doppler width (ku). While for magnetic field splitting larger than γ_a the left and right circular components of the signal interact with different atoms, for completely degenerate levels a coherence of the magnetic substates results from double quantum interactions of a single atom. This effect, already discussed in section 5.3.3, modifies the nonlinear characteristics of the medium. While the linear gain remains essentially unaltered, the nonlinear gain correction changes drastically as the sublevels become completely degenerate due to the magnetic field approaching zero. The sharp peak in the real part of the third order polarization at y or y' = 0 indicates the presence of a dip of the same width in the output of the amplifier. The existence of this dip for a strongly Doppler broadened transition has already been shown by Heer and Graft (22) and the analogous effect in a laser oscillator has been observed by Culshaw and Kannelaud (14). The Faraday rotation also shows an anomalous

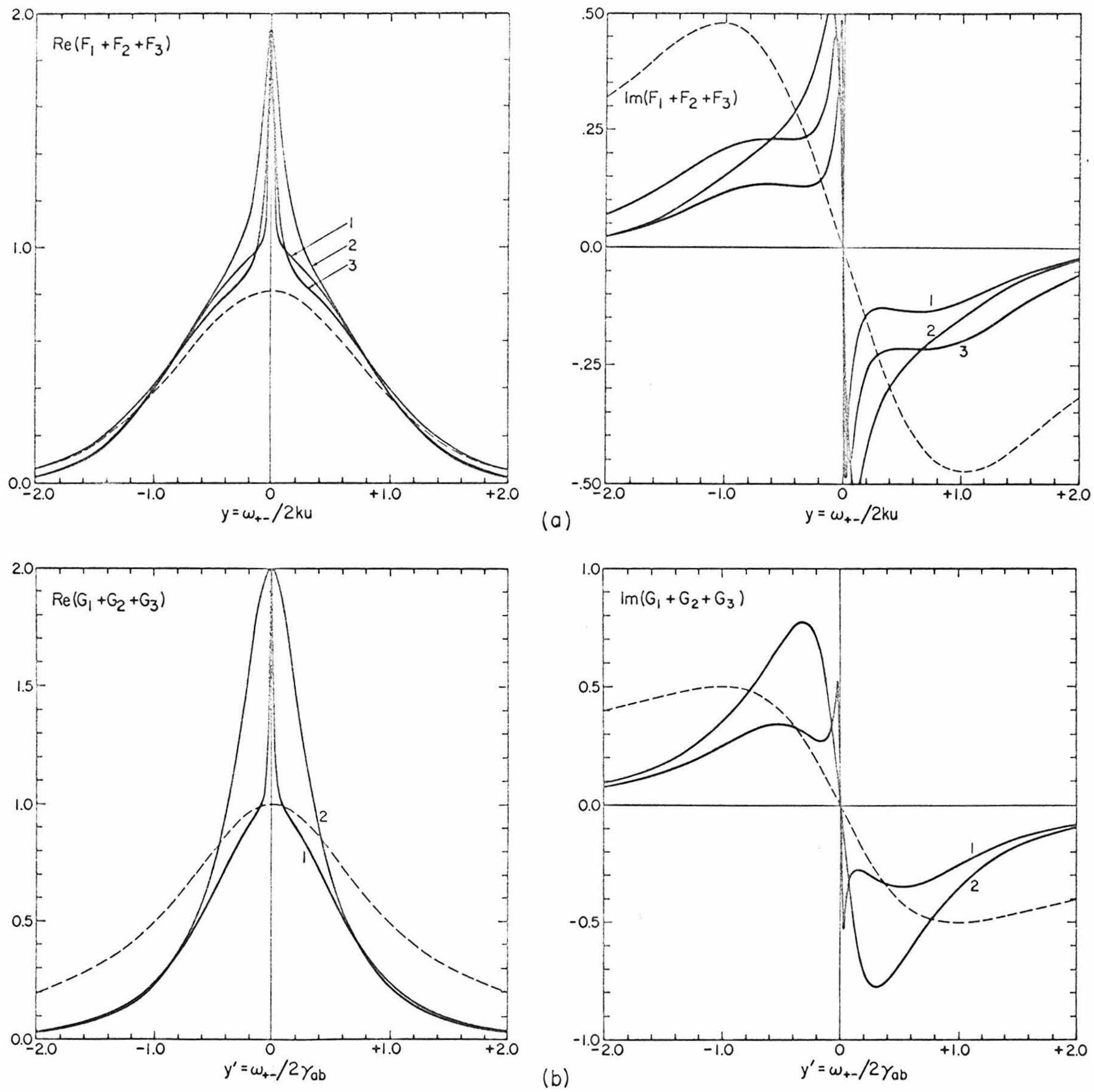


Figure 9.

The Total Nonlinear Polarization for a Single Linearly Polarized Wave: (a) Doppler Broadened Line 1. a = .2, A = .02, B = .18 2. a = .2, A = .1, B = .1 3. a = .5, A = .05, B = .45 (b) Naturally Broadened Line 1. $\gamma_a/2\gamma_{ab}$ = .02 2. $\gamma_a/2\gamma_{ab}$ = .3 Dashed Curves: Linear Polarization behavior near zero splitting as we can see by examining the imaginary part of $F_1 + F_2 + F_3$ or $G_1 + G_2 + G_3$. Due to the fact that the coherent double quantum interaction has a width γ_a about $\omega_{+-} = 0$ the imaginary part of the saturation function, has sharp peaks around $\omega_{+-} = \pm\gamma_a$. The imaginary part of the linear polarization function at this point is still small since it reaches maxima only at $\omega_{+-} = \pm ku$ (or $\omega_{+-} = \pm\gamma_{ab}$). For a = 0.2 and A = 0.02, for example, at y = 0.02 $Im(F_1 + F_2 + F_3) = -0.5$, while Im w* = -.016. Thus the incremental Faraday rotation changes sign in the region $0 < \omega_{+-} < \gamma_a$ for field intensities as low as $0.15\ E_o^2$.

Although, as indicated previously, equations 5.28 and 5.31 have only limited applicability, (only for as long as 5.28 gives values of E^2 smaller than approximately $0.5\ E_o^2$) we shall nevertheless integrate these equations now to obtain the field intensity and the rotation as a function of z. Our purpose in doing so is to show the behavior at or in the neighborhood of y = 0 where computational difficulties will be encountered in the derivation of solutions valid for arbitrarily strong fields.

Equation 5.28a can be written as

$$\frac{dE^2}{dz} = 2\alpha E^2 \left[\text{Re } w^*(y+ia) - \frac{E^2}{E_o^2} \text{Re } F(y) \right] , \tag{5.32}$$

where $F(y) = \frac{1}{4}\left[F_1(y,0) + F_2(y,0) + F_3(y,0)\right]$.

This is of the form $df/dz + c_1 f = c_2 f^2$ which is known as Bernoulli's equation and can be integrated if we let g = 1/f. The result is

$$\mathcal{I} = \frac{\mathcal{I}_o \exp[2\alpha \mathrm{Re}\ w^*(y+ia)z]}{1 + (\mathrm{Re}\ F(y)/\mathrm{Re}\ w^*(y+ia))\mathcal{I}_o \exp[2\alpha \mathrm{Re}\ w^*(y+ia)z]} \tag{5.33}$$

where $\mathcal{I} = E^2/E_o^2$, and $\mathcal{I}_o = \mathcal{I}(z=0)$.

The numerator exhibits the linear gain, while the denominator shows the saturation. From equation 5.30 we get for the Faraday rotation

$$\Phi = -2\alpha\{\mathrm{Im}\ w^*(y+ia)z - \mathrm{Im}\ F(y) \int_0^z \mathcal{I}\ dz\} \ . \tag{5.34}$$

Combining with 5.32, after performing the integration and some algebraic manipulation we obtain

$$\Phi = \alpha \{\mathrm{Im}\ w^*(y+ia)z - \mathcal{I}_o \frac{\mathrm{Im}\ F(y)}{\mathrm{Re}\ F(y)} [\mathrm{Re}\ w^*(y+ia)z$$
$$- \frac{1}{2\alpha} \ln \frac{[1+\mathrm{Re}\ F(y)/\mathrm{Re}\ w^*(y+ia)]\ \mathcal{I}_o \exp[2\alpha \mathrm{Re}\ w^*(y+ia)z]}{1+(\mathrm{Re}\ F(y)/\mathrm{Re}\ w^*(y+ia))\ \mathcal{I}_o \exp[2\alpha \mathrm{Re}\ w^*(y+ia)z]}]\} \ . \tag{5.35}$$

Similar equations can be obtained for the naturally broadened case, with $\alpha \rightarrow \alpha_o$, $\mathrm{Re}\ w^*(y+ia) \rightarrow \frac{1}{1+y'^2}$ and $F(y) \rightarrow \frac{1}{2}(G_1 + G_2 + G_3)$. Figure 10 shows the output intensity $\mathcal{I}_{out} = E_{out}^2/E_o^2$ and the total Faraday rotation Φ vs. magnetic field splitting for an input intensity $\mathcal{I}_{in} = E_{in}^2/E_o^2 = 0.15$. The two values of αz used, 0.7 and 0.95, correspond to linear power gains of 4 and 6 respectively. All the characteristics discussed above are evident. The depth of the central dip and the maximum value of the anomalous rotation is seen to increase as the ratio of γ_a/γ_{ab} becomes smaller. This is consistent with the behavior of the function F_3. For higher gains, i.e., stronger field intensities, we expect the dip to become deeper, although not as much as an application of the perturbational results would indicate. We shall, in

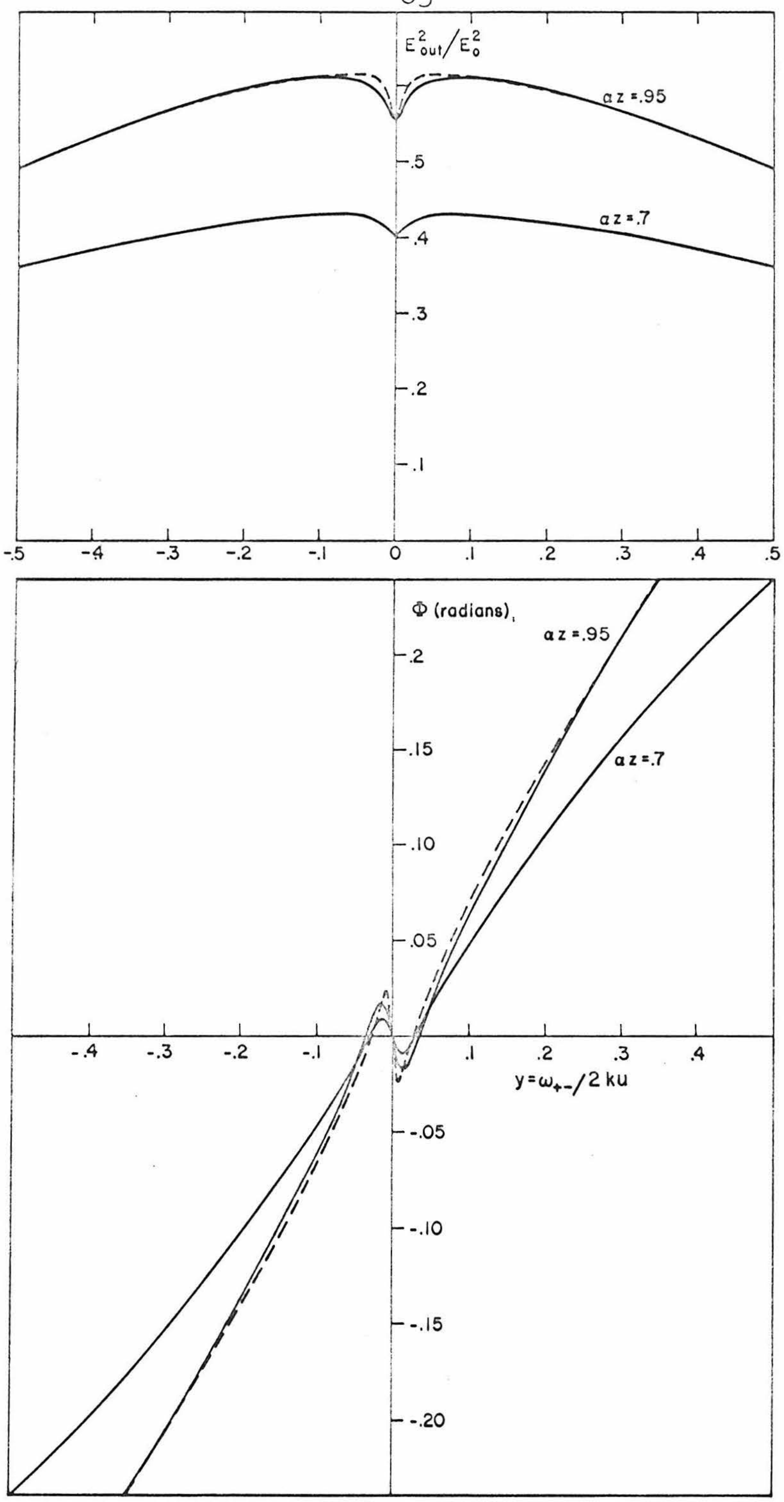


Figure 10.

Output Intensity and Faraday Rotation for a Linearly Polarized Wave.

E^2_{in}/E^2_o= .15, a = .2; Solid: A = .02, B = .18, Dashed: A = .01, B = .19

section 5.4.4 return to the special case of a single linearly polarized field and derive the depth of the dip for arbitrarily large field intensities.

5.4 The Strong Field Solutions

5.4.1 Introduction

We have seen in the previous sections for how limited a range of field intensity are the iterative solutions useful. Nonlinear effects are negligible for $E_{\pm}^2/E_o^2 \ll 1$ and the whole approach breaks down when the fields become comparable to the saturation field. For $E_{\pm}^2 > .5\, E_o^2$ the most we can expect is to draw some qualitative conclusions from the results of section 5.3.(Even these might be misleading: the gain, for example, never goes to zero as it would appear from the iterative results.) For quantitative results we must attempt to obtain solutions valid for strong fields. For the case discussed here, i.e., two opposite circularly polarized input signals, it is clear from equations 5.3 and 5.4 that $N_{\pm b}(z,t,v)$ is independent of t and $\rho_{+-}(z,t,v)$ has a known time dependence $(e^{i\Delta})$. Thus these quantities can be removed from the integrals resulting in a set of simultaneous linear equations. In principle these can be solved, $P_{\pm}(z,v)$ calculated and the integration over v performed to obtain $P_{\pm}(z)$ valid for fields of arbitrary strength. Unfortunately $P_{\pm}(z,v)$ in practice is such a complicated function of v, the fields, and atomic parameters that the integration is impractical since it is necessary that we have the polarization as a closed form function of the field intensities.

Fortunately, however, it is possible to obtain exact solutions for certain special cases and useful approximate solutions for still other specific cases. In this chapter we shall calculate and discuss these strong field solutions. Where necessary we shall use our knowledge of the characteristics of the iterative results to fill in some of the gaps in generality and to infer what the results would be like without the approximations, made out of necessity.

5.4.2 The Density Matrix for Strong Fields

From equations 5.3-5.5 we get, after performing the integrations over t_1, t_2, t_3 and setting $k_+ = k_- = k$ in the Doppler broadening terms:

$$\rho_{+-}(v) = -\frac{\gamma_a\gamma_b}{4}\frac{1}{\gamma_a+i(\omega_{+-}-\Delta\nu)}\left\{\frac{E_-E_+}{E_o^2}\left[\frac{N_{+b}}{\gamma_{ab}+i(\omega_{+b}-\nu_-+kv)}+\frac{N_{-b}}{\gamma_{ab}-i(\omega_{-b}-\nu_++kv)}\right]\right.$$

$$\left.\times e^{i\Delta}+\left[\frac{E_+^2/E_o^2}{\gamma_{ab}+i(\omega_{+b}-\nu_-+kv)}+\frac{E_-^2/E_o^2}{\gamma_{ab}-i(\omega_{-b}-\nu_++kv)}\right]\rho_{+-}\right\} \quad . \qquad (5.36)$$

Or letting $\rho_{+-} = \rho'_{+-}\, e^{-i\Delta}$

$$\rho'_{+-}(v) = -\frac{\gamma_a\gamma_b}{4}\frac{1}{\gamma_a+i(\omega_{+-}-\Delta\nu)}\left\{\frac{E_-E_+}{E_o^2}\left[\frac{N_{+b}}{\gamma_{ab}+i(\omega_{+b}-\nu_-+kv)}+\frac{N_{-b}}{\gamma_{ab}-i(\omega_{-b}-\nu_++kv)}\right]\right.$$

$$\left.+\left[\frac{E_+^2/E_o^2}{\gamma_{ab}+i(\omega_{+b}-\nu_-+kv)}+\frac{E_-^2/E_o^2}{\gamma_{ab}-i(\omega_{-b}-\nu_++kv)}\right]\rho'_{+-}\right\} \quad . \qquad (5.37)$$

Similarly,

$$N_{+b}(v) = N_o - \frac{E_-^2}{E_o^2}\frac{\gamma_{ab}^2}{\gamma_{ab}^2+i(\omega_{+b}-\nu_-+kv)^2}N_{+b} - \frac{E_+^2}{E_o^2}\frac{\gamma_{ab}\gamma_a/2}{\gamma_{ab}^2+(\omega_{-b}-\nu_++kv)^2}N_{-b} - \frac{E_-E_+}{E_o^2}$$

$$\times\left\{\frac{\gamma_{ab}}{2}\left[\frac{\rho'_{+-}}{\gamma_{ab}+i(\omega_{+b}-\nu_-+kv)}+\frac{\rho'^{*}_{+-}}{\gamma_{ab}-i(\omega_{+b}-\nu_-+kv)}\right]+\frac{\gamma_a}{4}\left[\frac{\rho'_{+-}}{\gamma_{ab}-i(\omega_{-b}-\nu_++kv)}\right.\right.$$

$$\left.\left.+\frac{\rho'^{*}_{+-}}{\gamma_{ab}+i(\omega_{-b}-\nu_++kv)}\right]\right\} \quad . \qquad (5.38)$$

$$N_{-b}(v) = N_o - \frac{E_+^2}{E_o^2}\frac{\gamma_{ab}^2}{\gamma_{ab}^2+(\omega_{-b}-\nu_++kv)^2} N_{-b} - \frac{E_-^2}{E_o^2}\frac{\gamma_{ab}\gamma_a/2}{\gamma_{ab}^2+(\omega_{+b}-\nu_-+kv)^2} N_{+b} - \frac{E_-E_+}{E_o^2}$$

$$\times\left\{\frac{\gamma_{ab}}{2}\left[\frac{\rho_{+-}'^*}{\gamma_{ab}+i(\omega_{-b}-\nu_++kv)} + \frac{\rho_{+-}'}{\gamma_{ab}-i(\omega_{-b}-\nu_++kv)}\right] + \frac{\gamma_a}{4}\left[\frac{\rho_{+-}'^*}{\gamma_{ab}-i(\omega_{+b}-\nu_-+kv)}\right.\right.$$

$$\left.\left.+\frac{\rho_{+-}'}{\gamma_{ab}+i(\omega_{+b}-\nu_-+kv)}\right]\right\} \quad . \qquad (5.39)$$

And finally ,

$$\frac{\nu}{2\epsilon_o c} P_-(v) = -i\frac{\alpha}{N_o}\left\{E_- \frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_-+kv)} + E_+ \frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_-+kv)}\rho_{+-}'\right\} N_{+b} \qquad (5.40a)$$

$$\frac{\nu}{2\epsilon_o c} P_+(v) = -i\frac{\alpha}{N_o}\left\{E_+ \frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{-b}-\nu_++kv)} + E_- \frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{-b}-\nu_++kv)}\rho_{+-}'^*\right\} N_{-b} \ . \qquad (5.40b)$$

The linear equations 5.36-5.39 are to be solved for $N_{\pm b}$, ρ_{+-}' and the polarization calculated from 5.40 and 5.41.

5.4.3 Two Special Cases

While the solutions of equations 5.36-5.41 in general are too complex to allow calculation of the behavior of the induced polarization for strong fields, useful information can be obtained by examining special cases. Let us now assume that the following conditions hold: $\omega_{-b}-\nu_+ = \omega_{+b}-\nu_- = 0$, $W(v) = \delta(v)$. Then, since $\omega_{+-} - \Delta\nu = 0$, we obtain

$$\rho'_{+-} = - \frac{(\gamma_b/2\gamma_{ab})\ (E_+E_-/E_o^2)}{1 + (\gamma_b/2\gamma_{ab})\ (E_+^2 + E_-^2)/2E_o^2} \frac{N_{+b} + N_{-b}}{2} , \tag{5.41}$$

and after substituting this into 5.38, 5.39

$$N_{+b} = N_o - \frac{E_-^2}{E_o^2} \frac{1 + (\gamma_b/2\gamma_{ab})\ (E_-^2/2E_o^2 - (\gamma_a/2\gamma_{ab})\ (E_+^2/2E_o^2))}{1 + (\gamma_b/2\gamma_{ab})\ (E_+^2 + E_-^2)/2E_o^2} N_{+b}$$

$$+ \frac{E_+^2}{E_o^2} \frac{(\gamma_b/2\gamma_{ab})\ (E_-^2/2E_o^2) - (\gamma_a/2\gamma_{ab})\ (1 + (\gamma_b/2\gamma_{ab})\ (E_+^2/2E_o^2))}{1 + (\gamma_b/2\gamma_{ab})\ (E_+^2 + E_-^2)/2E_o^2} N_{-b} \tag{5.42a}$$

$$N_{-b} = N_o - \frac{E_+^2}{E_o^2} \frac{1 + (\gamma_b/2\gamma_{ab})\ (E_+^2/2E_o^2 - (\gamma_a/2\gamma_{ab})\ (E_-^2/2E_o^2))}{1 + (\gamma_b/2\gamma_{ab})\ (E_+^2 + E_-^2)/2E_o^2} N_{-b}$$

$$+ \frac{E_-^2}{E_o^2} \frac{(\gamma_b/2\gamma_{ab})\ (E_+^2/2E_o^2) - (\gamma_a/2\gamma_{ab})\ (1+(\gamma_b/2\gamma_{ab})\ (E_-^2/2E_o^2))}{1 + (\gamma_b/2\gamma_{ab})\ (E_+^2 + E_-^2)/2E_o^2} N_{+b} . \tag{5.42b}$$

After some tedious algebra these equations can be shown to have the simple solutions

$$N_{+b} = \frac{1 + (\gamma_b/2\gamma_{ab})\ (E_+^2/E_o^2)}{1 + E_+^2/E_o^2 + E_-^2/E_o^2} N_o , \tag{5.43a}$$

$$N_{-b} = \frac{1 + (\gamma_b/2\gamma_{ab})\ (E_-^2/E_o^2)}{1 + E_+^2/E_o^2 + E_-^2/E_o^2} N_o , \tag{5.43b}$$

and by a simple substitution

$$\rho'_{+-} = - \frac{(\gamma_b/2\gamma_{ab})\,(E_+E_-/E_o^2)}{1 + E_+^2/E_o^2 + E_-^2/E_o^2}\, N_o = \rho'^{*}_{+-} \quad . \tag{5.44}$$

Combining 5.43-5.44 and 5.40a,b and making use of 2.10, the incremental gains are

$$\frac{dE_-}{dz} = \alpha_o E_- \left\{ \frac{1 + (\gamma_b/2\gamma_{ab})\,(E_+^2/E_o^2)}{1 + E_+^2/E_o^2 + E_-^2/E_o^2} - \frac{(\gamma_b/2\gamma_{ab})\,(E_+^2/E_o^2)}{1+E_+^2/E_o^2 + E_-^2/E_o^2} \right\}$$

$$= \alpha_o E_- \frac{1}{1 + E_+^2/E_o^2 + E_-^2/E_o^2} \tag{5.45a}$$

$$\frac{dE_+}{dz} = \alpha_o E_+ \frac{1}{1 + E_+^2/E_o^2 + E_-^2/E_o^2} \quad . \tag{5.45b}$$

These equations are to be compared with 5.20a,b for weak fields. From the form of the denominator, $1 + E_+^2/E_o^2 + E_-^2/E_o^2$, we see that the coupling between the left and right circularly polarized remains as it was for weak fields, i.e. critical at the special point $\omega_{+-} - \Delta\nu = 0$. Expanding 5.45a to first order in the intensities we get

$$\frac{dE_-}{dz} = \alpha_o E_- \left\{ 1 - E_+^2/E_o^2 - E_-^2/E_o^2 \right\} . \tag{5.46}$$

which agrees with the weak field results. More instructive is to expand 5.45a in its first form, before simplification. Then

$$\frac{dE_-}{dz} = \alpha_o E_- \left\{ 1 - E_-^2/E_o^2 - (\gamma_a/2\gamma_{ab})(E_+^2/E_o^2) - (\gamma_b/2\gamma_{ab})(E_-^2/E_o^2) \right\} \quad . \tag{5.47}$$

The first three terms come from the first term of 5.45a and include the linear gain, the self saturation and the so-called common level saturation (as discussed in sec. 5.3.3); the last term comes from the second term of 5.45a and is the contribution of ρ_{+-} , or the Raman term. The simplicity of the solution suggests that a transformation of variables is in order. This will now be done to obtain the gain for the Doppler broadened case (still for central tuning and $\omega_{+-} = \Delta\nu$). Since $kv \neq 0$ the gain dE_-/dz is given by

$$\frac{1}{E_-}\frac{dE_-}{dz} = \frac{1}{E_-} \mathrm{Re}\, \frac{\nu}{2\epsilon_o c} P_- = \frac{\alpha_o}{N_o}\frac{1}{\sqrt{\pi}u} \int_{-\infty}^{+\infty} e^{-v^2/u^2}$$

$$\left\{ \frac{\gamma_{ab}^2}{\gamma_{ab}^2+(kv)^2}\left[N_{+b} + \frac{E_+}{E_-} \mathrm{Re}\, \rho'_{+-}\right] - \frac{E_+}{E_-}\frac{\gamma_{ab} kv}{\gamma_{ab}^2+(kv)^2} \mathrm{Im}\, \rho'_{+-} \right\} dv ,$$

we try the new variables

$$x = N_{+b} + \frac{E_+}{E_-} \mathrm{Re}\, \rho'_{+-} + \frac{E_+}{E_-}\frac{kv}{\gamma_{ab}} \mathrm{Im}\, \rho'_{+-} \tag{5.49a}$$

$$y = N_{-b} + \frac{E_-}{E_+} \mathrm{Re}\, \rho_{+-} - \frac{E_-}{E_+}\frac{kv}{\gamma_{ab}} \mathrm{Im}\, \rho_{+-} \quad . \tag{5.49b}$$

Substituting the values from equations 5.37-5.39 into the right hand side of 5.49 we get for $\gamma_a \ll \gamma_b$, after grouping terms, the new equations

$$x = N_o - \left[\frac{E_-^2}{E_o^2} \frac{\gamma_{ab}^{\;2}}{\gamma_{ab}^{\;2}+(kv)^2} + \frac{E_+^2}{2E_o^2} \frac{\gamma_{ab}^{\;2}-(kv)^2}{\gamma_{ab}^2+(kv)^2}\right]x - \frac{E_+^2}{2E_o^2}\, y \qquad (5.50a)$$

$$y = N_o - \left[\frac{E_+^2}{E_o^2} \frac{\gamma_{ab}^{\;2}}{\gamma_{ab}^{\;2}+(kv)^2} + \frac{E_-^2}{2E_o^2} \frac{\gamma_{ab}^{\;2}-(kv)^2}{\gamma_{ab}^{\;2}+(kv)^2}\right]y - \frac{E_-^2}{2E_o^2}\, x \quad . \qquad (5.50b)$$

These simultaneous linear equations have the solution

$$x = \frac{N_o}{1 + (E_+^2 + E_-^2)\gamma_{ab}^{\;2}(\gamma_{ab}^{\;2}+(kv)^2)^{-1}} = y \quad , \qquad (5.51)$$

from which the Doppler broadened gain is calculated as

$$\frac{1}{E_\pm}\frac{dE_\pm}{dz} = \alpha \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{a e^{-\xi^2} d\xi}{(1+E_+^2+E_-^2)a^2+\xi^2}$$

$$= \frac{\alpha}{(1+E_+^2/E_o^2+E_-^2/E_o^2)^{1/2}}\, w(0+ib) \quad , \qquad (5.52)$$

where

$$b = a(1+E_+^2/E_o^2+E_-^2/E_o^2)^{1/2} \quad .$$

Saturation effects are seen to be weaker for the Doppler broadened case. For a strongly inhomogeneous line ($b \ll 1$) $w(0+ib) = 1$ giving the well known $(1+I)^{-1/2}$ behavior obtained for a single wave by several authors (6 , 10). Expanding to first order in $E_{\pm}^2/E_o^2$ the weak field result, equation 5.11, is obtained (with $x_o = y - \zeta = 0$). The important feature of equation 5.52 is that the natural linewidth is broadened by the strong fields, so that even a strongly inhomogeneous line becomes more and more homogeneous at high intensities. If $a \ll 1$ but $(E_+^2+E_-^2)/E_o^2 \rightarrow \infty$ b becomes large so that since

$$\lim_{b \rightarrow \infty} w(0+ib) = \frac{1}{\sqrt{\pi} b} \quad , \tag{5.53}$$

equation 5.52 becomes identical to the stationary atom solution 5.45. For $y - \zeta = (\omega_{+-} - \Delta\nu)/2ku = 0$ but $x_o = (\omega_o - \nu_o)/ku \neq 0$ the solutions can be written down by analogy. Only a "detuning" effect is present, all the results remaining essentially the same.

$$\frac{1}{E_\pm}\frac{dE_\pm}{dz} = \frac{\alpha_o \gamma_{ab}^2}{\gamma_{ab}^2(1+E_+^2/E_o^2+E_-^2/E_o^2) + (\omega_o - \nu_o)^2} \quad , \tag{5.54}$$

$$\frac{1}{E_\pm}\frac{dE_\pm}{dz} = \frac{\alpha}{(1+E_+^2/E_o^2+E_-^2/E_o^2)^{1/2}} \operatorname{Re} w(x_o+ib) \quad , \tag{5.55}$$

for natural and for a Doppler broadened line respectively.

To find out the behavior of the strong field solutions for $\omega_{+-} \neq \Delta\nu$, we compute numerically the values of the two contributions to the gain dE_-/dz , the part due to N_{+b} and the part due to ρ_{+-} . We call these respectively f_1 and f_2 . Thus

$$\frac{dE_-}{dz} = \alpha_o E_-(f_1 - f_2) \quad . \tag{5.48}$$

Since f_1 is the contribution of N_{+b} it includes the effects of self saturation and the common level mutual saturation. It is evident however, that since for strong fields N_{+b} is dependent on ρ_{+-} f_1 is also influenced by the other saturation processes. Figure 11 shows f_1 and f_2 as well as $f_1 - f_2$ for various values of field intensities and atomic parameters. We can immediately observe that the saturation effect of the + field is weaker than that of the - field (f_1-f_2 for $E_-^2/E_o^2 = 0.25$, $E_+^2/E_o^2 = 9$ is always larger than for $E_-^2/E_o^2 = 9$, $E_+^2/E_o^2 = 0.25$. Thus the coupling is weak and no polarization preference exists in the amplifier. In agreement with the perturbational results the nonlinear interaction of the two opposite circular fields is the strongest in the vicinity of the resonance $\omega_+ = \Delta\nu$, manifested by the dip in curves 2 and 3. For symmetrical location of the frequencies ν_+ and ν_- about the zero magnetic field line center the dip is exactly at $\omega_{+-} = \Delta\nu$, while for the asymmetrical case field strength dependent pulling effects exist causing the peak of the interaction to shift from the resonant value. Finally, we note that

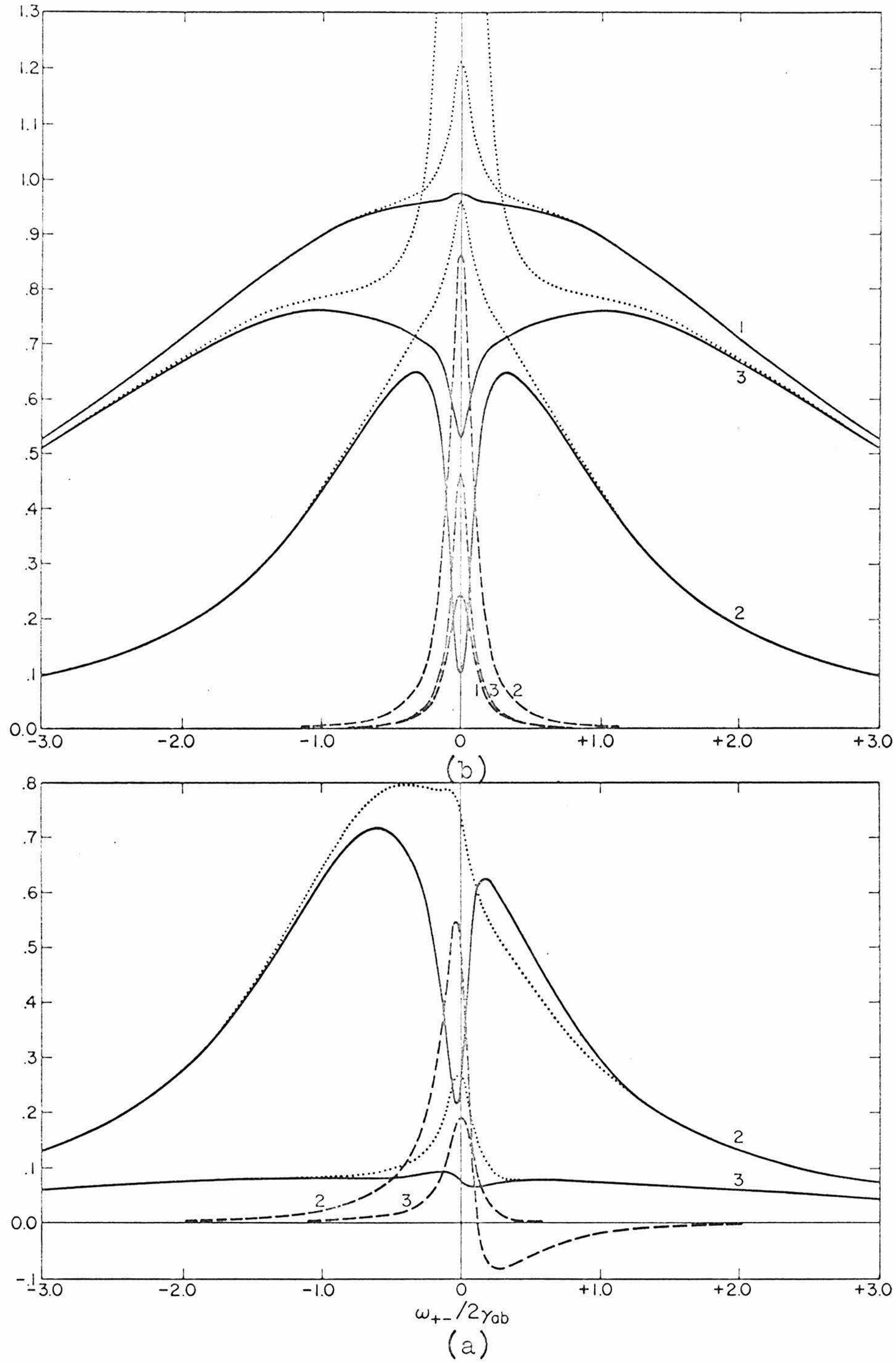


Figure 11.

The Strong Field Nonlinear Polarization Functions: Natural Broadening; γ_a/γ_{ab}= .02; (a) x_o'= 0, (b) x_o'= .5 1. E_-^2/E_o^2= 9, E_+^2/E_o^2= .25 2. E_-^2/E_o^2= .25, E_+^2/E_o^2= 9 3. E_-^2/E_o^2= E_+^2/E_o^2= 9; f_1(dotted), f_2(dashed), f_1-f_2(solid) (Scale factors, 1. (a) and 3. (a): 10f).

the width of the coherent double quantum interaction is broadened by the strong fields. While the width of the function G_3 in section 5.3 was approximately γ_a, that of f_3 is seen to be approximately $5\gamma_a$, $5\gamma_a$ and $3.5\gamma_a$ for the three cases shown respectively. The width of the total gain is also broadened. This broadening is due mostly to the same field whose gain is being examined. For case 2 ($E_-^2/E_o^2 = 0.25$, $E_+^2/E_o^2 = 9$) the width is roughly $2\gamma_{ab}$, the same as for weak field while for cases 1 and 3 ($E_-^2/E_o^2 = 9$, $E_+^2/E_o^2 = 0.25, 9$) it is of the order of $7\gamma_{ab}$ or about 3.5 times larger.

Short of performing lengthy numerical integrations for each given combination of field intensities and atomic parameters it is not possible to treat the Doppler broadened case in general. A few qualitative observations can, however, be made at this point. We have seen that for the case of weak fields the perturbational solutions have quite similar behavior for natural or Doppler broadening provided that for the former the frequencies are normalized to units of the natural linewidth γ_{ab} and in the latter to units of the Doppler parameter ku. Accordingly we expect that for strong fields the Doppler broadened solutions have characteristics similar to those for stationary atoms, with the following difference. The total width of the incremental gain should be approximately 2ku and be quite independent of the field intensities (since the Doppler width is constant) so long as the field broadened natural linewidth remains smaller than the Doppler width. No attempt will be made here to prove the validity of these assertions in a quantitative way. In the next section we return to the case of a single linearly polarized wave.

5.4.4 Single, Linearly Polarized Input Field

For this special case we set, as in section 5.3.4, $E_+ = E_- = E/\sqrt{2}$ and $\nu_- = \nu_+ = \nu = (\omega_{+b} + \omega_{-b})/2$. We have seen in section 5.3.4 that the behavior of the gain and of the Faraday rotation is particularly interesting in the region where the coherent double quantum processes are important. For weak fields this region had a width of γ_a about $\omega_{+-} = 0$. In the following we shall, for an arbitrarily Doppler broadened line, derive expressions for the induced polarization at the point $\omega_{+-} = 0$, and in the region where the contribution of the double quantum processes are negligible. This will give us the normally behaving part of the gain and Faraday rotation vs. magnetic field curve and also the depth of the dip caused by the Raman type interaction. The shape and width of the curve we calculate only approximately. For this, the previously calculated iterative solutions will also be found useful. The Faraday rotation is of course zero at $\omega_{+-} = 0$ and we shall not be able to obtain a useful expression for it in the interesting region where the double quantum processes are significant. A simple approximate method will be used to derive semiquantitative results. For the case of a naturally broadened homogeneous line a general solution, valid for arbitrary ω_{+-}, can be obtained since no integration over velocities is required. This will not be done, however, since the resulting expressions would be too complicated to have any practical value.

Let us then calculate the polarization for the Doppler broadened line. For $\omega_{+-} = 0$ it is evident that $N_{+b} = N_{-b} = N$, and we get from 5.37-5.39 after some straightforward manipulation

$$N(v) = N_o \frac{\gamma_{ab}^2 + (\gamma_{ab}\gamma_b/2)(E^2/2E_o^2 + (kv)^2)}{\gamma_{ab}^2 (1 + E^2/E_o^2) + (kv)^2} \quad , \text{ and} \tag{5.49}$$

$$\rho'_{+-} = - N_o \frac{(\gamma_b\gamma_{ab}/2)(E^2/2E_o^2)}{\gamma_{ab}^2 (1 + E^2/E_o^2) + (kv)^2} \quad . \tag{5.50}$$

These give for the polarization

$$\frac{\nu P_{\pm}}{2\epsilon_o c} = -i\alpha \frac{E}{\sqrt{2}} \int_o^{\infty} \frac{(a - i\xi)\, e^{-\xi^2}\, d\xi}{a^2 (1 + E^2/E_o^2) + \xi^2} \tag{5.51}$$

from which, using

$$\frac{dE}{dz} = \frac{\alpha E}{(1 + E^2/E_o^2)^{\frac{1}{2}}}\, w(0 + ib) \tag{5.52}$$

where $b = a(1 + E^2/E_o^2)^{\frac{1}{2}}$.

The effective linewidth is thus broadened by the factor $(1 + E^2/E_o^2)^{\frac{1}{2}}$. Equation 5.52 agrees with the results of Close (6, 7) derived with a model of scalar fields and non-degenerate levels, and is thus a useful check on the correctness of our method. In the limit as $b \rightarrow \infty$ 5.2 becomes

$$\frac{dE}{dz} = \frac{\alpha\, E_o}{1 + E^2/E_o^2} \tag{5.53}$$

which is well known homogeneous result for central tuning and no magnetic field.

In the iterative solutions the contribution of ρ_{+-} is seen to decrease as $\omega_{+-} - \Delta\nu$ changes from zero, with a width approximately γ_a. Because ρ_{+-} appears on the right hand side of equation 5.36 as well, the behavior of the contribution of the double quantum interaction to the polarization is somewhat different for strong fields. Without establishing how it falls off with increasing $\omega_{+-} - \Delta\nu$ let us, for now assume that there is a region where $\rho_{+-} \to 0$ and its influence on the induced polarization can be neglected. We shall return later to the question of the width of the coherent interaction region. For the region where $\rho_{+-} \simeq 0$ then the induced polarization is given by

$$\frac{\nu P_-(v)}{2\epsilon_o c} = -\, i\alpha_o \frac{E}{\sqrt{2}}$$

$$\frac{[\gamma_{ab}^2 - i\gamma_{ab}(\omega_{+b}-\nu+kv)][\gamma_{ab}^2+\gamma_{ab}\gamma_b E^2/4E_o^2+(\omega_{-b}-\nu+kv)^2]}{[\gamma_{ab}^2(1+E^2/2E_o^2)+(\omega_{-b}-\nu+kv)^2][\gamma_{ab}^2(1+E^2/2E_o^2)+(\omega_{+b}-\nu+kv)^2]-\gamma_a^2\gamma_{ab}^2 E^4/16E_o^4} \; . \tag{5.54}$$

Integrating over the velocity distribution

$$\frac{\nu}{2\epsilon_o c} P_- = i\alpha \frac{E}{\sqrt{2}} \frac{1}{\pi}$$

$$\int_{-\infty}^{+\infty} \frac{[a-i(y+\xi)][a^2(1+(B/a)(E^2/2E_o^2))+(y-\xi)^2]e^{-\xi^2}\,d\xi}{[a^2(1+E^2/2E_o^2)+(y+\xi)^2][a^2(1+E^2/2E_o^2)+(y-\xi)^2]-A^2a^2E^4/4E_o^4} \; . \tag{5.55}$$

For $\gamma_a \ll \gamma_b$, $B \simeq a$ and the above expression simplifies to

$$\frac{\nu}{2\epsilon_o c} P_- = - i\alpha \frac{E}{\sqrt{2}} \int_{-\infty}^{+\infty} \frac{[a-i(y+\xi)]e^{-\xi^2} d\xi}{a^2(1+E^2/2E_o^2)+(y+\xi)^2}$$

$$= \alpha \frac{E}{\sqrt{2}} \frac{1}{2b'} [(a+b')\, w^*(y+ib') + (a-b')\, w(y+ib')] \qquad (5.56)$$

where $b' = a(1+E^2/2E_o^2)^{1/2}$.

Comparing with 5.52 we see that there is, as the weak field results indicate, a dip in the gain at $\omega_{+-} = 0$, the saturation term $1 + E^2/2E_o^2$ changes to $1 + E^2/E_o^2$ as the contribution of ρ_{+-} becomes significant. The width of this dip, i.e., the region in which ρ_{+-} must be included, remains now to be determined.

We have seen from equation 5.52 that at $\omega_{+-} = 0$ the natural linewidth parameter γ_{ab} is broadened to $\gamma_{ab}(1 + E^2/E_o^2)^{1/2}$ by the strong fields. On the other hand, it is evident from 5.56 that in the region where $\rho_{+-} \simeq 0$ the broadening is somewhat less, γ_{ab} becoming $\gamma_{ab}(1 + E^2/2E_o^2)^{1/2}$. Since $\gamma_{ab} = \frac{1}{2}(\gamma_a + \gamma_b)$, the decay rates γ_a and γ_b are also broadened to $\gamma_a(1 + E^2/E_o^2)^{1/2}$ and $\gamma_b(1 + E^2/E_o^2)^{1/2}$ at $\omega_{+-} = 0$, and to $\gamma_a(1 + E^2/2E_o^2)^{1/2}$ and $\gamma_b(1 + E^2/2E_o^2)^{1/2}$ in the region where $\rho_{+-} \simeq 0$. Noting that the width of the double quantum interaction for weak fields was γ_a we can conclude that the width of the dip at $\omega_{+-} = 0$ is a quantity somewhere between $\gamma_a(1 + E^2/E_o^2)^{1/2}$ and $\gamma_a(1 + E^2/2E_o^2)^{1/2}$ This is confirmed by the numerical results of section 5.4.3. For case 3 on Figure 11 ($E_+^2/E_o^2 = E_-^2/E_o^2 = 9$) f_2 has a width approximately 3.5 γ_a . In view of the lack of a more accurate expression for it we shall henceforth take this width to be approximately $\gamma_a(1 + E^2/2E_o^2)^{1/2}$, and ρ_{+-} is then negligible when ω_{+-} is considerably larger than this quantity. The common level mutual saturation that is included in the integral of equation 5.55 should have a width roughly equal to $\gamma_b(1 + E^2/2E_o^2)^{1/2}$. It is difficult to determine, however, whether the contribution of ρ_{+-} and the effect of the common level mutual saturation combine, as they do for weak fields, to give a resultant Lorentzian interaction with width depending on γ_a alone and magnitude independent of the relative sizes of γ_a, γ_b. For the limiting case of $\gamma_a \ll \gamma_b$ the effect of the common level saturation is, of course, negligible as is seen from equation 5.56. The gain and Faraday rotation (for $\gamma_a \ll \gamma_b$, and $\omega_{+-} \gg \gamma_a(1 + E^2/2E_o^2)^{1/2}$) in that case is given by (from 5.56)

$$\frac{dE}{dz} = \frac{\alpha E}{(1 + E^2/2E_o^2)^{1/2}} \operatorname{Re} w^*(y+ib'), \tag{5.57a}$$

$$\frac{d\Phi}{dz} = \alpha \operatorname{Im} w^*(y+ib') , \tag{5.57b}$$

Let us first discuss the gain equations 5.52 and 5.57a. To get the total gain for a given amplifier and obtain the actual depth of the dip we must integrate these with respect to z. This, in general can only be done numerically, except for special cases. For a strongly inhomogeneous line, i.e., b, b' $\ll$ 1, to first order in b, b',

$$\frac{dE}{dz}\ (\omega_{+-} = 0) = \alpha E \left[\frac{1}{(1 + E^2/E_o^2)^{\frac{1}{2}}} - \frac{2a}{\sqrt{\pi}}\right] \quad , \text{ and} \qquad (5.58a)$$

$$\frac{dE}{dz}(|\omega_{+-}| > \gamma_a(1+E^2/2E_o^2)^{\frac{1}{2}}) = \alpha E \left[\frac{e^{-y^2}}{(1+E^2/2E_o^2)^{\frac{1}{2}}} - \frac{2a}{\sqrt{\pi}}\ (1-2y\ F(y))\right] \qquad (5.58b)$$

where $F(y) = e^{-y^2} \int_o^y e^{t^2}\ dt$.

These expressions can be compared with 5.28, valid for weak fields.

Equations 5.58a,b can be integrated. Rewriting them by letting $E^2/E_o^2 = I$ and $E^2/2E_o^2 = I'$ we have

$$\frac{1}{I}\frac{dI}{dz}\ (\omega_{+-} = 0) = \frac{2\alpha}{(1 + I)^{\frac{1}{2}}}\left[1 - \frac{2a}{\sqrt{\pi}}\ (1 + I)^{\frac{1}{2}}\right] \qquad (5.59a)$$

$$\frac{1}{I'}\frac{dI'}{dz}(|\omega_{+-}| > \gamma_a(1+E^2/2E_o^2)^{\frac{1}{2}}) = \frac{2\alpha\ e^{-y^2}}{(1+I')^{\frac{1}{2}}}\left[1 - \frac{2a}{\sqrt{\pi}}(1-2y\ F(y))e^{y^2}(1+I')^{\frac{1}{2}}\right] . \qquad (5.59b)$$

These are identical with equation 16 of Gordon, White and Rigden (10) with $a = \epsilon$ in 5.59a and $a(1-2y\ F(y))e^{y^2} = \epsilon$ in 5.59b. They can be integrated provided $\epsilon \ll 1$, giving

$$2\alpha L = 2(1+I_{out})^{\frac{1}{2}} - 2(1+I_{in})^{\frac{1}{2}} + \ln\left[\frac{(1+I_{in})^{\frac{1}{2}}+1}{(1+I_{in})^{\frac{1}{2}}-1}\right]\left[\frac{(1+I_{out})^{\frac{1}{2}}-1}{(1+I'_{out})^{\frac{1}{2}}+1}\right]$$

$$+ \frac{2a}{\sqrt{\pi}}\ (I_{out} - I_{in} + \ln I_{out}/I_{in}) , \qquad (5.60a)$$

for $\omega_{+-} = 0$ and

$$2\alpha L\ e^{-y^2} = 2(1+I'_{out})^{\frac{1}{2}} - 2(1+I'_{in})^{\frac{1}{2}} + \ln\left[\frac{(1+I'_{in})^{\frac{1}{2}}+1}{(1+I'_{in})^{\frac{1}{2}}-1}\right]\left[\frac{(1+I'_{out})^{\frac{1}{2}}-1}{(1+I'_{out})^{\frac{1}{2}}+1}\right]$$

$$+ \frac{2a}{\sqrt{\pi}}(1-2y\ F(y))e^{y^2}(I'_{out} - I'_{in} + \ln I'_{out}/I'_{in}) , \qquad (5.60b)$$

for $\omega_{+-} > \gamma_a(1+E^2/2E_o^2)^{\frac{1}{2}}$. This second equation is valid only if we are not too far off line center, since if y becomes large, $\frac{2a}{\sqrt{\pi}}(1-2y\ F(y))\ e^{y^2}$ can become comparable to unity even though a is small. It is sufficient to restrict y to be smaller than unity. Figure 12 shows the overall gain for various values of I_o. The "dip" at $\omega_{+-} = 0$ is 1.9 to 2.0 db deep. On the other hand if $a \to \infty$, the line is homogeneously broadened, the gain equations become

$$\frac{dI'}{dz}\ (\omega_{+-} = 0) = 2\alpha_o \frac{I}{1 + I} \quad \text{and} \qquad (5.61a)$$

$$\frac{dI'}{dz}\ (|\omega_{+-}| > \gamma_a(1+E^2/2E_o^2)^{\frac{1}{2}}) = 2\alpha \frac{I'}{1 + I' + \omega_{+-}^2/4\gamma_{ab}^2} . \qquad (5.61b)$$

These can also be integrated giving

$$2\alpha L = \ln(I/I_o) + I - I_o , \text{ and} \qquad (5.62a)$$

$$2\alpha L = [1 + \omega_{+-}^2/4\gamma_{ab}^2] \ln(I'/I'_o) + I' - I'_o . \qquad (5.62b)$$

This case is also shown on Figure 12; the "dip" is somewhat deeper. We note that even for small a as the field intensities become very strong b, b' $\to \infty$, we have, effectively, homogeneous broadening and equations 5.62a and 5.62b are valid and for very strong fields

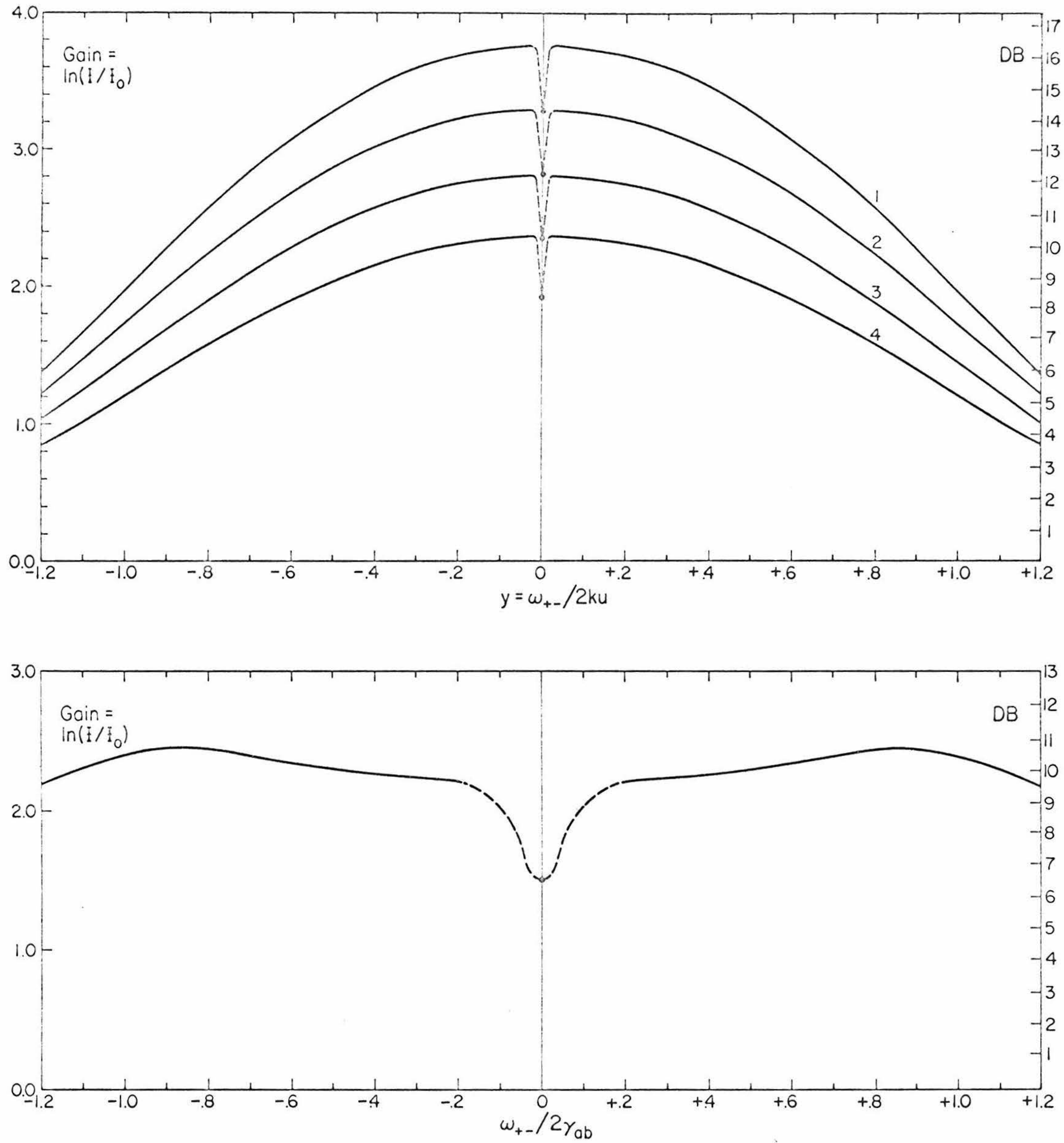


Figure 12.

Gain vs. Magnetic Field Splitting for a Linearly Polarized Wave: $\gamma_a \ll \gamma_b$; Upper: Doppler Broadened, a = 0.01, α = 3.5, 1. E_{in}^2/E_o^2 = .25 2. E_{in}^2/E_o^2 = .5 3. E_{in}^2/E_o^2 = 1 4. E_{in}^2/E_o^2 = 2; Lower; Naturally Broadened, α_o = 3.5, E_{in}^2/E_o^2 = 0.25.

the depth of the zero magnetic field dip approaches that for homogeneous broadening. For intermediate values of a this will happen for intensities that are not excessively high. (e.g., if $a = 0.4$ for $E^2/E_o^2 = 24$, $b = 2.0$ and the line is effectively homogeneously broadened.)

The Faraday rotation in the region $\omega_{+-} \gg \gamma_a(1 + E^2/E_o^2)^{\frac{1}{2}}$ is given by

$$\frac{d\Phi}{dz} = \alpha \text{ Im } w^*(y + ib') \quad . \tag{5.63}$$

Examination of this expression shows that for a Doppler broadened line the Faraday rotation is less susceptible to saturation than the gain. The parameter governing saturation of the Faraday rotation is $b' = a(1 + E^2/2E_o^2)^{1/2}$, saturation effects becoming noticeable for $b' > .05$. For a strongly inhomogeneous line this takes place only at extremely high field intensities. For $a = .01$ for example, E^2 must be as large as $50\ E_o^2$ before any decrease in the incremental rotation is noticeable. For $a = .1$ on the other hand the rotation begins to saturate for $E^2 \approx E_o^2$. This is consistent with the weak field results where the third order correction to the phase shift is negligible for small values of a (To the third order in the perturbational solutions b is identical with a). On Figure 13 we have plotted $d\Phi/dz$ the incremental rotation vs. E^2/E_o^2, the normalized field intensity, for various values of a and a fixed value of the magnetic field such that $\omega_{+-} = ku$. On the same figure is shown the incremental gain vs. E^2/E_o^2, also for the same values of a and ω_{+-}. It is seen that the saturation of the gain is essentially independent of a, while that

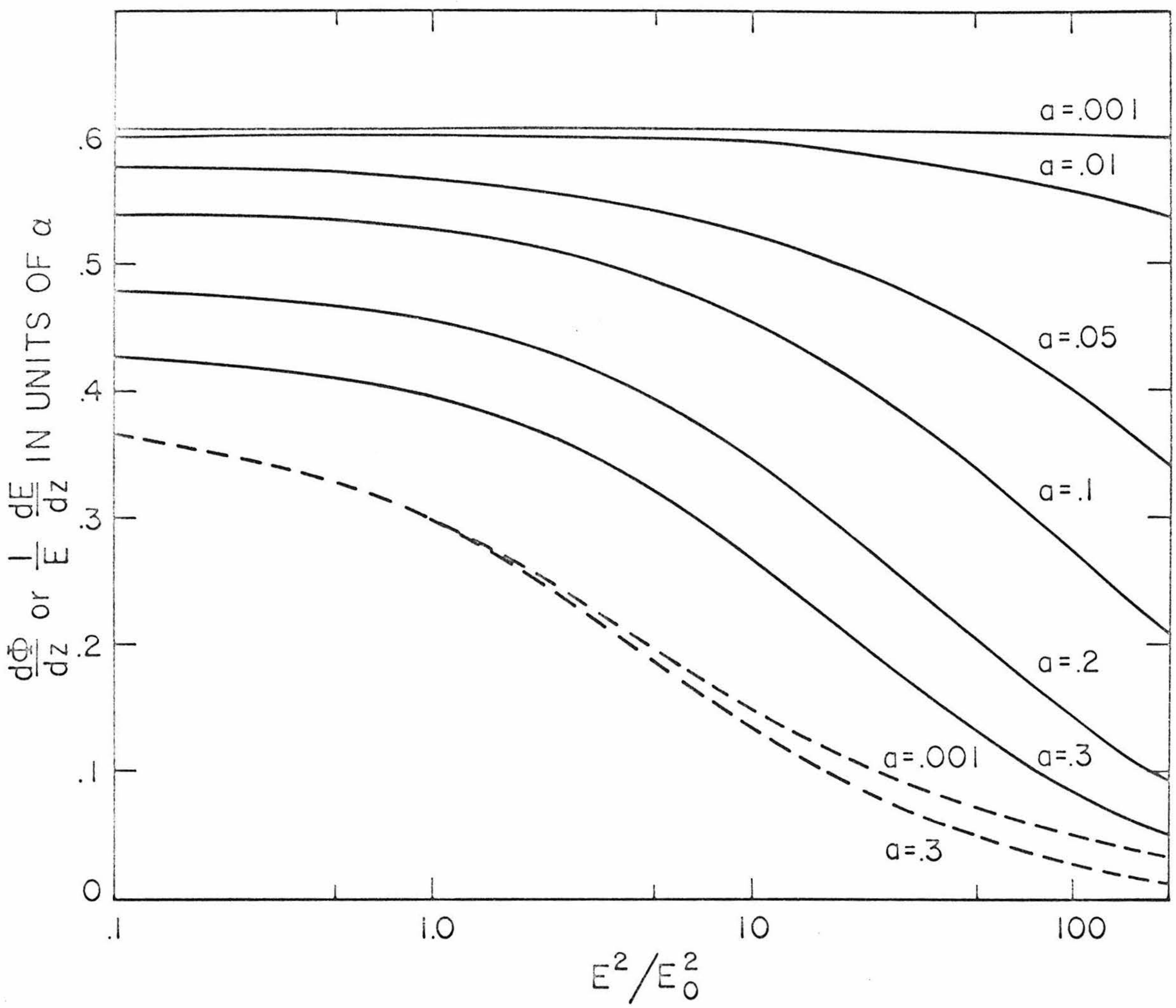


Figure 13.

Incremental Gain (dashed) and Faraday Rotation (solid) vs Field Intensity, for various values of a , and $\omega_{+-} \gg \gamma_a(1+E^2/2E_o^2)^{1/2}$.

of the rotation is a strong function of that parameter.

In the discussion of the iterative results we have noted that due to the coherent double quantum processes the Faraday rotation has an anomalous behavior in the region $|\omega_{+-}| < \gamma_a$, changing sign in the vicinity of $\omega_{+-} = \gamma_a$. For arbitrarily strong fields this region broadens to $|\omega_{+-}| < \gamma_a(1 + E^2/E_o^2)^{1/2}$. In the following we use an approximate method to estimate the magnitude of the rotation, and its dependence on the magnetic field in this region, for a laser amplifier with input field comparable to E_o^2 and output such that the perturbational results are not valid. On the basis of our discussion of the behavior of the iterative solutions and of the strong field results we shall assume that in the region $\omega_{+-} < \gamma_a(1 + E^2/2E_o^2)$ the induced polarization has a "dip" that can be approximately accounted for by subtracting a Lorentzian

$$C \frac{1}{\gamma_a(1 + E^2/2E_o^2)^{1/2} + i\omega_{+-}}$$

from the value computed without the contribution of the coherent interaction ρ_{+-}. The value of C is adjusted to give the proper depth for the central dip in the incremental gain curve. The depth of the incremental gain dip, from equation 5.52 and 5.57 is approximately $\alpha[(1 + E^2/2E_o^2)^{-1/2} w(0 + ib') - (1 + E^2/E_o^2)^{-1/2} w(0 + ib)]$, and the incremental Faraday rotation for $0 < |\omega_{+-}| < \gamma_a(1 + E^2/2E_o^2)^{1/2}$, and $\gamma_a/2ku \ll 1$ is then given approximately by

$$\frac{d\Phi}{dz} \simeq \alpha \frac{\gamma_a(1+E^2/2E_o^2)^{1/2}\omega_{+-}}{\gamma_a^2(1+E^2/2E_o^2)+\omega_{+-}^2}[(1+E^2/2E_o^2)^{-1/2}w(0+ib')-(1+E^2/E_o^2)^{-1/2}w(0+ib)], \tag{5.64}$$

since for $\gamma_{ab}/2ku \ll 1$ the rotation due to the "normal" part of the polarization is negligible in the region discussed. Equation 5.64 for $E^2/E_o^2 \ll 1$ can be expanded to first order to give

$$\frac{d\Phi}{dz} \simeq \alpha \frac{E^2}{4E_o^2} \frac{\gamma_a \ \omega_{+-}}{\gamma_a^2 + \omega_{+-}^2} \ ,$$ which agrees with the weak field result (5.31)

if in the latter we neglect the contributions of the linear part, and of F_1 , (the analogous approximation was made in obtaining 5.64) and $F_2 + F_3$ are contracted and simplified as in equation 5.27. For a given E^2 the incremental rotation over the whole range of ω_{+-} thus has the shape shown on Figure 14. On the same figure the dependence on E^2/E_o^2 of the incremental rotation in the anomalous region is plotted for various values of ω_{+-} . It must be emphasized that these curves are only approximate as is equation 5.64. It is seen that the maximum incremental rotation in this region is of the order of .06. For a linear gain of 1000 ($\alpha \simeq 3.5$) and an input field intensity of 0.5 E_o^2, the total gain from equation 5.60 is approximately 25 near $\omega_{+-} = 0$; then, without any complicated calculations, we can estimate the rotation at $\omega_{+-} = 1.6\ \gamma_a$ as α times the mean value of curve (b) of Figure 14 between E^2/E_o^2 = .5 and 12.5. Very approximately $\Phi \approx 3.5 \times 0.052 \approx .185$ radians $\approx$ 10 degrees. The - sign indicates that the rotation here is in the opposite direction from that in the normal region where $\omega_{+-} \gg \gamma_a(1 + E^2/E_o^2)^{1/2}$. The observability of this

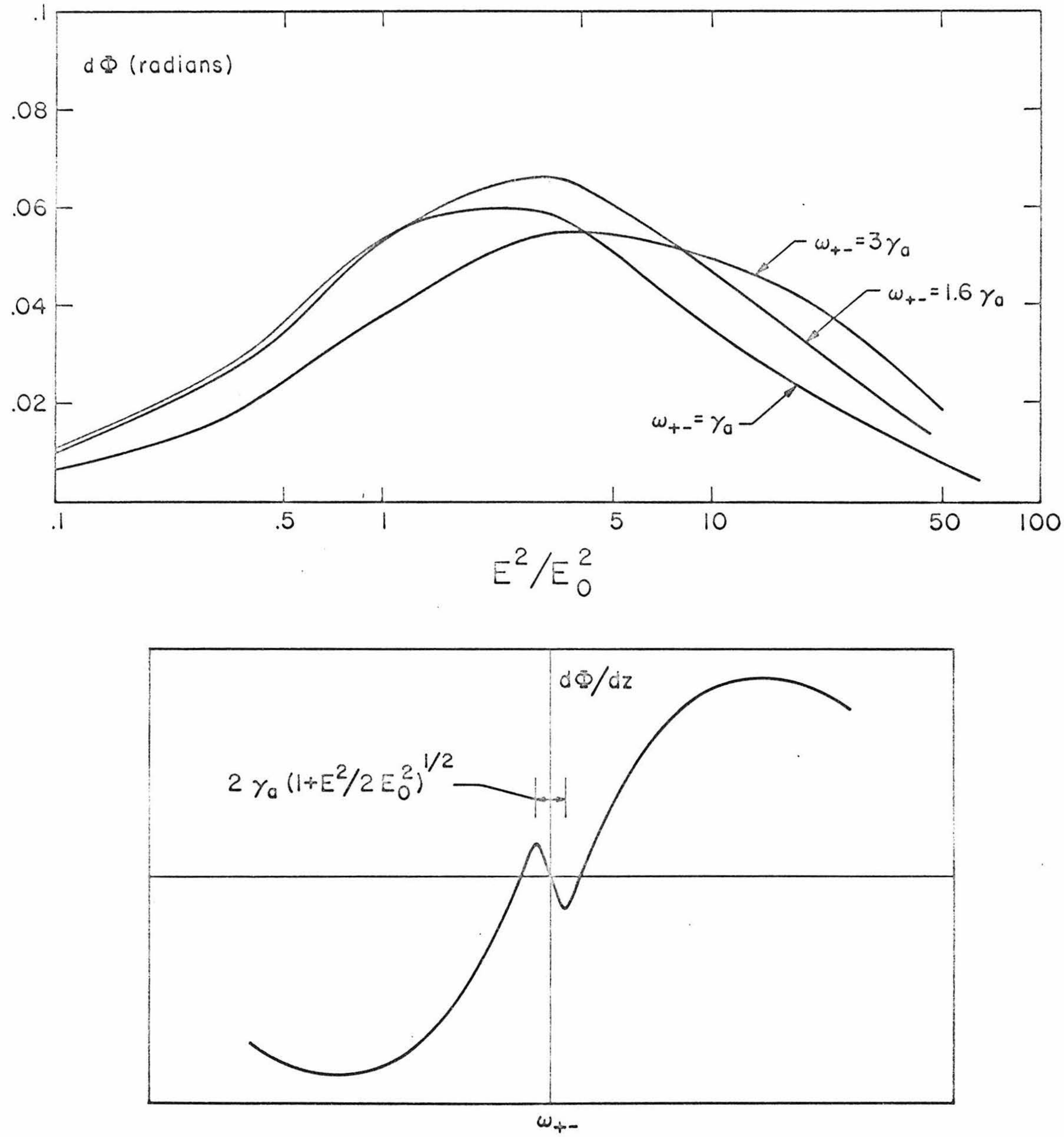


Figure 14.

The Incremental Faraday Rotation vs. Magnetic Field Splitting and vs. Field Intensity in the Anomalous Region ($\omega_{+-} \approx \gamma_a$).

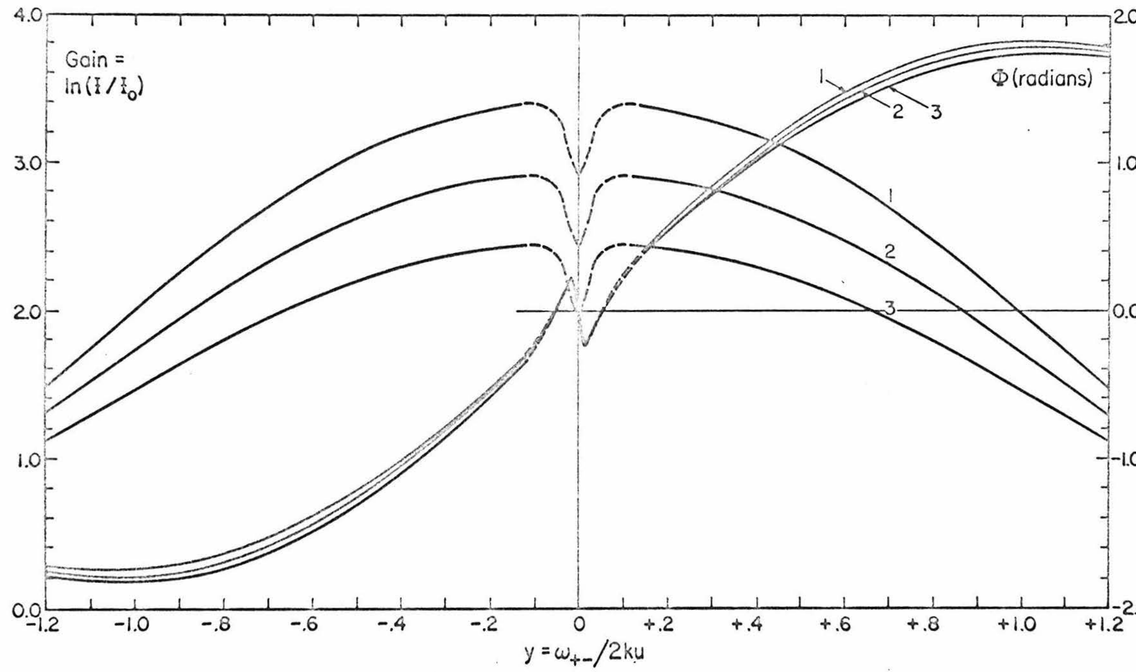


Figure 15.

Gain and Faraday Rotation vs. Magnetic Field Splitting for an Intermediate a : a = 0.3; 1. $E_{in}^2/E_o^2 = 0.5$ 2. $E_{in}^2/E_o^2 = 1.0$ 3. $E_{in}^2/E_o^2 = 2.0$; A = 0.03, B = 0.27 .

anomalous rotation is thus rather marginal. Figure 15 shows the complete curves of the total gain and Faraday rotation vs. magnetic field splitting for intermediate values of a and various input intensities. These were obtained by numerical integration of equations 5.57a and 5.57b. The dashed portions are approximate except for the point at $y = \omega_{+-}/2ku = 0$ on the gain curve which is determined by equation 5.52. The shape and magnitude of the anomalous rotation is only very roughly approximated using equation 5.60 and Figure 14. The similarity with the perturbational results is evident.

5.5 Waves of the Same Helicity Travelling in Opposite Directions

5.5.1 Introduction

So far in this chapter we have treated two opposite circularly polarized waves (opposite helicity) travelling in the same direction. These two waves interact with different transitions which have a common level, resulting in a weak interaction between the two fields. Two circularly polarized fields of the same helicity that are travelling in opposite directions also interact with different transitions since at a given point the electric vectors rotate in the opposite direction. This is also an interesting case and by a very simple modification we can derive solutions for it. To do this it is not necessary to modify the integral equations 5.2-5.4 but only to assume that either k_+ or k_- is negative. This reverses the helicity of the wave and we have exactly the case discussed. The results are useful in furthering the understanding of cavity mode interactions.

5.5.2 Homogeneously Broadened Line

No new calculations are necessary for this case, since the quantitites $k_\pm$ do not appear in the expressions for $P_\pm$ and consequently the solutions are identical to those for opposite circularly polarized waves in the same direction, derived in section 5.3 and 5.4, the only difference being that the variable is replaced by -z in one of the two nonlinear gain equations. Thus, for example, equations 5.45a,b become

$$\frac{dE_1}{dz} = \alpha E_1 \frac{1}{1 + E_2^2/E_o^2 + E_1^2/E_o^2} \qquad (5.65a)$$

$$\frac{dE_1}{dz} = -\alpha E_2 \frac{1}{1 + E_2^2/E_o^2 + E_1^2/E_o^2} \quad . \qquad (5.65b)$$

The subscripts - and + have also been replaced by 1 and 2 respectively. The identity of the expressions for the induced polarization with that for the case of two opposite circular waves in the same direction shows that the atoms being stationary they "see" no difference between a wave of a given helicity running the positive z direction and a wave of opposite helicity travelling in the negative z direction.

5.5.3 Inhomogeneously Broadened Line

For this case, since the atoms are in motion the interactions are different from those of section 5.3 and we calculate the iterative solutions. Our starting point is equation 5.9 which is modified by replacing k_+ by $-k_+$; or, since we assume as in section 5.3 that $|k_1| = |k_2|$, we set $k_+ = -k$ and $k_- = k$. The integration over v is carried out next. The details of this computation are given together with the other iterative type calculations, in Appendix I. The results are

$$\frac{\nu}{2\epsilon_o c} P_{1,2} = \alpha E_{1,2} \{w^*(x_o \pm y \mp \zeta + ia) - \frac{E_{1,2}^2}{2E_o^2} F_1(\pm y, \pm\zeta)$$

$$- \frac{E_{2,1}^2}{2E_o^2} [F_{20}(\pm y, \pm\zeta) + F_{30}(\pm y, \pm\zeta)]\} \qquad (5.66)$$

where $F_1(y,\zeta)$ is the same as the corresponding function for waves in the same direction (5.12a) and F_{20}, F_{30} are given by

$$F_{20}(y,\zeta) = A\{\frac{1}{2(a+ix_o)} [w^*(x_o+y-\zeta+ia) + w^*(x_o-y+\zeta+ia)]$$

$$+ \frac{i}{2x_o} [w^*(x_o+y-\zeta+ia) - w(x_o-y+\zeta+ia)] \qquad (5.67a)$$

$$F_{30} = AB \left\{\frac{1}{(B+ix_o)^2} [w^*(y-\zeta+iA) - w^*(x_o+y-\zeta+ia)]\right.$$

$$- \frac{1}{B+ix_o} [\frac{2}{\sqrt{\pi}} - 2(a+i(x_o+y-\zeta)) w^*(x_o+y-\zeta+ia)] + \frac{1}{B^2+x_o^2} w^*(y-\zeta+iA)$$

$$- \frac{B}{B^2+x_o^2}\,\frac{1}{2}\,[w^*(x_o+y-\zeta+ia) - w(x_o-y+\zeta+ia)]$$

$$- \frac{1}{B^2+x_o^2}\,\frac{1}{2}\,[w^*(x_o+y-\zeta+ia) + w(x_o-y+\zeta+ia)] \qquad (5.67b)$$

with the following limiting case for $x_o = 0$

$$\lim_{x_o \to 0} \frac{i}{2x_o}\,[w^*(x_o+y-\zeta+ia) - w(x_o-y+\zeta+ia)] = \frac{2}{\sqrt{\pi}} - 2(a+i(y-\zeta))\,w^*(y-\zeta+ia)\ . \qquad (5.67c)$$

The functions F_{20} and F_{30} correspond to F_2 and F_3 of section 5.3 (two opposite circularly polarized waves in the same direction). We shall discuss the properties of these functions in the same manner as we have done earlier for F_2 and F_3.

Inspection of F_{20}, the common level mutual saturation, reveals that F_{20} is identical to F_2, if in the latter we interchange x_o and $y-\zeta$. Consequently its dependence on the parameter a and on the relative magnitudes of γ_a and γ_b (A,B) is the same as that of F_2, while its variation with x_o has width $2a$ about $x_o = 0$ and its dependence on $y-\zeta$ is slow with width 2 about $y-\zeta = 0$. Once again these properties can be given simple physical interpretations in terms of holeburning. Figure 16 shows the Doppler gain curves. Only the holes made by negative helicity wave running in the positive z direction and interacting with the $J=1$, $M = +1 \rightarrow J = 0$ transition (ω_{+b}) are shown. A hole is burned by this wave in its own gain curve at the location ν_1. The atoms involved are those travelling with velocity v_R

such that $\nu_1(1-v_R/c) = \omega_{+b}$, i.e., $\nu_1 = \omega_{+b} + \nu v_R c$ (within $\pm\gamma_{ab}$). Because the common lower level is being filled up, a corresponding hole is burned into the population inversion of the other (2) wave as well. Since, however, this (2) wave is running in the opposite direction the location of this hole is at $\omega_{-b} - \nu v_R/c$, on the opposite side of the Doppler gain curve. It is evident that only when the two frequencies are within γ_{ab} of being symmetrically located about ω_o is there a significant interaction between the two waves that are travelling in opposite directions. This means $|(\nu_1+\nu_2)/2 - (\omega_{+b}+\omega_{-b})/2| < \gamma_{ab}$ which is the variation that we observe. As in section 5.3, the ratio of the depth of the hole made by a given signal in the gain curve

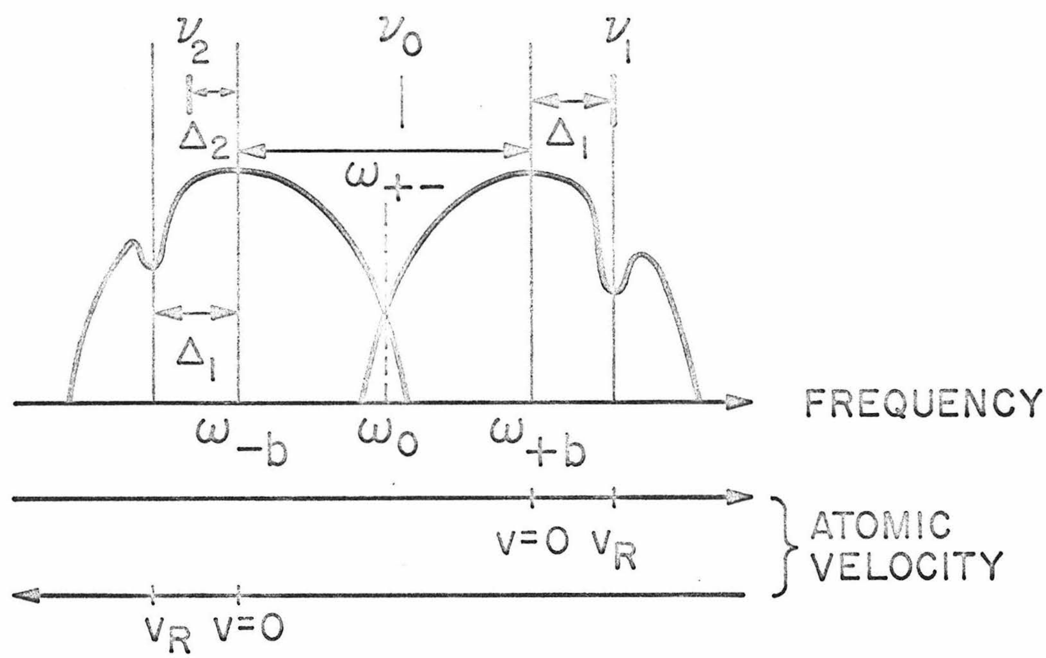


Figure 16.

of the other signal to the depth of the hole it burns in its own gain curve is equal to $\gamma_a/2\gamma_{ab}$, the ratio of the decay rate of the upper level to the sum of the decay rates. Hence the dependence of the magnitude of F_{20} on the relative sizes of γ_a and γ_b.

The function F_{30}, describing the contribution of the double quantum interaction to the third order induced polarization, has a more complicated form than the corresponding function F_3 for waves in the same direction, due to the fact that F_{20} is a product of three Doppler shifted resonance denominators. This expression can be simplified considerably for $x_o = 0$. Then

$$F_{30} = 2\,\frac{A}{B}\,\{w^*(y-\zeta+iA) - w^*(y-\zeta+ia)$$

$$- B[\,\frac{2}{\sqrt{\pi}} - 2(a+i(y-\zeta))\,w^*(y-\zeta+ia)]\} \quad . \qquad (5.68)$$

Evidently for strongly inhomogeneous line $(a, A \to 0)$ $F_{30} \to 0$. This is in contrast with the behavior of F_3 which actually increased somewhat with decreasing a, A. The reason for this can be seen from the examination of the integrals constituting F_{30} in Appendix I. For $x_- = x_+ = y-\zeta = 0$ these take the form

$$F_{30} = \int_{-\infty}^{+\infty} \frac{e^{-\xi^2}\,d\xi}{(a+i\xi)^2}\,\frac{1}{A+i\xi} \quad . \qquad (5.69)$$

$\dfrac{e^{-\xi^2}}{(a+i\xi)^2}$ can be written as $\dfrac{e^{-2i\phi(\xi)}e^{-\xi^2}}{a^2+\xi^2}$, where $\phi(\xi) = \tan^{-1}\dfrac{\xi}{a}$.

This shows that atoms moving with different velocities contribute with varying phase to the third order polarization and these contributions tend to cancel each other resulting in a small value for F_{30}. F_3, for opposite circular waves in the same direction (and for $x_- = x_+ = y-\zeta = 0$), contains the term

$$B \int_{-\infty}^{+\infty} \frac{e^{-\xi^2}}{a^2+\xi^2} d\xi \quad ,$$

where atoms moving with various velocities all contribute in phase, with a large resulting F_3. F_{30} is further decreased by the presence of the additional Doppler term $(A+i\xi)^{-1}$. Figure 17 shows the variation of F_{30} with x_o and $y-\zeta$ with an expanded ordinate. The maximum of the real part occurs at $x_o = 0$ and $y = 0$.

In the limit as a, A, B $\rightarrow \infty$ the function F_{30} should become identical to the homogeneous results. To show this we made use of the asymptotic form for large arguments (47):

$$w(z) \simeq \frac{i}{\sqrt{\pi}} \left(\frac{1}{z} - \frac{1}{2z^3}\right) \quad . \tag{5.70}$$

Keeping only the lowest order terms after considerable algebra we obtain equation 5.16c from 5.67b.

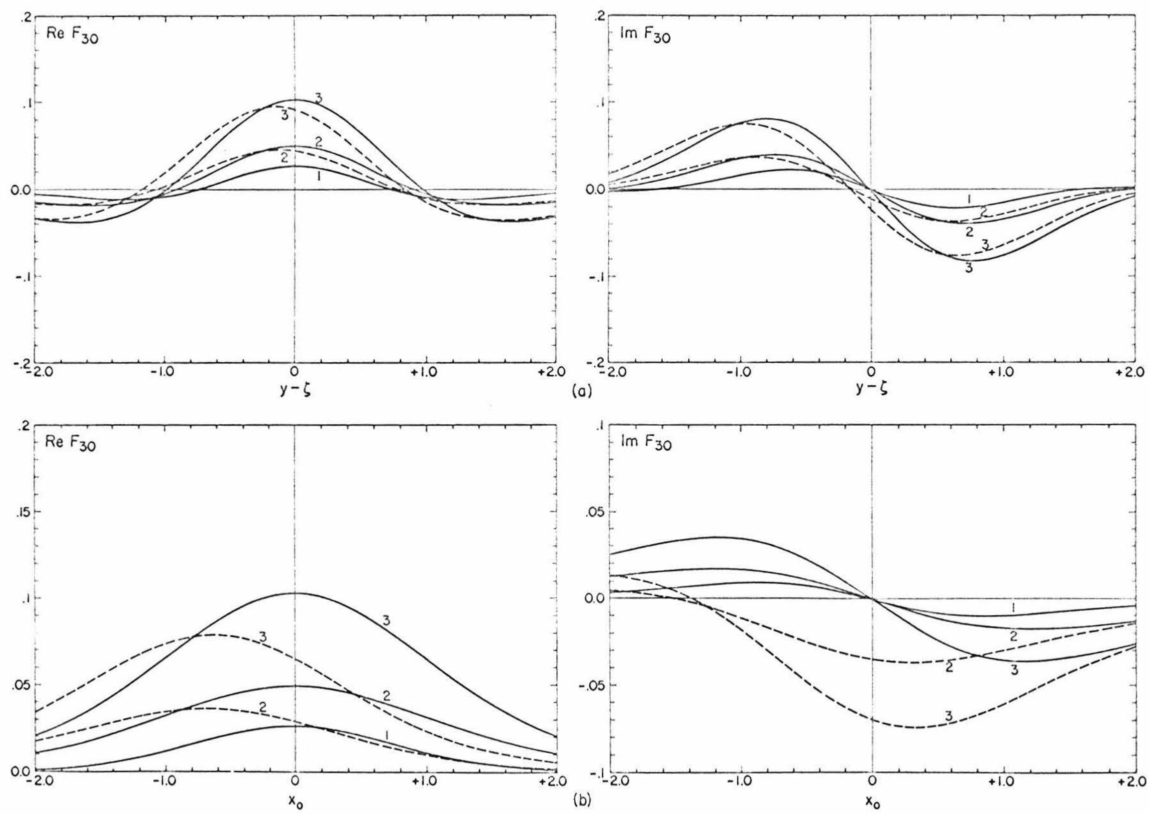


Figure 17.

The Nonlinear Polarization Function F_{30} for Oppositely Running Waves of the Same Circular Polarization: (a) vs. $y - \zeta$, $x_o = 0$ (solid), $x_o = .5$ (dashed) (b) vs. x_o, $y - \zeta = 0$ (solid), $y - \zeta = .5$ (dashed); 1. a = 0.2, A = 0.02, B = 0.18 (scale factor: $5F_{30}$) 2. a = 1.0, A = 0.1, B = 0.9 3. a = 1.0, A = 0.5, B = 0.5 .

CHAPTER SIX

TWO LINEARLY POLARIZED WAVES

6.1 Introduction

In this chapter we examine the interaction of two arbitrary linearly polarized waves in the laser amplifier by deriving, as in chapter five, the induced polarization of the medium. As it was discussed in chapter four, for the case of two arbitrary linearly polarized input fields two types of nonlinear effects are present; saturation effects of the type discussed in detail in chapter five for two opposite circularly polarized waves, and combination tone generation. Due to the fact that the induced polarization contains frequency components not present in the input, the use of the electromagnetic field equations 2.10a,b requires more careful consideration. Let us reproduce here for convenience these equations.

$$\left[\frac{\partial\varphi_{\nu m}}{\partial z} + \frac{1}{c}\frac{\partial\varphi_{\nu m}}{\partial t}\right] E_{\nu m} = \frac{\nu}{2\epsilon_o c} P_{\nu cm} \tag{6.1a}$$

$$\frac{\partial E_{\nu m}}{\partial z} + \frac{1}{c}\frac{\partial E_{\nu m}}{\partial t} = \frac{\nu}{2\epsilon_o c} P_{\nu sm} \tag{6.1b}$$

The term $\frac{\nu m}{c}(n_m - 1)$ has now been omitted from 6.1a. To calculate an index of refraction is useful only for the lowest order linear results. These having already been discussed in chapter 5 we consider only the phase shift φ .

Two possible approaches can be taken to the use of equations 6.1, 6.2. One is to include the new frequency components of the induced polarization as modulated terms at the input frequencies. In this case parts of $P_{\nu m}$ will have slow harmonic time dependence (at the difference frequencies $\Delta\nu = \nu_i - \nu_j$). Both the space and the time derivatives must be considered and rather complex nonlinear equations result. The second method, which will be used in this chapter, is to separate out the new frequency components of the polarization. In that case it is evident that fields at the new frequencies will be generated in the medium and it is necessary to include these fields in the total field E . To keep the problem from becoming too complex we can assume that these new fields will be so weak as to cause no nonlinear effects themselves. That this is a reasonable assumption has been shown by Close (6,7). In any case when any of the new fields become strong enough to cause nonlinear effects themselves the problem can be reformulated and the input fields redefined at a new spatial location. If we further assume the input fields to have narrow spectral width that can for all practical purposes be considered to be monochromatic, then we can set the time derivatives equal to zero and the electromagnetic field equations become

$$\frac{1}{c}\frac{d\varphi_{\nu m}}{dt} E_{\nu m} = \frac{\nu}{2\epsilon_o c} P_{\nu c m} \quad , \qquad (6.2a)$$

$$\frac{dE_{\nu m}}{dz} = \frac{\nu}{2\epsilon_o c} P_{\nu c m} \quad , \qquad (6.2b)$$

where the indices ν,m run over the set of the input fields, only, and we have the additional equations for the new frequency components:

$$\frac{dE\mu}{dz} = \frac{\nu}{2\epsilon_o c}\left(|P_\mu| + P'_{\mu s}\right) \quad . \tag{6.3a}$$

$$\left[\frac{\nu_\mu}{c}(n_\mu - 1)\right] E_\mu = \frac{\nu}{2\epsilon_o c} P'_{\mu c} \tag{6.3b}$$

The $|P_\mu|$'s are the absolute values of the new frequency components of the induced polarization due to the original input fields and $P'_{\mu c}$ and $P'_{\mu s}$ are the in and out of phase components of the polarization due to the new frequency fields themselves, using the linear approximation only. The initial condition on the new fields of course is $E_\mu(0) = 0$. One has to be cautious, however, about the use of equation 6.3. The absolute value $|P_\mu|$ is the significant quantity since originally the field being zero it will build up 90° out of phase with the driving term. Once a field at the new frequency is generated, at any given point it will propagate with the linear propagation constant $k_\mu = \frac{\nu_\mu n_\mu}{c}$. The polarization P_μ, on the other hand, acquires a nonlinear phase that is a combination of the phases of the input fields. This may not be equal to the phase of E_μ and the generated field and the driving term can get in phase. This problem will be examined in more detail in section 6.4 where we derive expressions for the induced polarization at the combination tone frequencies.

In the next section we shall write down the integral equations for two arbitrary linearly polarized input fields, and in sections 6.3 and 6.4 we shall calculate the polarization of the medium. Only the iterative solutions will be developed in quantitative detail, but a few qualitative remarks for strong fields will also be made with the help of the integral equations. First the components of the induced polarization oscillating at the input frequencies only will be calculated giving the interaction of the two waves in the medium. Following this the new frequency components will be derived and the dependence of the combination tone generation on various atomic parameters and on the frequencies and intensities of the input fields obtained.

6.2 The Integral Equations

For non-zero magnetic field on the laser both the gain and the phase shift will be different for the left and right circular components of each linearly polarized field. It is necessary therefore that we consider these components separately. Thus we write for the total field

$$E(t,z) = \mathrm{Re}\Big\{-\hat{e}_+ E_{1-}\, e^{i(k_1 z-\nu_1 t+\varphi_{1-})} - \hat{e}_- E_{1+} e^{i(k_1 z-\nu_1 t+\varphi_{1+})} - \hat{e}_+ E_{2-}\, e^{i(k_2 z-\nu_2 t+\varphi_{2-})} - \hat{e}_- E_{2+} e^{i(k_2 z-\nu_2 t+\varphi_{2+})}\Big\} \quad . \tag{6.4}$$

Initially, at $z = 0$, $E_{1-} = E_{1+}$, $E_{2-} = E_{2+}$ and $\varphi_{1\pm}$, $\varphi_{2\pm}$ are such as to give the desired linear polarization at the input. The propagation constants $k_{1,2} = \frac{\nu_{1,2}}{c}$ have been assumed equal for both circular components, all the dispersive effects being accounted by the nonlinear phase shifts $\varphi_{1\pm}$ and $\varphi_{2\pm}$. With the field defined by equation 6.4 the interaction matrix elements become

$$V_{-b} = (\langle a\|\rho\|b\rangle/\sqrt{3}\hbar)\left\{\frac{1}{2}\left[E_{1-}e^{+i(k_1z-\nu_1t+\varphi_{1-})}-E_{1+}e^{-i(k_1z-\nu_1t+\varphi_{1+})}\right]\right.$$
$$\left.+\frac{1}{2}\left[E_{2-}e^{+i(k_2z-\nu_2t+\varphi_{2-})}-E_{2+}e^{-i(k_2z-\nu t+\varphi_{2+})}\right]\right\} \qquad (6.5a)$$

$$V_{+b} = (\langle a\|\rho\|b\rangle/\sqrt{3}\hbar)\left\{\frac{1}{2}\left[-E_{1-}e^{-i(k_1z-\nu_1t+\varphi_{1-})}+E_{1+}e^{+i(k_1z-\nu_1t+\varphi_{1+})}\right]\right.$$
$$\left.+\frac{1}{2}\left[-E_{2-}e^{-i(k_2z-\nu_2t+\varphi_{2-})}+E_{2+}e^{+i(k_2z-\nu_2t+\varphi_{2+})}\right]\right\} . \qquad (6.5b)$$

Using these matrix elements in the integral equations we obtain the new set valid for two arbitrary linearly polarized input fields:

$$N_{+b}(z,t,v) = N^{o}_{+b}W(v) - \frac{\gamma_a\gamma_b}{4}\int_0^\infty d\tau'\int_0^\infty d\tau''$$
$$\left\{\frac{E_{1-}^2}{E_o^2}\left[e^{-\gamma_a\tau'}+e^{-\gamma_b\tau'}\right]e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_1+k_1v)]\tau''}\right.$$
$$+\frac{E_{1-}E_{2-}}{E_o^2}\left[e^{-[\gamma_a+i(\nu_2-\nu_1-(k_2-k_1)v)]\tau'}+e^{-[\gamma_b+i(\nu_2-\nu_1-(k_2-k_1)v]\tau'}\right]$$
$$\times e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_1+k_1v)]\tau''}e^{+i[(k_1-k_2)z+(\nu_2-\nu_1)t]}e^{i(\varphi_1-\varphi_{2-})}$$
$$\left.+\text{ same with } 1\rightleftarrows 2\right\}N_{+b}(z,t-\tau'-\tau'',v)-\frac{\gamma_a\gamma_b}{4}\int_0^\infty d\tau'\int_0^\infty d\tau''$$

$$\left\{\frac{E_{1+}^2}{E_o^2}\, e^{-\gamma_b\tau'}\, e^{-[\gamma_{ab}+i(\omega_{-b}-\nu_1+k_1v)]\tau''} + \frac{E_{1+}E_{2+}}{E_o^2}\, e^{-[\gamma_b+i(\nu_2-\nu_1-(k_2-k_1)v)]\tau'}\right.$$

$$\times\, e^{-[\gamma_{ab}+i(\omega_{-b}-\nu_1+k_1v)\tau''}\, e^{+i[(k_1-k_2)z+(\nu_2-\nu_1)t]}\, e^{i(\varphi_{1+}-\varphi_{2+})}$$

$$\left.+\text{ same with } 1\rightleftarrows 2\right\} N_{-b}(z,t-\tau'-\tau'',v) - \frac{\gamma_a\gamma_b}{4}\int_0^\infty d\tau'\int_0^\infty d\tau''\left\{\frac{E_{1+}E_{1-}}{E_o^2}\left[\left(e^{-\gamma_a\tau'}+e^{-\gamma_b\tau'}\right)\right.\right.$$

$$\left.\times\, e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_1+k_1v)]\tau''} + e^{-\gamma_b\tau'}\, e^{-[\gamma_{ab}-i(\omega_{-b}-\nu_1+k_1v)]\tau''}\right] e^{i(\varphi_{1+}-\varphi_{1-})}$$

$$+\frac{E_{1+}E_{2-}}{E_o^2}\left[\left(e^{-[\gamma_a+i(\nu_2-\nu_1-(k_2-k_1)v)]\tau'}+e^{-[\gamma_b+i(\nu_2-\nu_1-(k_2-k_1)]\tau'}\right)\right.$$

$$\times\, e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_1+k_1v)]\tau''} + e^{-[\gamma_b+i(\nu_2-\nu_1-(k_2-k_1)v]\tau'}$$

$$\left.\times\, e^{-[\gamma_{ab}-i(\omega_{-b}-\nu+k_1v)]\tau''}\right] e^{i[(k_1-k_2)z+(\nu_2-\nu_1)t]}\, e^{i(\varphi_{1+}-\varphi_{2-})}$$

$$\left.+\text{ same with } 1\rightleftarrows 2\right\}\rho_{+-}(z,t-\tau'-\tau'',v) + \text{c.c.}\,, \qquad (6.6)$$

$N_{-b}(z,t,v)$ = same as $N_{+b}(z,t,v)$ but with + and - interchanged in all subscripts,

$$\rho_{+-}(z,t,v) = \frac{\gamma_a\gamma_b}{4}\int_0^\infty d\tau'\int_0^\infty d\tau''$$

$$\left\{\frac{E_{1-}E_{1+}}{E_o^2}\, e^{-(\gamma_a+i\omega_{+-})\tau'}\, e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_1+k_1v)]\tau''}\, e^{i(\varphi_{1-}-\varphi_{1+})}\right.$$

$$+\frac{E_{1-}E_{2+}}{E_o^2}\, e^{-[\gamma_a+i(\omega_{+-}-(\nu_1-\nu_2)+(k_1-k_2)v)]\tau'}\, e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_1+k_1v]\tau''}$$

$$\times e^{i[(k_1-k_2)z+(\nu_2-\nu_1)t]} e^{i(\varphi_{1-}-\varphi_{2+})} + \text{same with } 1 \rightleftarrows 2 \Big\} N_{+b}(z,t-\tau'-\tau'',v)$$

$$- \frac{\gamma_a\gamma_b}{4} \int_0^\infty d\tau' \int_0^\infty d\tau'' \Big\{ \frac{E_{1-}E_{1+}}{E_0^2} e^{-(\gamma_a+i\omega_{+-})\tau'} e^{-[\gamma_{ab}-i(\omega_{-b}-\nu_1+k_1v)]\tau} e^{i(\varphi_{1-}-\varphi_{1+})}$$

$$+ \frac{E_{1-}E_{2+}}{E_0^2} e^{-[\gamma_a+i(\omega_{+-}-(\nu_1-\nu_2)+(k_1-k_2)v)]\tau'} e^{-[\gamma_{ab}-i(\omega_{-b}-\nu_2+k_2v)]\tau''} e^{i(\varphi_{1-}-\varphi_{2+})}$$

$$+ \text{same with } 1 \rightleftarrows 2 \Big\} N_{-b}(z,t-\tau'-\tau'',v)$$

$$- \frac{\gamma_a\gamma_b}{4} \int_0^\infty d\tau' \int_0^\infty d\tau'' \Big\{ \frac{E_{1+}^2}{E_0^2} e^{i(\gamma_a+i\omega_{+-})\tau'} e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_1+k_1v)]\tau''}$$

$$+ \frac{E_{1+}E_{2+}}{E_0^2} e^{-[\gamma_a+i(\omega_{+-}-(\nu_1-\nu_2)+(k_1-k_2)v)]\tau'} e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_1+k_1v)]\tau''}$$

$$\times e^{i[(k_1-k_2)z+(\nu_2-\nu_1)t]} e^{i(\varphi_{1+}-\varphi_{2+})} + \text{same with } 1 \rightleftarrows 2 \Big\} \rho_{+-}(z,t-\tau'-\tau'',v)$$

$$- \frac{\gamma_a\gamma_b}{4} \int_0^\infty d\tau' \int_0^\infty d\tau'' \Big\{ \frac{E_{1-}^2}{E_0^2} e^{i(\gamma_a+i\omega_{+-})\tau'} e^{-[\gamma_{ab}-i(\omega_{-b}-\nu_1+k_1v)]\tau''}$$

$$+ \frac{E_{1-}E_{2-}}{E_0^2} e^{-[\gamma_a+i(\omega_{+-}-(\nu_1-\nu_2)+(k_1-k_2)v)]\tau'} e^{-[\gamma_{ab}-i(\omega_{-b}-\nu_1+k_1v)]\tau''}$$

$$\times e^{i[(k_1-k_2)z+(\nu_2-\nu_1)t]} e^{i(\varphi_{1-}-\varphi_{2-})} + \text{same with } 1 \rightleftarrows 2 \Big\} \rho_{+-}(z,t-\tau'-\tau'',v) \quad . \tag{6.7}$$

And finally, in terms of the above quantities,

$$P_-(z,t,v) = -i\,\frac{|\langle a\|p\|b\rangle|^2}{3\hbar}\int_0^\infty d\tau\Big\{E_{1-}\,e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_1+k_1v)]\tau}\,e^{i(k_1z-\nu_1t+\varphi_{1-})}$$

$$+E_{2-}\,e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_2+k_2v)]\tau}\,e^{i(k_2z-\nu_2t+\varphi_{2-})}\Big\}N_{+b}(z,t-\tau,v)$$

$$-i\,\frac{|\langle a\|p\|b\rangle|^2}{3\hbar}\int_0^\infty d\tau\Big\{E_{1+}e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_1+k_1v)]\tau}\,e^{i(k_1z-\nu_1t+\varphi_{1+})}$$

$$+E_{2+}e^{-[\gamma_{ab}+i(\omega_{+b}-\nu_2+k_2v)]\tau}\,e^{i(k_2z-\nu_2t+\varphi_{2+})}\Big\}\rho_{+-}(z,t-\tau,v)\quad. \qquad (6.8)$$

$P_+(z,t,v)$ = same with + and - interchanged in all subscripts.
All the symbols have been previously defined. The note "same with $1 \rightleftarrows 2$" calls for repeating all the preceding terms within the same integral but with subscripts 1 and 2 interchanged everywhere.

6.3 The Iterative Solutions. Saturation Effects

Using the same method as in chapter five we can now in a straightforward manner find the first order, linear and the third, lowest, order nonlinear solution for the polarization. It is evident from examining equations 6.6-6.8 that, as predicted in chapter four, the induced polarization has new frequency components. These are due to the modulation of the population inversions $N_{\pm b}$ at the difference frequency $\nu_2 - \nu_1$ and are also influenced by the multiple quantum interactions through the contributions of ρ_{+-} . In addition, the components of the polarization oscillating at the input frequencies contain terms that were not present for two opposite circular waves. We expect these new terms to influence the coupling between the two input fields.

6.3.1 Saturation Effects

The following zeroeth and second order terms contribute to the first and third order polarization at the input frequency ν_1 (for simplicity we set, as in chapter five, $N^o_{+b} = N^o_{-b} = N_o$ and $k_1 = k_2 = k$ anywhere but in the exponentials).

$$N_{+b}^{(0)+(2)} = N_o W(v)\Big\{1 - \frac{E_{1-}^2}{2E_o^2}\left(\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)} + \frac{\gamma_{ab}}{\gamma_{ab}-i(\omega_{+b}-\nu_1+kv)}\right)$$

$$- \frac{E_{2-}^2}{2E_o^2}\left(\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_2+kv)} + \frac{\gamma_{ab}}{\gamma_{ab}-i(\omega_{+b}-\nu_2+kv)}\right) - \frac{E_{1-}E_{2-}}{E_o^2}\,\frac{\gamma_a\gamma_b}{4}$$

$$\times\left(\frac{1}{\gamma_a+i(\nu_2-\nu_1)} + \frac{1}{\gamma_b+i(\nu_2-\nu_1)}\right)\left(\frac{1}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)} + \frac{1}{\gamma_{ab}-i(\omega_{+b}-\nu_2+kv)}\right)$$

$$\times e^{i[(k_1-k_2)z-(\nu_1-\nu_2)t]}\, e^{i(\varphi_1-\varphi_2)}$$

$$- \frac{E_{1+}^2}{2E_o^2}\,\frac{\gamma_a}{2\gamma_{ab}}\left(\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{-b}-\nu_1+kv)} + \frac{\gamma_{ab}}{\gamma_{ab}-i(\omega_{-b}-\nu_1+kv)}\right)$$

$$- \frac{E_{2+}^2}{2E_o^2}\frac{\gamma_a}{2\gamma_{ab}}\left(\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{-b}-\nu_2+kv)}+\frac{\gamma_{ab}}{\gamma_{ab}-i(\omega_{-b}-\nu_2+kv)}\right)$$

$$- \frac{E_{1+}E_{2+}}{E_o^2}\frac{\gamma_a\gamma_b}{4}\frac{1}{\gamma_b+i(\nu_2-\nu_1)}\left(\frac{1}{\gamma_{ab}+i(\omega_{-b}-\nu_1+kv)}+\frac{1}{\gamma_{ab}-i(\omega_{-b}-\nu_2+kv)}\right)$$

$$\times e^{i[(k_1-k_2)z-(\nu_1-\nu_2)t]}e^{i(\varphi_{1+}\varphi_{2+})} \quad . \tag{6.9}$$

$N_{-b}^{(0)+(2)}(z,t,v)$ = same, but with + and - interchanged in all subscripts.

$$\rho_{+-}^{(2)}(z,t,v) = -N_o W(v)\left\{\frac{E_{1-}E_{1+}}{E_o^2}\frac{\gamma_a\gamma_b}{4}\frac{1}{\gamma_a+i\omega_{+-}}\right.$$

$$\times\left(\frac{1}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}+\frac{1}{\gamma_{ab}-i(\omega_{-b}-\nu_1+kv)}\right)e^{i(\varphi_{1-}-\varphi_{1+})}$$

$$+\frac{E_{2-}E_{2+}}{E_o^2}\frac{\gamma_a\gamma_b}{4}\frac{1}{\gamma_a+i\omega_{+-}}\left(\frac{1}{\gamma_{ab}+i(\omega_{-b}-\nu_2+kv)}+\frac{1}{\gamma_{ab}-i(\omega_{-b}-\nu_2+kv)}\right)$$

$$\times e^{i[(k_1-k_2)z-(\nu_1-\nu_2)t]}e^{i(\varphi_{1-}-\varphi_{2+})}+\frac{E_{1-}E_{2+}}{E_o^2}\frac{\gamma_a\gamma_b}{4}\frac{1}{\gamma_a+i(\omega_{+-}-(\nu_1-\nu_2))}$$

$$\times\left(\frac{1}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}+\frac{1}{\gamma_{ab}-i(\omega_{-b}-\nu_2+kv)}\right)e^{i[(k_1-k_2)z-(\nu_1-\nu_2)t]}e^{i(\varphi_{1-}-\varphi_{2+})} . \tag{6.10}$$

There are other terms in $N_{+b}^{(0)+(2)}$ and $\rho_{+-}^{(2)}$ but they do not contribute to the polarization at ν_1. Finally, combining 6.8, 6.9 and 6.10 we get for the left circular component of the polarization at the frequency ν_1

$$\frac{\nu}{2\epsilon_o c} P_{1-}^{(1)+(3)}(v) = -i\alpha_o W(v) E_{1-}\left\{\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\right.$$

$$-\frac{E_{1-}^2}{2E_o^2}\left[\left(\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\right)^2+\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\,\frac{\gamma_{ab}}{\gamma_{ab}-i(\omega_{+b}-\nu_1+kv)}\right]$$

$$-\frac{E_{2-}^2}{2E_o^2}\left[\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\,\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_2+kv)}+\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\right.$$

$$\left.\times\frac{\gamma_{ab}}{\gamma_{ab}-i(\omega_{+b}-\nu_2+kv)}\right]-\frac{E_{2-}^2}{2E_o^2}\,\frac{\gamma_a\gamma_b}{2\gamma_{ab}}\left[\frac{1}{\gamma_a+i(\nu_2-\nu_1)}+\frac{1}{\gamma_b+i(\nu_2-\nu_1)}\right]$$

$$\times\left[\left(\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\right)^2+\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\,\frac{\gamma_{ab}}{\gamma_{ab}-i(\omega_{+b}-\nu_2+kv)}\right]$$

$$-\frac{E_{1+}^2}{2E_o^2}\,\frac{\gamma_a}{2\gamma_{ab}}\left[\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\,\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{-b}-\nu_1+kv)}\right.$$

$$\left.+\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\,\frac{\gamma_{ab}}{\gamma_{ab}-i(\omega_{-b}-\nu_1+kv)}\right]$$

$$-\frac{E_{2+}^2}{2E_o^2}\,\frac{\gamma_a}{2\gamma_{ab}}\left[\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\,\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{-b}-\nu_2+kv)}\right.$$

$$\left.+\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\,\frac{\gamma_{ab}}{\gamma_{ab}-i(\omega_{-b}-\nu_2+kv)}\right]-\frac{E_{1+}^2}{2E_o^2}\,\frac{\gamma_a\gamma_b}{2\gamma_{ab}}\,\frac{1}{\gamma_a+i\omega_{+-}}$$

$$\times\left[\left(\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\right)^2+\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\frac{\gamma_{ab}}{\gamma_{ab}-i(\omega_{-b}-\nu_1+kv)}\right]$$

$$-\frac{E_{2+}^2}{2E_o^2}\frac{\gamma_a\gamma_b}{2\gamma_{ab}}\frac{1}{\gamma_a+i(\omega_{+-}-(\omega_{+-}-(\nu_1-\nu_2))}\left[\left(\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\right)^2\right.$$

$$\left.\left.+\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\frac{\gamma_{ab}}{\gamma_{ab}-i(\omega_{-b}-\nu_2+kv)}\right]\right\}-i\alpha_o W(v)\frac{E_{2-}E_{1+}E_{2+}}{2E_o^2}$$

$$\times\left\{\frac{\gamma_a\gamma_b}{2\gamma_{ab}}\frac{1}{\gamma_b+i(\nu_2-\nu_1)}\left[\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{-b}-\nu_1+kv)}\right.\right.$$

$$\left.+\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\frac{\gamma_{ab}}{\gamma_{ab}-i(\omega_{-b}-\nu_2+kv)}\right]$$

$$+\frac{\gamma_a\gamma_b}{2\gamma_{ab}}\frac{1}{\gamma_a+i\omega_{+-}}\left[\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_2+kv)}\right.$$

$$\left.\left.+\frac{\gamma_{ab}}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\frac{\gamma_{ab}}{\gamma_{ab}-i(\omega_{-b}-\nu_2+kv)}\right]\right\}e^{i[(\varphi_{2-}-\varphi_{2+})-(\varphi_{1-}-\varphi_{1+})]}\quad . \quad (6.11)$$

The right circular component $\frac{\nu}{2\varepsilon_o c}P_{1+}^{(1)+(3)}(v)$ is same with + and - subscripts interchanged. Similar expressions hold for $\frac{\nu}{2\varepsilon_o c}P_{2\pm}$ which we can also get from the above by replacing 1 by 2 and vice versa in all subscripts. Using the same method as in the previous chapter we calculate the final expression for the polarization by integrating over the velocity distribution. Only the case of an arbitrary Doppler broadening will be considered. (For details of calculations see Appendix I) The result is :

$$\frac{\nu}{2\varepsilon_o c} P_{1-} = i\alpha E_{1-}\Big\{w^*(x_o+y-\eta+ia) - \frac{E_{1-}^2}{2E_o^2} H_1(x_o,y,\eta) - \frac{E_{1+}^2}{2E_o^2}\Big[H_2(x_o,y,\eta)+H_3(x_o,y,\eta)\Big]$$

$$+\frac{E_{2-}^2}{2E_o^2}\Big[H_4(x_o,y,\eta)+H_5(x_o,y,\eta)\Big] - \frac{E_{2+}^2}{2E_o^2}\Big[H_6(x_o,y,\eta)+H_7(x_o,y,\eta)\Big]\Big\}$$

$$-i\alpha\frac{E_{1+}E_{2-}E_{2+}}{2E_o^2}\Big[H_8(x_o,y,\eta)+H_9(x_o,y,\eta)\Big] \tag{6.12}$$

where $H_1 = F_1$, $H_6 = F_2$, $H_7 = F_3$ with $\zeta \to \eta$ and the other six functions are given by

$$H_2(x_o,y,\eta) = A\Big\{\frac{i}{2y}\Big[w^*(x_o+y-\eta+ia)-w^*(x_o-y-\eta+ia)\Big]$$

$$+\frac{1}{2(a+iy)}\Big[w^*(x_o+y-\eta+ia)+w(x_o-y-\eta+ia)\Big]\Big\} \tag{6.13}$$

$$H_3(x_o,y,\eta) = \frac{AB}{A+iy}\Big\{\frac{2}{\sqrt{\pi}} - 2\big(a+i(x_o+y-\eta)\big)w^*(x_o+y-\eta+ia)$$

$$+\frac{1}{2(a+iy)}\Big[w^*(x_o+y-\eta+ia)+w(x_o-y-\eta+ia)\Big]\Big\} \tag{6.14}$$

$$H_4(x_o,y,\eta) = a\left\{\frac{i}{2\eta}\left[w^*(x_o+y-\eta+ia) - w^*(x_o+y-\eta+ia)\right]\right.$$

$$\left.+\frac{1}{2(a-i\eta)}\left[w^*(x_o+y-\eta+ia)+w(x_o+y+\eta+ia)\right]\right\} \qquad (6.15)$$

$$H_5(x_o,y,\eta) = AB\left(\frac{1}{A-i\eta}+\frac{1}{B-i\eta}\right)\left\{\frac{2}{\sqrt{\pi}} - 2(a+i(x_o+y-\eta))w^*(x_o+y-\eta+ia)\right.$$

$$\left.+\frac{1}{2(a-i\eta)}\left[w^*(x_o+y-\eta+ia)+w(x_o+y+\eta+ia)\right]\right\} \qquad (6.16)$$

$$H_8(x_o,y,\eta) = \frac{AB}{B-i\eta}\left\{\frac{i}{2y}\left[w^*(x_o+y-\eta+ia)-w^*(x_o-y-\eta+ia)\right]+\frac{1}{2[a+i(y-\eta)]}\right.$$

$$\left.+\left[w^*(x_o+y+\eta+ia)+w(x_o-y+\eta+ia)\right]\right\}e^{i[(\varphi_{2-}-\varphi_{2+})-(\varphi_{1-}-\varphi_{1+})]} \qquad (6.17)$$

$$H_9(x_o,y,\eta) = \frac{AB}{A+iy}\left\{\frac{i}{2\eta}\left[w^*(x_o+y-\eta+ia)-w^*(x_o+y-\eta+ia)\right]\right.$$

$$\left.\frac{1}{2[a+i(y-\eta)]}\left[w^*(x_o+y-\eta+ia)+w(x_o-y+\eta+ia)\right]\right\}e^{i[(\varphi_{2-}-\varphi_{2+})-(\varphi_{1-}-\varphi_{1+})]}$$

$$(6.18)$$

With the following limiting cases

$$\lim_{y\to 0}\frac{i}{2y}\left[w^*(x_o+y-\eta+ia)-w^*(x_o-y-\eta+ia)\right]=\frac{2}{\sqrt{\pi}}-2(a+i(x_o-\eta))w^*(x_o-\eta+ia) \qquad (6.19)$$

$$\lim_{y\to 0}\frac{i}{2\eta}\left[w^*(x_o+y+\eta+ia)-w^*(x_o+y-\eta+ia)\right]=\frac{2}{\sqrt{\pi}}-2(a+i(x_o+y))w^*(x_o+y+ia) \qquad (6.20)$$

The quantity η is defined by $\eta = (\nu_1 - \nu_2)/2ku$, half the separation of the input signal frequencies. All others are identical to those defined and used in chapter five. The superscript (1) + (3) indicating that first and third order terms are included has now been omitted. To obtain the other three polarization components P_{1+}, P_{2-}, and P_{2+} the following simple rules can be used.

1) $P_{1m} \rightarrow P_{2m}$ Interchange subscripts 1 and 2
Let $\eta \rightarrow -\eta$

2) $P_{i-} \rightarrow P_{i+}$ Interchange subscripts + and -
Let $y \rightarrow -y$

6.3.2 Discussion

Since we are free to specify arbitrary initial conditions on the phases $\varphi_{1\pm}$, $\varphi_{2\pm}$, equations 6.12-6.20 are valid not only for two linearly but also for two arbitrary elliptically polarized input fields. In fact, even for linearly polarized inputs the outputs will in general be elliptically polarized because the left and right circular components experience different gains and phase shifts. We shall examine the following two cases, where the fields remain (at least approximately) linearly polarized, in some detail.

1) Zero magnetic field, arbitrary tuning of input frequencies ($\omega_{+-}=0$)
2) Nonzero magnetic field, signals tuned symmetrically about zero magnetic field line center and their separation small compared to the Doppler width ($\omega_{+-}\neq 0$; $(\nu_1+\nu_2)/2 = (\omega_{+b}+\omega_{-b})/2$; $\nu_1-\nu_2 \ll ku$)

First, however, let us briefly discuss the general solutions. Although at first glance the equation for P_{1-} appears exceedingly complicated the separate functions $H_1, \ldots 9$, have easily recognizably characteristics and for these physical explanations can also be given. In doing this we are aided by the knowledge gained in chapter five about the interaction of two opposite circularly polarized waves.

Each of the nine functions contain two terms (each in a square bracket) and certain multiplying factors that depend mostly on the individual decay rates. For strong Doppler broadening the first term is negligibly small while for homogeneous broadening ($a \to \infty$) the two terms contribute equally. Thus the nonlinear effects for the former are only half as strong as for the latter.* H_1 is the self saturation term of the - component of the field at ν_1. It is exactly equal to F_1 and has the same physical explanation.

H_2 describes the common level mutual saturation interaction between the - and the + components of one linearly polarized wave (ν_1), and H_3 is the contribution of the corresponding double quantum interaction of the atoms. Accordingly, the properties of these functions are identical with F_2 and F_3 respectively with $y - \zeta$ replaced by y (since the left and right circular components have the same frequency) and x_o replaced by $x_o - \eta$ (since $x_o - \eta$ is the detuning of the frequency ν_1 from line center).

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* This is of course valid for weak fields only. In chapter five we have seen that for strong fields the difference in saturation characteristics shows up as the $(1+E^2/E_o^2)^{-1/2}$ and $(1+E^2/E_o^2)^{-1}$field strength dependence for the inhomogeneous and homogeneous cases respectively. Expanding to first order gives the factor of 2 difference when the fields are weak.

H_4 is a new term not corresponding to any of the terms of two opposite circular fields in chapter five. It is caused by the competition for the same population inversion by two different fields of the same polarization. Thus it has an equivalent in the scalar theory of Bennett (12) and in the two nondegenerate level approach of Close (6). The hole burned by one field in the gain curve for the same polarization influences the gain of the other field if the frequency difference is less than the natural linewidth. Since the same transition is involved the magnitude of the interaction is independent of the relative size of the decay rates. H_4 thus has a width of 2a about $\eta=0$. In addition, the center of the transition being at $x_o+y=0$, plotted vs. that variable a slow variation with a width equal to the Doppler width about $x_o+y=0$ will be observed.

The function H_5 again describes an interaction not previously encountered, the parallel of which is found in Close's theory (6). It results from the coherent modulation of the population inversion density by the two field at the difference frequency $\Delta\nu = \nu_1-\nu_2$, giving rise to "sideband" generation as discussed in chapter four. Two of the sidebands coincide with the original frequencies (for example, $\nu_1-\Delta\nu = \nu_2$) and contribute to the induced polarization and thus to the gain and phase correction of the input fields. H_5 describes this contribution. The important characteristic of this term is the appearance of the factor $[(A-i\eta)^{-1}+(B-i\eta)^{-1}]$ which cause it to decrease as η becomes larger than A and B (i.e., $\Delta\nu > \gamma_a$, γ_b). Physically, this is due to the fact that only when the frequency difference is smaller than the decay rates is the population inversion capable of following the pulsations of the field. The width of H_5 with η is thus some sort of mean of the two decay rates, about $\eta=0$. As expected a Doppler variation, identical to that of H_4, with x_o+y is also present.

H_6 and H_7 describe the interaction with the opposite circularly polarized component of the other field. Predictably the functions are identical to F_2 and F_3 respectively and all the discussion of chapter five applies.

H_8 and H_9 are the contributions of the interaction of three field components to the induced polarization of the fourth (e.g., of E_{1+}, E_{2-} and E_{2+} to P_{1-}). These functions contain a multiplying factor that is dependent on the phase difference of the circularly polarized components and obviously these are the terms that determine the dependence of the interaction on the angle between the polarization vectors of the input fields (or more generally, on the polarization characteristics of the elliptical input waves). H_8 is a "sideband" term somewhat similar to H_5, but quite different too at the same time. The modulation of one circularly polarized population inversion (of N_{-b} by E_{1+} and E_{2+} for the case described by 6.12 and 6.17) induces, because of the common lower level, a sideband with the opposite circular polarization (-) at the input frequency (at ν_1 for the case discussed). Accordingly, the characteristics of both the common level mutual saturation (e.g. F_2, H_6) and of "sideband" terms appear together in H_8. The interaction is proportional to the ratio $\gamma_a/2\gamma_{ab}$, i.e., it is small when the decay rate γ_b is large and the common level empties rapidly, and has the characteristic dependence on $(\omega_{+-}-\Delta\nu)/2$ with width $2\gamma_{ab}$ about zero, shown by the $[a+i(y-\eta)]^{-1}$ multiplier of the second, significant term. In addition, however it contains the multiplier $(B-i\eta)^{-1}$. This shows that the common level population must be following the pulsations of the fields for this term to remain significant. The composite dependence on the two variables y,η is rather complicated. It is evident however that the magnetic field must be small for if y is large the two conditions $y-\eta=0$ and $\eta=0$ cannot be simultaneously fulfilled.

H_9, finally, is the somewhat complimentary interaction to H_8, being proportional to γ_b/γ_{ab}. It can be described as follows. A simultaneous, Raman type, interaction of the atom with the fields E_{2+} and E_{2-}, for example, takes place resulting in a coherence between the upper sublevels. The resulting ρ_{+-} contributes to the polarization at P_{1-} by another interaction of the atom with E_{1+}. For the double quantum interaction the Raman condition $\omega_{+-}=0$ $(y=0)$ has to be satisfied. (The condition is as shown, since $\nu_+-\nu_- = \nu_2-\nu_2 = 0$). Since the lower level acts as a virtual level the width here is γ_a. The additional condition that $\nu_1-\nu_2=\omega_{+-}$ $(y-\eta=0)$ must also be fulfilled for the induced coherence to contribute to P_{1-}, the width here being the natural linewidth. The resulting final dependence on y and η is again complex, as it was for H_8. The frequency separation must be small, however, since otherwise the two conditions $y = 0$ and $y = \eta$ cannot be simultaneously satisfied. A Doppler variation with x_o is additionally present in both H_8 and H_9.

In conclusion we note that for strong Doppler broadening (a=0, pure inhomogeneous case) and the limiting case of $x_o=y=0$, $\eta\to 0$ we have $H_1 = H_4 = H_5 = 1$, $H_2 = H_6 = H_8 = A/a = \gamma_a/2\gamma_{ab}$ and $H_3 = H_7 = H_9 = B/a = \gamma_b/2\gamma_{ab}$, while for natural broadening ($a \to \infty$, homogeneous case) the functions have twice the above values, provided in the latter case we replace α by α_o.

6.3.3 Zero Magnetic Field

This is an important limiting case which has been examined to some extent previously by Haken and Sauermann (8) and by Close (6), using a model of two nondegenerate levels and a method of averaging over possible atomic dipole orientations. We shall make a quantitative comparison with their results.

Let us assume that the phases accumulated by the left and right circular components will be of the form

$$\varphi_{j-}(z) = -\pi/2 + \Phi_j(z) + \varphi_j(z) \tag{6.21a}$$

$$\varphi_{j+}(z) = -\pi/2 - \Phi_j(z) + \varphi_j(z) \quad . \tag{6.21b}$$

We shall see below why these are both suitable and appropriate. Then the fields are

$$\bar{E}_j(t,z) = \mathrm{Re}\left\{\left\{(\hat{e}_x/\sqrt{2})\left[(E_{j-}+E_{j+})\sin\Phi_j - i(E_{j-}-E_{j+})\cos\Phi_j\right]\right.\right.$$
$$\left.\left. + (\hat{e}_y/\sqrt{2})\left[(E_{j-}+E_{j+})\cos\Phi_j + i(E_{j-}-E_{j+})\sin\Phi_j\right]\right\} e^{i(k_j z - \nu_j t + \varphi_j)}\right\} \; . \tag{6.22}$$

If $E_{j-} = E_{j+} = E_j/\sqrt{2}$ this reduces to

$$\overline{E}_j(t,z) = \text{Re}\left\{[\hat{e}_x \sin \Phi_j + \hat{e}_y \cos \Phi_j]E_j e^{i(k_j z-\nu_j t+\varphi_j)}\right\} , \tag{6.23}$$

which is a linearly polarized wave with polarization $\hat{e}_x \sin \Phi_j + \hat{e}_y \cos \Phi_j$ and an accumulated nonlinear phase shift φ_j .

We shall now assume that $E_{j-} = E_{j+} = E_j/\sqrt{2}$, i.e., the waves remain linearly polarized, but we allow Φ_j to be a function of z, that is the direction of polarization may be changed by the nonlinear interactions in the medium. (It will be seen later that in fact E_{j-} and E_{j+} do not remain equal except under special circumstances) It is seen from equations 6.12-6.18 that for the case discussed

$$\frac{\nu}{2\epsilon_o c}P_{1\pm}(z,t) = \left[A+Be^{\mp i2(\Phi_2-\Phi_1)}\right]e^{i(k_1 z-\nu_1 t+\varphi_{1\pm})} , \tag{6.24}$$

where we have omitted the Re , indicating the real part. In the above ,

$$A = -i\alpha \frac{E_1}{\sqrt{2}} \left\{w^*(x_o-\eta+ia)- \frac{E_1^2}{4E_o^2}\left[H_1(x_o,0,\eta)+H_2(x_o,0,\eta)\right.\right.$$
$$\left.\left. + H_3(x_o,0,\eta)\right]- \frac{E_2^2}{4E_o^2}\left[H_4(x_o,0,\eta)+H_5(x_o,0,\eta)+H_6(x_o,0,\eta)+H_7(x_o,0,\eta)\right]\right\} \tag{6.25a}$$

$$B = -i\alpha \frac{E_1}{\sqrt{2}} \left\{- \frac{E_2^2}{4E_o^2}\left[H_8(x_o,0,\eta)+H_9(x_o,0,\eta)\right] \right\} \quad . \tag{6.25b}$$

After using a relation analogous to 6.22 and some trigonometry we obtain

$$\frac{\nu}{2\epsilon_o c} P_{1p}(z,t) = \left[\sqrt{2}\, A + \sqrt{2}\, B \cos 2(\Phi_2 - \Phi_1)\right] e^{i(k_1 z - \nu_1 t + \varphi_1)} \qquad (6.26a)$$

$$\frac{\nu}{2\epsilon_o c} P_{1q}(z,t) = \left[\sqrt{2}\, B \sin 2(\Phi_2 - \Phi_1)\right] e^{i(k_1 z - \nu_1 t + \varphi_1)} \quad . \qquad (6.26b)$$

The subscripts p and q indicate the projections parallel and perpendicular to the polarization vector of the field $E_1(\hat{e}_{p1} = \hat{e}_x \sin\Phi_1 + e_y \cos\Phi_1)$. The angle $\Phi_2 - \Phi_1$ is the angle between the polarization vectors of the two fields. The wave dependence of P has been included to show that it has the same propagation characteristics as E_1 and the field equations 6.2a,b, can be used. Equation 6.26a describes the dependence of the saturation characteristics of the medium on the angle between the polarization vectors of the two fields. The significance of equation 6.26b is that it shows that unless this angle is zero or 90° new fields, polarized perpendicularly to the direction of the fields are generated. We shall discuss this further after more detailed examination of 6.26a.

After noting that when y=0 $H_2 + H_3 = H_1$, using the identity $\cos 2\theta = 2\cos^2\theta - 1$ and observing that (again for y=0) $H_4 - H_9 = H_6$ and $H_5 - H_8 = H_7$, equation 6.38a can be rewritten as

$$\frac{\nu}{2\epsilon_o c} P_{1p} = -i\alpha E_1 \left\{ w^*(x_o - \eta + ia) - \frac{E_1^2}{2E_o^2} Q_1(x_o, \eta) - \frac{E_2^2}{2E_o^2} \left[Q_2(x_o, \eta) + Q_3(x_o, \eta)\cos^2\theta \right] \right\} , \quad (6.27)$$

where

$$Q_1(x_o, \eta) = a\left[\frac{2}{\sqrt{\pi}} - 2(a+i(x_o-\eta))w^*(x_o-\eta+ia) + \frac{1}{a}\mathrm{Re}\ w^*(x_o-\eta+ia))\right] , \quad (6.28)$$

$$Q_2(x_o, \eta) = a\left[\frac{i}{2\eta}(w^*(x_o-\eta+ia) - w^*(x_o-\eta+ia)) + \frac{1}{a-i\eta}(w^*(x_o+\eta+ia) + w(x_o+\eta+ia))\right] + \frac{AB}{A-i\eta}\left[\frac{2}{\sqrt{\pi}} - 2(a+i(x_o-\eta))w^*(x_o-\eta+ia) + \frac{1}{a-i\eta}(w^*(x_o-\eta+ia) + w(x_o+\eta+ia))\right] , \quad (6.29)$$

$$Q_3(x_o, \eta) = \frac{AB}{B-i\eta}\left[\frac{2}{\sqrt{\pi}} - 2(a+i(x_o-\eta))w^*(x_o-\eta+ia) + \frac{1}{a-i\eta}(w^*(x_o-\eta+ia) + w(x_o+\eta+ia))\right] + B\left[\frac{i}{2\eta}(w^*(x_o+\eta+ia) - w^*(x_o-\eta+ia)) + \frac{1}{a-i\eta}(w^*(x_o-\eta+ia) + w(x_o+\eta+ia))\right] , \quad (6.30)$$

$\theta = \Phi_2 - \Phi_1$, the angle between the polarization vectors of the two fields E_1 and E_2 . We note that Q_2 and Q_3 are the same but with A and B interchanged. In exactly the same manner we obtain for the field at ν_2

$$\frac{\nu}{2\epsilon_o c} P_{2p} = -i\alpha E_1 \Big\{ w^*(x_o+\eta+ia) - \frac{E_2^2}{2E_o^2} Q_1(x_o-\eta) - \frac{E_1^2}{2E_o^2}\Big[Q_2(x_o,-\eta)+Q_3(x_o,-\eta)\cos^2\theta\Big]\Big\} \quad . \tag{6.31}$$

These equations can now be compared to those of Close (6) and of Haken and Sauermann (8). The function Q_1 describing the self saturation, which, not surprisingly, is identical with the corresponding function F_1 for opposite circularly polarized fields, is the same as that of Close. The interaction terms $Q_2+Q_3 \cos^2\theta$, however, are somewhat different. A factor $(1+2\cos^2\theta)/3$ was obtained by that author as multiplying the whole interaction term. In the results obtained by our more rigorous method, because of the different frequency dependence of Q_2 and Q_3, such simplification is not possible. Moreover we see that in the limit of strong Doppler broadening, $x_o = 0$ and $\eta \to 0$, i.e., central tuning and small frequency separation, $Q_2 = Q_3 = 1$ and the interaction term equals $1 + \cos^2\theta$ rather than the value $(2/3)(1+\cos^2\theta)$ his equations give at the same limit. Haken and Sauermann, using similar methods have obtained the same factor $(1 + 2 \cos^2\theta)/3$ and thus exactly the same comments apply.

The gain and the phase shift for the fields at ν_1 and ν_2 are given by

$$\frac{1}{E_{1,2}} \frac{dE_{1,2}}{dz} = \alpha \text{ Re} \left\{ \begin{matrix} \text{from} \\ 6.37, \ 6.41 \end{matrix} \right\} \tag{6.32a}$$

$$\frac{d\varphi_{1,2}}{dz} = \alpha \text{Im} \left\{ \begin{matrix} \text{from} \\ 6.37, \ 6.41 \end{matrix} \right\} \quad . \tag{6.32b}$$

Let us now examine the strength of the interaction between two linearly polarized waves. In the limit as $x_o = 0$, $\eta \to 0$ (central tuning, small frequency separation) $Q_1 = Q_2 = Q_3 = 1$ and 2 for strong Doppler broadening and for homogeneous broadening respectively (for the latter one must replace α by α_o) In either case we see that, by the definition of chapter five, the coupling is weak if $\theta = 90^\circ$ (the fields are perpendicularly polarized), while for $\theta = 90^\circ$ the coupling is strong. The strongest coupling occurs at $\theta = 0^\circ$ (parallel polarization) when the effect of E_2 on the gain of E_1 is twice as strong as that of E_1 on itself. This is in agreement with the results of Close (6) and also agrees with those of Doyle and White (25) derived for a laser oscillator. To obtain the detailed frequency dependence of this phenomenon we first specialize our results for strong Doppler broadening. For that case equations 6.27-6.32 further simplify and we obtain the remarkably simple result:

$$\frac{1}{E_{1,2}}\frac{dE_{1,2}}{dz} = \alpha\left\{1-\frac{E_{1,2}^2}{2E_o^2}-\frac{E_{2,1}^2}{2E_o^2}\left[\frac{\gamma_a^2}{\gamma_a^2+(\Delta\nu)^2}+\frac{\gamma_b^2}{\gamma_a^2+(\Delta\nu)^2}\cos^2\theta\right]\right\}e^{-(\omega_o-\nu_o)^2/(ku)^2} . \tag{6.33}$$

For given γ_a, γ_b, $\Delta\nu$ the angle at which transition from strong to weak coupling occurs is given by

$$\cos^2\theta = \frac{(\Delta\nu)^2}{\gamma_a^2+(\Delta\nu)}\frac{\gamma_b^2+(\Delta\nu)^2}{\gamma_b^2} . \tag{6.34}$$

If the two polarizations are parallel $(\theta = 0)$ the transition from strong to weak coupling occurs at $\eta = (AB)^{1/2}$, i.e., $\Delta\nu = (\gamma_a\gamma_b)^{1/2}$ in agreement with the results of Lamb (5) and of Doyle and White (25) for oscillators. Figure 18 shows the variation of the coupling with $\Delta\nu$ for various values of θ.

6.3.3A Anisotropic Effects

We now examine the hitherto neglected question whether E_{j+} and E_{j-} remain equal or not as the fields propagate through the medium and whether Φ_j, describing the orientation of the polarization vectors, is changed by the nonlinear interactions. To do this we must go back to the expressions for the circular components of the induced polarization, equation 6.24. Since

$$\frac{dE_{1\pm}}{dz} = -\mathrm{Im}\,\frac{\nu}{2\epsilon_o c}P_{1\pm} = -\left[\mathrm{Im}A+\mathrm{Im}B\cos 2\theta\mp\mathrm{Re}\sin 2\theta\right] , \tag{6.35}$$

$$\frac{1}{E_{1\pm}}\frac{d\varphi_{1\pm}}{dz} = \mathrm{Re}\,\frac{\nu}{2\epsilon_o c}P_{1\pm} = \mathrm{Re}A + \mathrm{Re}B\cos 2\theta \pm \mathrm{Im}B\sin 2\theta , \tag{6.36}$$

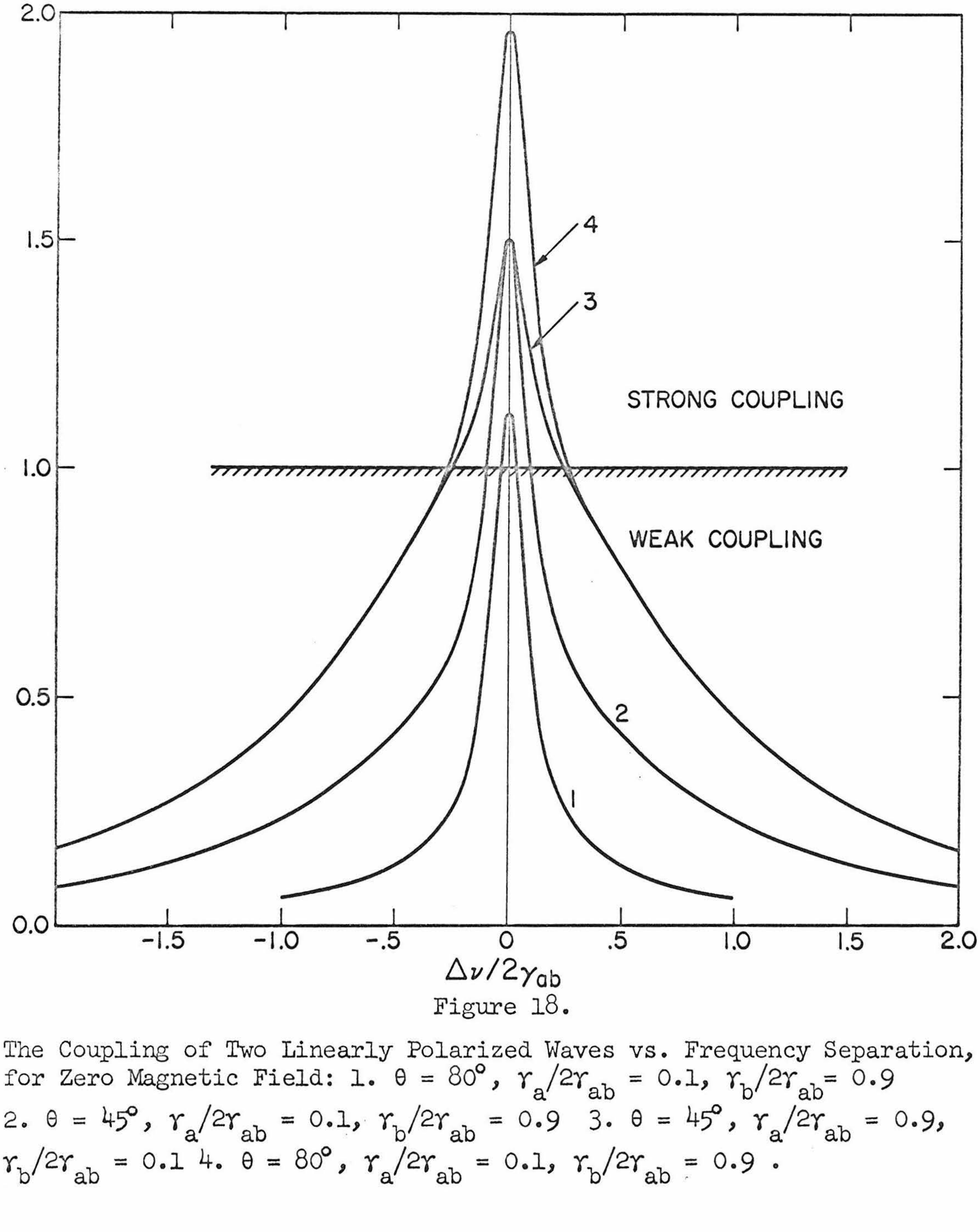

Figure 18.

The Coupling of Two Linearly Polarized Waves vs. Frequency Separation, for Zero Magnetic Field: 1. $\theta = 80°$, $\gamma_a/2\gamma_{ab} = 0.1$, $\gamma_b/2\gamma_{ab} = 0.9$ 2. $\theta = 45°$, $\gamma_a/2\gamma_{ab} = 0.1$, $\gamma_b/2\gamma_{ab} = 0.9$ 3. $\theta = 45°$, $\gamma_a/2\gamma_{ab} = 0.9$, $\gamma_b/2\gamma_{ab} = 0.1$ 4. $\theta = 80°$, $\gamma_a/2\gamma_{ab} = 0.1$, $\gamma_b/2\gamma_{ab} = 0.9$.

It is evident that even if initially $E_{j+} = E_{j-}$ (as they are in the above equations) neither the gain nor the phase shift is the same for opposite circularly polarized components. The medium is rendered effectively anisotropic for one field by the presence of the other. The effect of E_{j+} and E_{j-} becoming unequal is that the linearly polarized fields slowly change into elliptical, and of Φ_j changing is that their orientation changes. If the first of these two effects is large then the method of combining the circular components to obtain linearly polarized fields is incorrect. In the foregoing we shall show that under certain conditions it is legitimate to neglect this effect and we can treat the fields as remaining approximately linearly polarized. The magnitude of the change in Φ_j will also be estimated.

From equation 6.35 we get for $E_{2+} = E_{2-}$

$$d(E_{1-}-E_{1+})/dz = \alpha E_1 (E_2^2/2E_o^2)\mathrm{Im}Q_3(x_o,\eta)\sin 2\theta \ , \tag{6.37}$$

which for strong Doppler broadening can be written as

$$d(E_{1-}-E_{1+})/dz = \alpha E_1 \frac{E_2^2}{E_o^2} \frac{\gamma_b \Delta\nu}{\gamma_b^2+(\Delta\nu)^2} e^{-(\omega_o-\nu_o)^2/(ku)^2} \sin 2\theta \ . \tag{6.38}$$

The value of this function is zero for $\theta = 0$ or $\pi/2$ and is maximum for $\pi/4$. $d(E_{1-}-E_{1+})/dz$ also goes to zero if $\Delta\nu \ll \gamma_b$, i.e., if the frequency separation is small. To estimate the magnitude of the unbalance between the circular components we note that if $\Delta\nu = \gamma_b$, $\theta = \pi/4$, $E_2^2/E_o^2 = .5$ and $\omega_o - \nu_o = 0$, then, very approximately, $E_{1-} - E_{1+} \simeq .125\, \alpha z (E_1)_{mean}$. Take $\alpha z = 3.5$, corresponding to a linear amplitude gain of 33. The laser is saturated so assume the actual gain is only 16. Then $E_{1-} - E_{1+} \simeq .2 E_{1\ final}$, i.e. the "unbalance" between + and - components is about 20%. This is of course under conditions optimum for this effect. For small $\Delta\nu$ and/or θ close to 0 or 90 degrees it is much smaller. We shall henceforth neglect this effect and continue to treat the fields as being linearly polarized. This assumption will break down seriously only for $\Delta\nu \sim \gamma_b$ and θ close to 45 degrees.

To compute the change of orientation we write, from equations 6.36 and the corresponding ones for $\varphi_{2\pm}$, since $\theta = \frac{1}{2}[(\varphi_{2-}-\varphi_{2+}) -(\varphi_{1-}-\varphi_{1+})]$

$$\frac{d\theta}{dz} = \alpha \operatorname{Re} \frac{E_1^2+E_2^2}{4E_o^2} Q_3(x_o,\eta) \sin 2\theta , \qquad (6.39)$$

which for strong Doppler broadening becomes

$$\frac{d\theta}{dz} = \alpha \frac{E_1^2+E_2^2}{4E_o^2} \frac{\gamma_b^2}{\gamma_b^2+(\Delta\nu)^2} e^{-(\omega_o-\nu_o)^2/(kv)^2} \sin 2\theta . \qquad (6.40)$$

This indicates that the fields tend to rotate apart, unless the angle between them is zero or 90°. This tendency is largest for $\Delta\nu \ll \gamma_b$

(this is also the condition for the fields to remain truly linearly polarized). For such a case, with $\alpha z = 3.5$, $(E_1/E_o^2)_{mean} = (E_2^2/E_o^2)_{mean} \simeq .5$ and $\theta(0) = \pi/4$ we estimate

$$\theta - \theta(0) \simeq .44 \text{ radians} = 25^\circ \ .$$

Thus the rotation should be easily detectable for gains of 30 db or higher provided we are not too far from the optimum conditions assumed here.

Finally, we turn our attention to equation 6.26b which predicts the generation of a new field at the frequency ν_1 with polarization perpendicular to that of E_1 . This expression can be rewritten as

$$\frac{\nu}{2\epsilon_o c} P_{1q} = i\alpha E_1 \frac{E_2^2}{4E_o^2} Q_3(x_o, \eta) \sin 2\theta ,$$

which equals

$$i\alpha E_1 \frac{E_2^2}{4E_o^2} \frac{\gamma_b}{\gamma_b - i\Delta\nu} e^{-(\omega_o - \nu_o)^2/(ku)^2} \sin 2\theta , \qquad (6.41)$$

for strong Doppler broadening. There being initially no field E_{1q} it will build up out of phase with the driving term, leading it by 90° . Thus the initial phase of the new field is

$$\varphi_{1q} = \varphi_{1p} + \tan^{-1} \frac{-\Delta\nu}{\gamma_b} + \frac{\pi}{2} \quad . \qquad (6.42)$$

In general this is not equal to φ_{1p} and the new field is not in phase with the original one. If $\Delta\nu \ll \gamma_b$, however, $\varphi_{1q} = \varphi_{1p}$ and the new field adds to the original one causing a rotation of the resultant field which remains linearly polarized. For $\Delta\nu \sim \gamma_b$ the rotation is still present except that the resultant field has become slightly elliptical. We now assert that this is not a different effect but only a restatement of the rotation of the polarizations calculated on the previous pages and that equation 6.41 expresses the same information, in a different form, as equation 6.40. To show this first we note that in circularly polarized form we have eight independent equations, four for gain and four for phase shift. In the previous calculations we have in fact considered exactly that number: two equations for $d\varphi_{1,2}/dz$ (by adding $d\varphi_{j-}$ and $d\varphi_{j+}$), one for $d\theta/dz = d(\Phi_2 - \Phi_1)/dz$ (by combining all four phase differentials); a fourth one, $d(\Phi_2 + \Phi_1)/dz$ is evidently identically zero; two gain equations for dE_{1p}/dz, dE_{2p}/dz (by adding and subtracting according to formula 6.22); and two others for $d(E_{1-}-E_{1+})/dz$, $d(E_{2-}-E_{2+})dz$ making a total of eight. Thus we have considered all eight independent variables and equation 6.41 can only be restating an already calculated one in a different form. Next we show that in fact 6.40 can be obtained from 6.41. The new orientation of $\hat{e}_{p1}$ is given by $\Phi_1 = \Phi_1 + \tan(E_{1q}/E_{1p}) = \Phi_1 + (E_{1q}/E_{1p})$, since E_{1q} is small. Then

$$\frac{d\theta}{dz} = \frac{d(E_{2q}/E_{2p})}{dz} - \frac{d(E_{1q}/E_{1p})}{dz} = \alpha\, \frac{E_1^2+E_2^2}{4E_o^2}\, \frac{\gamma_b^2}{\gamma_b^2+(\Delta\nu)^2}\, e^{-[(\omega_o-\nu_o)/ku]^2} \sin 2\theta, \tag{6.43}$$

which is identically equal to equation 6.40. We should also remark at this point that even if $\Delta\nu \simeq \gamma_b$ expression 6.43 should remain essentially correct. Although the fields will become somewhat elliptically polarized due to the unbalance between the left and right circular components of E_{1p}, E_{2p}, or equivalently, because the new fields E_{1q}, E_{2q} are partially out of phase with the input fields, the major axes of the polarization ellipses will rotate apart and their incremental separation should still be given by equation 6.44.

6.3.3B Summary and Conclusions

Several interesting conclusions can be drawn from the results derived and discussed in the preceding section. To begin with, the strength of the coupling between two linearly polarized fields is seen to be a function of the upper and lower laser level decay rates, the frequency separation of the signals, and the angle between the polarization vectors. For any angle other than 90 degrees there is a critical frequency separation, below which strong coupling exists. Physically this is due to the fact that for parallel polarization of the fields (and for any angle other than 90° there exists finite projection of one parallel to the other) the signals compete for the same population inversions if their frequency separation is within the natural linewidth.

The most interesting result that emerges from the examination of the no magnetic field case is the effective anisotropy of the medium due to the interaction of the two linearly polarized optical signals. This is manifested by a characteristic rotation of the electric field vectors in such a manner that the angle between them increases. This effect has a sin 2θ dependence which also characterizes the second isotropic effect namely the conversion of the fields from linear to elliptical polarization states. We have shown however that for small frequency separations this second effect can be ignored. The conclusion can be drawn from these results that perpendicular polarization of the fields is in some manner stable while any other relative orientation is unstable. For perpendicular polarization of the signals the coupling between them is also weak which fact further suggests high stability. Parallel polarization of the fields is less stable in the sense that although the medium is isotropic and no rotation or change of polarization exists, the coupling is the strongest and persists for nearly the whole interaction width ($\Delta\nu < 2\gamma_{ab}$), resulting in eventual dominance of one of the signals.

6.3.4 Nonzero Magnetic Field

The case of central tuning and small frequency separation of the signals will now be considered for $\omega_{+-} \neq 0$ ($y \neq 0$). Because $x_o = 0$ and η is small the fields remain approximately linearly polarized. Equations 6.21-6.26 still apply but with the above conditions on the variables. Φ_1 and Φ_2 now change rapidly since thereis a Faraday rotation experienced by each field. Let us first examine the solutions for $y \gg a$, i.e., $\omega_{+-} \gg \gamma_{ab}$.

6.3.4A Well-Separated Magnetic Sublevels

If $\omega_{+-} \gg \gamma_{ab}$ all the saturation functions with the exception of H_1, H_4 and H_5 are negligibly small. Then if $x_o = 0$ and $\eta \ll y$

$$\frac{\nu}{2\epsilon_o c} P_{1p} = i\alpha E_1 \left\{ \frac{1}{2}\left[w^*(y-\eta+ia)+w^*(-y-\eta+ia) \right] \right.$$

$$- \frac{E_1^2}{4E_o^2}\, \frac{1}{2}\left[H_1(0,y,\eta)+H_1(0,-y,\eta) \right]$$

$$\left. - \frac{E_2^2}{4E_o^2}\, \frac{1}{2}\left[H_4(0,y,\eta)+H_4(0,-y,\eta)+H_5(0,y,\eta)+H_5(0,-y,\eta) \right] \right\} \qquad (6.44)$$

and a similar equation for $P_{2p}(\eta \rightarrow -\eta)$.

For very strong Doppler broadening and $\omega_{+-} \gg \Delta\nu$ these become

$$\frac{dE_1}{dz} = \alpha\, E_1 \left\{ 1 - \frac{E_1^2}{4E_o^2} - \frac{E_2^2}{4E_o^2}\left[\frac{(\gamma_a+\gamma_b)^2}{(\gamma_a+\gamma_b)^2+(\Delta\nu)^2} \right.\right.$$

$$\left.\left. + \frac{\gamma_a\gamma_b}{2\gamma_{ab}}\left(\frac{\gamma_a}{\gamma_a^2+(\Delta\nu)^2} + \frac{\gamma_b}{\gamma_b^2+(\Delta\nu)^2} \right) \right] \right\} e^{-(\omega_{+-}/2ku)^2} \qquad (6.45a)$$

$$\frac{dE_2}{dz} = \alpha\, E_2 \left[1 - \frac{E_2^2}{4E_o^2} - \frac{E_1^2}{4E_o^2}\left[\frac{(\gamma_a+\gamma_b)^2}{(\gamma_a+\gamma_b)^2+(\Delta\nu)^2} \right.\right.$$

$$\left.\left. + \frac{\gamma_a\gamma_b}{2\gamma_{ab}}\left(\frac{\gamma_a}{\gamma_a+(\Delta\nu)^2} + \frac{\gamma_b}{\gamma_b^2+(\Delta\nu)^2} \right) \right] \right\} e^{-(\omega_{+-}/2ku)^2} \quad . \qquad (6.45b)$$

For small $\Delta\nu$ the coupling is uniformly strong, regardless of the magnetic field splitting, but the saturation effects are only half as strong as for the no magnetic field case. The interaction of the two signals is also independent of the relative orientations of the polarization vectors. The function in the square brackets which determines how the coupling falls off with increasing $\Delta\nu$ is similar to the one for zero magnetic field (for $\theta = 0$), the transition to weak coupling occuring at $\Delta\nu$ roughly equal to the geometric mean of the decay rates. The equations for the Faraday rotation are equally straightforward

$$\frac{d\Phi_1}{dz} = \alpha \, \mathrm{Im}\left\{\frac{1}{2}\left[w^*(+y-\eta+ia) - w^*(-y-\eta+ia)\right] - \frac{E_1^2}{4E_o^2}\,\frac{1}{2}\left[H_1(0,y,\eta)-H_1(0,-y,\eta)\right]\right.$$

$$\left. - \frac{E_2^2}{4E_o^2}\,\frac{1}{2}\left[H_4(0,y,\eta)-H_4(0,-y,\eta)+H_5(0,y,\eta)-H_5(0,-y,\eta)\right]\right\} \quad , \qquad (6.46)$$

and a similar equation for $d\Phi_2/dz$ $(\eta \rightarrow -\eta)$. We already know from the results of chapter five that self saturation has an effect on the Faraday rotation only for homogeneous or nearly homogeneous broadening. The imaginary part of H_1 is neglible for $a \rightarrow 0$ (inhomogeneous broadening). In terms of hole burning this is equivalent to the statement that "a hole does not have a first order effect on itself" (12) . The presence of the second field, however, does result in saturation of the Faraday rotation. This is seen more readily by writing 6.46 for a strongly inhomogeneous line and $y \gg \eta$ as

$$\frac{d\Phi_1}{dz} = \alpha \left\{\mathrm{Im}\, w^*(y+ia) - \frac{E_2^2}{4E_o^2}\left[\frac{\eta a}{a^2+\eta^2} + \frac{AB}{a}\left(\frac{\eta}{A^2+\eta^2} + \frac{\eta}{B^2+\eta^2}\right)\right]e^{-y^2}\right\} \quad . \qquad (6.47)$$

The saturation function (in square brackets) is odd in η , that is the effect of the interaction is to increase or decrease the angle between the two fields leaving the mean rotation unchanged from the unsaturated value. This can be seen from writing

$$d\left[\frac{1}{2}(\Phi_1+\Phi_2)\right]/dz = \alpha \ \mathrm{Im}\ w^*(y+ia) \tag{6.48}$$

$$d\left[\frac{1}{2}(\Phi_2-\Phi_1)\right]/dz = \alpha \frac{E_2^2}{4E_o^2}\left[\frac{A\eta}{A^2+\eta^2} + \frac{AB}{a}\left(\frac{\eta}{A^2+\eta^2} + \frac{\eta}{B^2+\eta^2}\right)\right] e^{-y^2} \ . \tag{6.49}$$

6.3.4B Small Magnetic Fields

This case is considerably more difficult to handle since all the saturation functions are important. Notably H_8 and H_9 are significant, with the result that the nonlinear effects are dependent on θ , the angle between the polarization vectors. We shall examine the dependence of the coupling on the magnetic field splitting, the frequency separation and the angle θ . The Faraday rotation will not be considered, the equations being too complex to warrant quantitative analysis here.

$$\frac{\nu}{2\epsilon_o c}P_{1p} = i\alpha E_1 \left\{\frac{1}{2}\left[w^*(y-\eta+ia)+w^*(-y-\eta+ia)\right]\right.$$

$$\left. - \frac{E_1^2}{4E_o^2}\sum_{i=1}^{3} R_i(y,\eta) - \frac{E_2^2}{4E_o^2}\sum_{i=4}^{9} R_i(y,\eta)\right\} \tag{6.50}$$

where

$$R_i(y,\eta) = \frac{1}{2}\left[H_i(0,y,\eta)+H_i(0,-y,\eta)\right] \quad . \tag{6.51}$$

In order to keep the results tractable we again specialize to very strong Doppler broadening. This means that the first portion of all the H functions is negligible and since both y and η are small the value of the second square bracketed quantity equals 2 in all of them. With this simplification 6.46 becomes

$$\frac{\nu}{2\epsilon_o c}P_{1p} = i\alpha E_1\left\{1-\frac{E_1^2}{4E_o^2}\left[1+\frac{A^2}{A^2+y^2}\right]-\frac{E_2^2}{4E_o^2}\left[\frac{AB}{a-i\eta}\left(\frac{1}{A-i\eta}+\frac{1}{B-i\eta}\right)+\frac{a}{a-i\eta}\right.\right.$$

$$+\frac{1}{2}\left(\frac{A}{A+i(y-\eta)}+\frac{A}{A-i(y+\eta)}\right)+\frac{1}{2}\left(\frac{AB}{a+i(y-\eta)}\left(\frac{1}{A+iy}+\frac{1}{B-i\eta}\right)e^{i2\theta}\right.$$

$$\left.\left.\left.+\frac{AB}{a-i(y+\eta)}\left(\frac{1}{A-iy}+\frac{1}{B+i\eta}\right)e^{i2\theta}\right)\right]\right\} \tag{6.52}$$

And a similar equation for $P_{2p}(\eta \rightarrow -\eta)$.

Equation 6.48 can be compared with the corresponding one for zero magnetic field, 6.33. We see that the nonlinear functions are considerably more complicated, both the self saturation and the interaction being now functions of y . The coupling strength is determined by the relative sizes of the real parts of the quantities multiplying $E_1^2/2E_o^2$ and $E_2^2/2E_o^2$. Figure 19 shows these vs. y for various values of $\Delta\nu$ and θ . The region of strong coupling is where the self saturation function (β_1) is smaller than the interaction function (θ_{12}) . The interesting feature of these curves is the existence of strong coupling regions around the resonance points $\omega_{+-} = \Delta\nu$. For very small frequency separation the coupling is always strong, regardless of the angle θ and of the magnetic field, except at y = 0 for $\theta = \pi/2$ where weak coupling exists as discussed in section 6.3.3. As the frequency separation is increased the interaction decreases in the central region (around $\omega_{+-} = 0$) but for both $\theta = 0$ and $\pi/2$ symmetric strong coupling regions about $\omega_{+-} = \pm \Delta\nu$ can be observed. These resonances can be attributed at least in part to the coherent double quantum interactions which play an important role in all nonlinear processes. For $\Delta\nu > \gamma_{ab}$ the coupling is weak everywhere as expected. The curves for $\theta = \pi/4$ are asymmetric about y = 0 but otherwise have qualitative behavior similar to the other two cases. For $\omega_{+-} > \gamma_{ab}$ these solutions agree with the strong magnetic field case discussed in 6.3.4A, that is the interaction is determined by the frequency separation only and is independent of either the magnetic field or the angle θ .

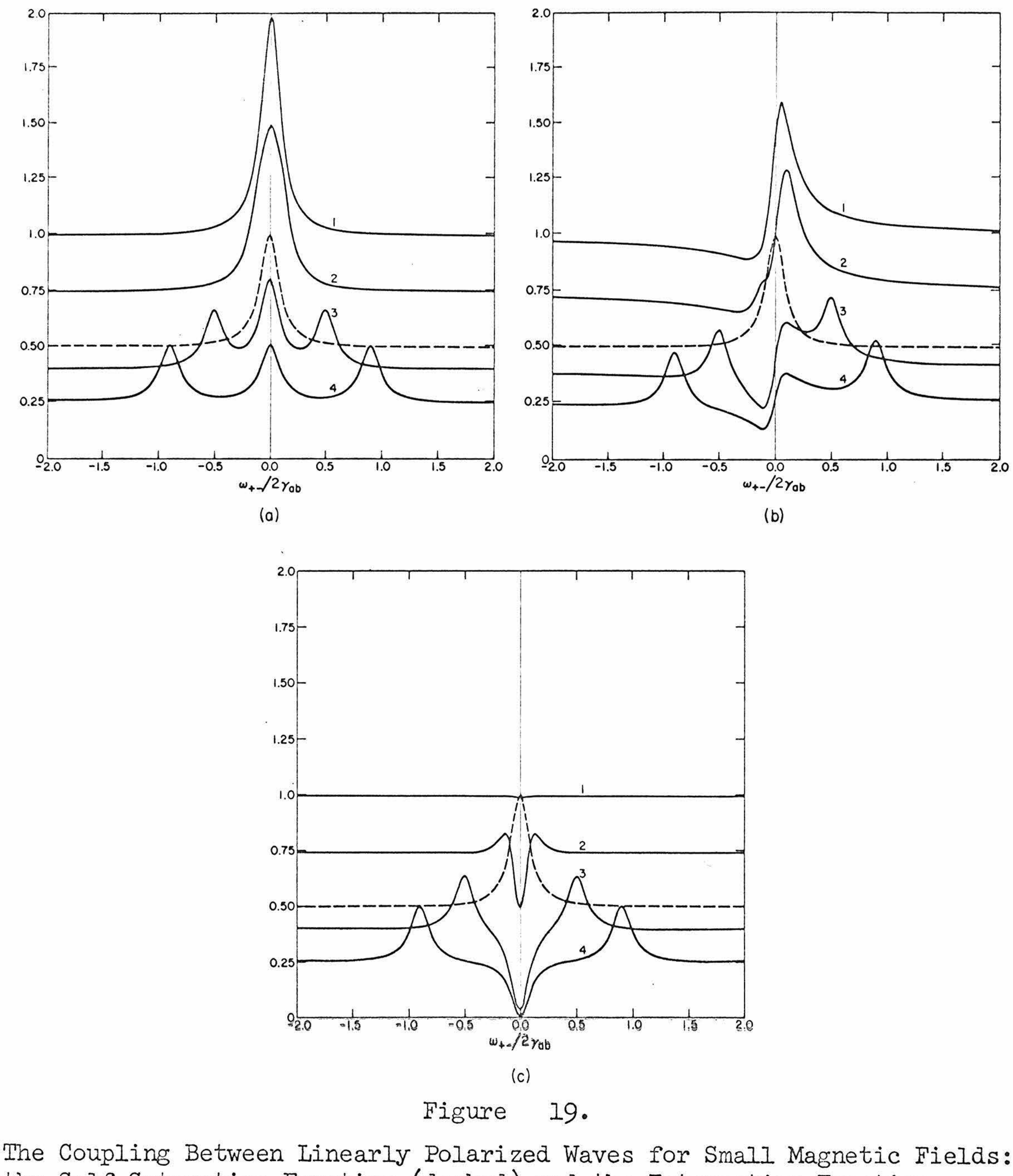

Figure 19.

The Coupling Between Linearly Polarized Waves for Small Magnetic Fields: the Self Saturation Function (dashed) and the Interaction Function (solid); $\gamma_a/2\gamma_{ab} = 0.1$, $\gamma_b/2\gamma_{ab} = 0.9$; (a) $\theta = 0^\circ$ (b) $\theta = 45^\circ$ (c) $\theta = 90^\circ$; 1. $\Delta\nu/2\gamma_{ab} = 0.0$ 2. $\Delta\nu/2\gamma_{ab} = 0.1$ 3. $\Delta\nu/2\gamma_{ab} = 0.5$ 4. $\Delta\nu/2\gamma_{ab} = 0.9$.

6.4 Combination Tone Generation

The generation of new frequencies in the laser and its dependence on the magnetic field and on the angle between the polarization vectors of the input fields are discussed in this section. Some of the results have been reported earlier (48). The following second order terms contribute to the third order polarization at the new frequency $2\nu_1-\nu_2$

$$N_{+b}^{(2)} = -N_o W(v) \left\{ \frac{E_{1-}E_{2-}}{E_o^2} \frac{\gamma_a\gamma_b}{4} \left(\frac{1}{\gamma_a - i(\nu_1-\nu_2)} + \frac{1}{\gamma_b - i(\nu_1-\nu_2)} \right) \right.$$

$$\times \left(\frac{1}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)} + \frac{1}{\gamma_{ab}-i(\omega_{+b}-\nu_2+kv)} \right) e^{i(\varphi_{1-}-\varphi_{2-})}$$

$$+ \frac{E_{1+}E_{2+}}{E_o^2} \frac{\gamma_a\gamma_b}{4} \frac{1}{\gamma_b - i(\nu_1-\nu_2)} \left(\frac{1}{\gamma_{ab}+i(\omega_{-b}-\nu_1+kv)} + \frac{1}{\gamma_{ab}-i(\omega_{-b}-\nu_2+kv)} \right)$$

$$\times e^{i(\varphi_{1+}-\varphi_{2+})} e^{i[(k_1-k_2)z-(\nu_1-\nu_2)]t} \qquad (6.53)$$

$N_{-b}^{(2)}$ = same but with subscripts + and - interchanged.

$$\rho_{+-}^{(2)} = -N_o W(v) \frac{E_{1-}E_{2+}}{E_o^2} \frac{\gamma_a\gamma_b}{4} \frac{1}{\gamma_a + i[\omega_{+-}+(\nu_2-\nu_1)]}$$

$$\times\left[\frac{1}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}+\frac{1}{\gamma_{ab}-i(\omega_{-b}-\nu_2+kv)}\right] e^{i(\varphi_{1-}-\varphi_{2+})}$$

$$\times e^{i[(k_1-k_2)z-(\nu_1-\nu_2)t]} \quad . \tag{6.54}$$

Other terms in $N_{\pm b}$ and ρ_{+-} contribute to the polarization at the other sideband frequency $2\nu_2-\nu_1$. This term will be written down by analogy. Combining 6.53, 6.54 we obtain

$$\frac{\nu}{2\epsilon_o c}P_{o-}^{(3)}(v,z,t) = i\alpha_o W(v)\left\{\frac{E_{1-}^2 E_{2-}}{E_o^2}\,\frac{\gamma_a\gamma_b}{4}\left(\frac{1}{\gamma_a-i(\nu_1-\nu_2)}+\frac{1}{\gamma_b-i(\nu_1-\nu_2)}\right)\right.$$

$$\times\left(\frac{1}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\,\frac{1}{\gamma_{ab}+i(\omega_{+b}-2\nu_1+\nu_2+kv)}\right.$$

$$\left.+\frac{1}{\gamma_{ab}-i(\omega_{+b}-\nu_2+kv)}\,\frac{1}{\gamma_{ab}+i(\omega_{+b}-2\nu_1+\nu_2+kv)}\right)e^{i(2\varphi_{1-}-\varphi_{2-})}$$

$$+\frac{E_{1+}E_{1-}E_{2+}}{E_o^2}\,\frac{\gamma_a\gamma_b}{4}\,\frac{1}{\gamma_b-i(\nu_1-\nu_2)}\left(\frac{1}{\gamma_{ab}+i(\omega_{-b}-\nu_1+kv)}\,\frac{1}{\gamma_{ab}+i(\omega_{+b}-2\nu_1+\nu_2+kv)}\right.$$

$$\left.+\frac{1}{\gamma_{ab}-i(\omega_{-b}-\nu_2+kv)}\,\frac{1}{\gamma_{ab}+i(\omega_{+b}-2\nu_1+\nu_2+kv)}\right)e^{i(\varphi_{1-}+\varphi_{1+}-\varphi_{2+})}$$

$$+\frac{E_{1+}E_{1-}E_{2+}}{E_o^2}\frac{\gamma_a\gamma_b}{4}\frac{1}{\gamma_a+i[\omega_{+-}-(\nu_1-\nu_2)]}\left(\frac{1}{\gamma_{ab}+i(\omega_{+b}-\nu_1+kv)}\frac{1}{\gamma_{ab}+i(\omega_{+b}-2\nu_1+\nu_2+kv)}\right.$$

$$\left.+\frac{1}{\gamma_{ab}-i(\omega_{-b}-\nu_2+kv)}\frac{1}{\gamma_{ab}+i(\omega_{+b}-2\nu_1+\nu_2+kv)}\right)e^{i(\varphi_{1-}+\varphi_{1+}-\varphi_{2+})}\Big\}$$

$$\times e^{i[(2k_1-k_2)z-(2\nu_1-\nu_2)]} \quad . \qquad (6.55)$$

The wave characteristics of P have been included. The subscript o indicates that it is a new frequency not present in the input. The other combination tone frequency $2\nu_2-\nu_1$ will be indicated by the subscript 3. After performing the integration over the velocity distribution (details in Appendix I) the following expression is obtained.

$$\frac{\nu}{2\epsilon_o c}P_{o-}(z,t) = i\alpha\left\{\frac{E_{1-}E_{2-}^2}{E_o^2}U_1(x_o,y,\eta)+\frac{E_{1+}E_{2+}E_{2-}}{E_o^2}U_2(x_o,y,\eta)e^{i[2(\Phi_2-\Phi_1)]}\right\}$$

$$\times e^{i(2\varphi_{1-}-\varphi_{2-})}e^{i[(2k_1-k_2)z-(2\nu_1-\nu_2)t]} \qquad (6.56)$$

where

$$U_1(x_o,y,\eta) = \frac{AB}{2}\left[\frac{1}{A-i\eta}+\frac{1}{B-i\eta}\right]\left[\frac{i}{2\eta}\Big(w^*(x_o+y-\eta+ia)-w^*(x_o+y-3\eta+ia)\Big)\right.$$

$$\left.+\frac{1}{2(a-2i\eta)}\Big(w^*(x_o+y-3\eta+ia)+w(x_o+y+\eta+ia)\Big)\right] \quad . \qquad (6.57)$$

$$U_2(x_o,y,\eta) = \frac{AB}{2}\frac{1}{B-i\eta}\left[\frac{i}{2(y-\eta)}\Big(w^*(x_o+y-3\eta+ia)-w^*(x_o-y-\eta+ia)\Big)\right.$$

$$+\left.\frac{1}{2(a+i(y-2\eta))}\Big(w^*(x_o+y-3\eta+ia)+w(x_o-y+\eta+ia)\Big)\right]$$

$$+\frac{AB}{2}\frac{1}{A+i(y-\eta)}\left[\frac{i}{2\eta}\Big(3^*(x_o+y-\eta+ia)-w^*(x_o+y-3\eta+ia)\Big)\right.$$

$$+\left.\frac{1}{2(a+i(y-2\eta))}\Big(w^*(x_o+y-3\eta+ia)+w(x_o-y+\eta+ia)\Big)\right] \quad . \qquad (6.58)$$

The rules for obtaining P_{o+} , P_{3+} and P_{3-} are

1.) $P_{om} \rightarrow P_{3m}$ Interchange subscripts 1 and 2

Let $\eta \rightarrow -\eta$

2.) $P_{i-} \rightarrow P_{i+}$ Interchange subscripts + and -

Let $y \rightarrow -\eta$

6.4.1 Zero Magnetic Field

As we have done for the saturation effects, first we examine the important limiting case of zero magnetic field. Since $U_1(x_o,0,\eta) = U_2(x_o,0,\eta)$ the polarization at the new frequency can be written

$$\frac{\nu}{2\epsilon_o c}P_{o\pm}(z,t) = i\alpha\frac{E_1}{\sqrt{2}}\frac{E_2^2}{2E_o^2}\left[1+e^{\mp i2\theta}\right]U_1(x_o,0,\eta)$$

$$\times e^{i(2\varphi_{1\pm}-\varphi_{2\pm})}e^{i[(2k_1-k_2)z-(2\nu_1-\nu_2)t]} \quad . \qquad (6.59)$$

In the above $E_{1,2+}$ and $E_{1,2-}$ have been assumed to remain equal. The validity of this assumption has been discussed in section 6.3. It is immediately obvious that for $\theta = \pi/2$ $P_{o\pm} = 0$; that is, no combination tone generation occurs when the fields are perpendicularly polarized. This is in accordance with the discussion of chapter four, and holds only for zero magnetic field.*

The variation of $U_1(x_o, 0, \eta)$ with η is somewhat similar to that of H_5 . This is as it should be since H_5 also describes a modulation effect, except not at a new frequency, but at one of the original frequencies. The difference is in the appearance of an extra frequency shift in U_1 since the "sideband" field is at $\nu_1 + \Delta\nu$. For very strong Doppler broadening, i.e., $\eta \ll ku$ this difference is negligible and the variation of U_1 is due to the $AB\left[(A-i\eta)^{-1}+(B-i\eta)^{-1}\right]$ factor, at $\eta = (AB)^{1/2}$ the magnitude of the induced polarization P_o decreasing to half its maximum value.

To find the polarization characteristics of the combination tone we note that at the input $(z = 0)$, there is no field at the frequency $\nu_o = 2\nu_1 - \nu_2$ and the new field will build up out of phase with the driving term, leading it by 90°. Thus the initial phases $(z = 0)$ of the + and - components of the new field are, from 6.59,

$$\varphi_{o+} = \frac{\pi}{2} + \Phi_2 - 2\Phi_1 - \varphi' + 2\varphi_1 - \varphi_2 \qquad (6.60a)$$

$$\varphi_{o-} = \frac{\pi}{2} - \Phi_2 + 2\Phi_1 + \varphi' + 2\varphi_1 - \varphi_2 \qquad (6.60b)$$

---

* For orthogonal elliptical polarizations $\Phi_2 - \Phi_1 = \pi/2$ and $E_{1\pm} = E_{2\mp}$; from equation 6.56 P_o is seen to vanish for this case as well.

where

$$\varphi' = \tan^{-1} \frac{\sin 2(\Phi_2-\Phi_1)}{1+\cos 2(\Phi_2-\Phi_1)} = \Phi_2-\Phi_1 , \tag{6.61}$$

and therefore, defining $\varphi_{12} = 2\varphi_1-\varphi_2+\pi$

$$\varphi_{o+} = -\frac{\pi}{2} - \Phi_1 + \varphi_{12} , \tag{6.62a}$$

$$\varphi_{o-} = -\frac{\pi}{2} + \Phi_1 + \varphi_{12} , \quad \text{and} \tag{6.62b}$$

$$\frac{\nu}{2\epsilon_o c}\bar{P}_o(0,t) = i\alpha(\hat{e}_x \sin\Phi_1 + \hat{e}_y \cos\Phi_1)E_1\frac{E_2^2}{2E_o^2}\left[2+2\cos 2\theta\right]U_1(x_o,0,\eta) \, e^{-i(2\nu_1-\nu_2)t + i\varphi_{12}} . \tag{6.63}$$

Comparing with 6.21a,b we see that the field at $\nu_o = 2\nu_1 - \nu_2$ is parallel to E_1. Similarly we can show that the field at $\nu_3 = 2\nu_2 - \nu_1$ will be parallel to E_2. Since for $z > 0$ there exists now fields at the new frequencies it is proper that they be included in the total electromagnetic field. Neglecting the nonlinear effects caused by these very weak signals we can write the additional gain equation

$$\frac{dE_o}{dz} = \alpha E_o \operatorname{Re} w^*(x_o+y-3\eta+ia) + \frac{\nu}{2\epsilon_o c}|P| . \tag{6.64}$$

The problem with using equation 6.48 that it is in fact correct only at z = 0, for several reasons. First, as we know from section 6.3 the polarization of the fields E_1 and E_2 does not remain fixed, but they rotate apart due to an induced anisotropy in the medium (In addition there is also the tendency to convert to elliptical polarization. This, however, we have shown to be negligible if the frequency separation is small) To show that the new field E_o is parallel to E_1 we assumed that there being no field E_o at $z = 0$ it builds up out of phase with $P_{o\pm}$. This condition does not continue to hold for $z = 0$ since there is dispersion in the medium. The accumulated nonlinear phase of $P_{o\pm}(z,t)$ is not the same as the accumulated phase of $E_{o\pm}$. The only proper way to handle the equations for the new fields is to keep them in circularly polarized form. The correct gain and phase equations valid for all z therefore are

$$\frac{dE_{o\pm}}{dz} = \alpha E_{o\pm} \operatorname{Re} w^*(x_o+y-3\eta+ia)+|P_o|\cos(\varphi_{120\pm}-\varphi_{120\pm}(0)) , \tag{6.65}$$

$$\frac{d\varphi_{o\pm}}{dz} E_{o\pm} = \alpha E_{o\pm} \operatorname{Im} w^*(x_o+y-3\eta+ia)+|P_o|\sin(\varphi_{120\pm}-\varphi_{120\pm}(0)) , \tag{6.66}$$

where $\varphi_{120\pm} = 2\varphi_{1\pm}- \varphi_{2\pm}- \varphi_{o\pm}$. These are four coupled equations for $dE_{o\pm}/dz$ and $d\varphi_{o\pm}/dz$. In addition, the quantities $\varphi_{1\pm}$ and $\varphi_{2\pm}$ are given by equation 6.36 of section 6.3. Obviously the complete problem is extremely complex and we will not attempt to solve it here. It will suffice to say that the gain of the new field E_o is less than that given by 6.48. The present analysis is also unable to answer the question whether the field E_o remains parallel to E_1 as the latter rotates away from E_2 in its propagation through the medium. For parallel polarization of the input signals there is of course no rotation and the new fields $2\nu_1 - \nu_2$ and at $2\nu_2 - \nu_1$ are also polarized in the same direction as E_1 , E_2 . The building up of a phase mismatch between E_o and P_o , however, is still present. The correct equations for the combination tone fields are 6.49 and 6.50 (without the ± subscripts, since + and - components combine to give one phase and one gain equation). For this case our equations agree with those of Close (6) who has considered the effects of the mismatch in some detail. That author has also recently reported observations of combination tones, for parallel polarized inputs, at the 3.39μ He-Ne transition. The effects of nonparallel polarizations discussed here should be observable using similar techniques.

6.4.2 Nonzero Magnetic Field

For nonzero magnetic fields U_1 and U_2 are not equal, and $P_{o,3\pm}$ are given by equations 6.40-6.42 and the rules following these. The actual solutions for the combination tone fields are even more complicated than those for nonzero magnetic fields. Rather than attempt to find these we shall be content with examining the characteristics of $\frac{\nu}{2\epsilon_o c}|P_o|$ which gives an idea of how the combination tone generation varies with $\Delta\nu$ and ω_{+-} . From 6.59,

$$\frac{\nu}{2\epsilon_o c}|P_{o\pm}| \simeq \alpha E_1 \frac{E_2^2}{E_o^2}|U_1(x_o,\mp y,\eta) + U_2(x_o,\mp y,\eta)e^{\mp i2\theta}| \quad . \tag{6.67}$$

The unbalance that builds up between the + and - components of the input fields have also been neglected in the above. The characteristics of U_1 are essentially the same as for the zero magnetic field case: the maximum occurs for $\eta \rightarrow 0$ (closely spaced input frequencies), U_1 has decreased to half its value for $\eta = (AB)^{1/2}$ and continues to decrease as the frequency separation is increased. This term is due physically to the modulation of the N_{+b} population inversion density by E_{1-} and E_{2-} and the decrease with increasing frequency separation is consistent with the description of the process given in chapter four. U_{2-} contains two parts, the first resulting from the modulation of the common lower level's population by E_{1+} and E_{2+} and the second from a coherent double quantum process involving E_{1-} and E_{2+}. The notable feature of U_2 is the presence of a resonance at $y = \eta$ due to the above mentioned coherent Raman type process, shown by the factor $[A+i(y-\eta)]^{-1}$. The magnitude of the resonance, however, is limited by the factor $a+i(y-2\eta)$ which causes the peak to become smaller as η increases. The complete behavior of $|P_\pm|$ is shown on Figure 20 for the two extreme cases, parallel and perpendicular polarization of the input signals. $C_\pm(2\nu_1-\nu_2)$ is the quantity between the absolute value signs in equation 6.67. The same curves hold for $C_\pm(2\nu_2-\nu_1)$ but with + and - components interchanged. For a fixed frequency separation of the input signals the left and right circular components of P_o are seen to increase as the magnetic fields are increased from zero. The two components are not the

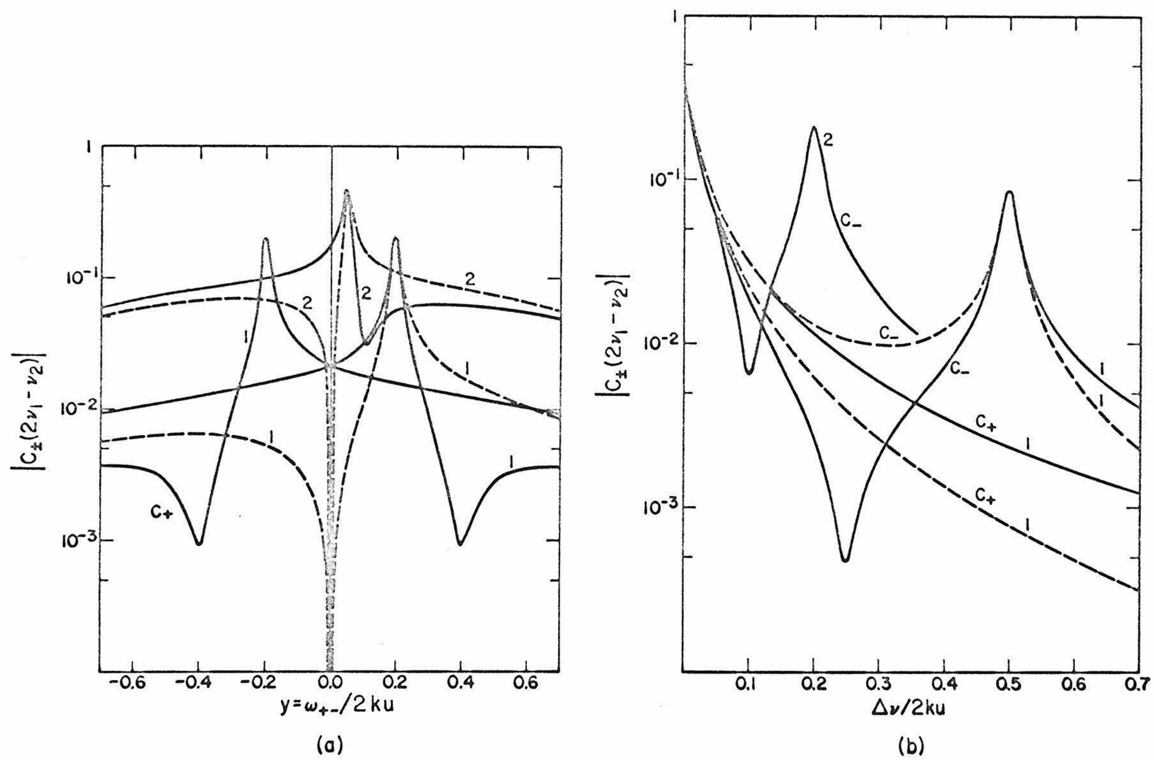

Figure 20.

The Polarization at $2\nu_1-\nu_2$ Due to Fields at ν_1 and ν_2 : a = 0.1, A = 0.01, B = 0.09, x_o = 0; Dashed Line: Perpendicular Polarizations, Solid Line: Parallel Polarizations; (a) vs. Magnetic Field Splitting 1. $\Delta\nu/2ku = 0.2$ 2. $\Delta\nu/2ku = 0.05$ (b) vs. Frequency Separation 1. $\omega_{+-}/2ku = 0.5$ 2. $\omega_{+-}/2ku = 0.2$.

same size, however, except at $y = 0$. P_{o-} peaks at $y = \eta$ while P_{o+} at $y = -\eta$, and at these points the other components are smaller, indicating essentially circularly polarized combination tones. For perpendicularly polarized inputs there is of course no combination tone generation for $y = 0$ and the increase in $P_{o\pm}$ as y is changed from zero is more sudden. These peaks in the sideband generating interaction at $\omega_{+-} = \pm \Delta\nu$ correspond to the peaks in the saturation interaction at the same points which can be observed on Figure 19. Both are caused by Raman type, double quantum interactions. If the frequency separation is varied, for a given magnetic field separation of the transitions for small $\Delta\nu$ the behavior is essentially the same as for the zero magnetic field case, $P_{o\pm}$ decreasing rapidly with increasing $\Delta\nu$. Provided y is not too large, however, at the resonance point $\Delta\nu = \omega_{+-}$ P_{o-} has a resonance large enough that combination tone generation again becomes significant. P_{+}, on the other hand continues to decrease. For E_1 and E_2 parallel polarized, in addition there is a dip in the interaction at $\Delta\nu = \omega_{+-}/2$. Asymmetrical location of ν_1 and ν_2 about line center $(x_o \neq 0)$ tends to wipe out this dip while leaving the resonance peak essentially unchanged. All the curves on Figure 20 are for $\gamma_b = 9\gamma_a$; for $\gamma_a \gg \gamma_b$ the coherent double quantum interactions that cause the resonance at $\Delta\nu = \omega_{+-}$ are smaller, with the result that the peaks of $P_{o\pm}$ are broader and less significant.

6.5 Higher Order Effects

In chapter five some results that are valid for arbitrarily strong fields were calculated for two opposite circularly polarized waves. Similar calculations for linearly polarized fields are exceedingly complex and will not be attempted here. On the bases of previously derived results some conclusions can still be drawn, however, regarding the nature of the strong field solutions. The following two general remarks apply to all of them: 1.) the nonlinear effects are weaker than predicted by an application of the iterative results, 2.) the frequency, magnetic field, and polarization dependence is similar to that of the iterative solutions, as given by equations 6.27-6.43 (zero magnetic field), 6.45-6.49 (large magnetic fields) and 6.48 (small magnetic fields) for saturation effects and by equations 6.56-6.58 for combination tone generation. The only difference is that the decay rates γ_a, γ_b and the natural linewidth γ_{ab} are broadened by the strong fields as we have seen in chapter five.

For the limiting case of zero magnetic field the strong field solutions can be written down without derivation, for some special cases. When the two input fields are perpendicularly polarized the iterative result obtained in section 6.3.3 (equations 6.27-6.29) is identical to the corresponding one for opposite circularly polarized waves, derived in chapter five. The physical reasons for this have already been stated. It is logical to expect that the strong field results are also identical, so long as the magnetic field is zero. The equations for opposite circularly polarized fields of arbitrary intensity were calculated with the condition $\omega_{+-} = \Delta\nu$. For zero magnetic field this corresponds to zero frequency separation. The same results hold, however, in the limit of small frequency difference. Consequently it follows that for $\nu_o = \omega_o$, $\Delta\nu \ll \gamma_a , \gamma_b$

$$\frac{dE_{1,2}}{dz} \simeq \frac{\alpha_o E_{1,2}}{1+E_1^2/E_o^2+E_2^2/E_o^2} \tag{6.68}$$

for a naturally broadened line (stationary atoms), and

$$\frac{dE_{1,2}}{dz} \simeq \frac{\alpha E_{1,2}}{(1+E_1^2/E_o^2+E_2^2/E_o^2)^{1/2}} \operatorname{Re} w^* (0 + ib) , \tag{6.69}$$

for an arbitrary amount of Doppler broadening. In the second equation $b = a(1+E_1^2/E_o^2+E_2^2/E_o^2)^{1/2}$. Also, by analogy we can write down the solutions for $\nu_o \neq \omega_o$.

$$\frac{dE_{1,2}}{dz} = \frac{\alpha_o E_{1,2}}{1+E_1^2/E_o^2+E_2^2/E_o^2+(\omega_o-\nu_o)^2/\gamma_{ab}^{\;2}} , \tag{6.70}$$

$$\frac{dE_{1,2}}{dz} = \frac{\alpha E_{1,2}}{(1+E_1^2/E_o^2+E_2^2/E_o^2)^{1/2}} \operatorname{Re} w^*(x_o+ib) . \tag{6.71}$$

For parallel polarized input fields (and zero magnetic fields) our iterative solutions agree with those of Close (6,7) who used a model of two nondegenerate levels. That author has calculated in some detail results valid for strong fields including generation of higher order sidebands. Unfortunately his method is not suitable for our more complicated level system, but for the case discussed his results should apply. For the derivation the reader is referred to Close's paper (6,7) . The main results will be stated here without derivation, converted to our formalism. For $\Delta\nu \ll \gamma_a , \gamma_b$ (and of course $\omega_{+-} = 0$)

$$\frac{\nu P(z,t)}{2\epsilon_o c} = \frac{\alpha_o \gamma_{ab}}{\gamma_{ab}^2\left(1+\frac{E_1^2+E_2^2+2E_1E_2\cos\Delta_{12}}{E_o^2}\right)+(\omega_o-\nu_o)^2}$$

$$\times\left\{\left[\gamma_{ab}-i(\omega_o-\nu_1)\right]E_1 e^{i(k_1 z-\nu_1 t+\varphi_1)}+\left[\gamma_{ab}-i(\omega_o-\nu_2)\right]E_2 e^{i(k_2-\nu_2 t+\varphi_2)}\right\} , \tag{6.72}$$

for natural broadening, and

$$\frac{\nu P(z,t)}{2\epsilon_o c} = \frac{\alpha}{\pi}\int_{-\infty}^{+\infty}\frac{e^{-\xi^2}d\xi}{a^2\left(1+\frac{E_1^2+E_2^2+2E_1E_2\cos\Delta_{12}}{E_o^2}\right)+(x_o+\xi)^2}$$

$$\times\left\{\left[a-i(x_o-\eta)\right]E_1 e^{i(k_1 z-\nu_1 t+\varphi_1)}+\left[a-i(x_o+\eta)\right]E_2 e^{i(k_2 z-\nu_2 t+\varphi_2)}\right\} , \tag{6.73}$$

for the Doppler broadened case. In these equations $\Delta_{12} = (k_1 - k_2) z - (\nu_1 - \nu_2)t + \varphi_1 - \varphi_2$.

These are the complete expressions for the polarization and include all the combination tones in addition to the input frequencies ν_1 and ν_2 . Close expands these equations in a Fourier series and thus obtains the various frequency components. Experimental verification of these results has been reported (30). We may assume that (still for zero magnetic field) somewhat similar results hold for other than parallel polarization of the input fields but that the quantity

$$\frac{E_1^2 + E_2^2 + 2E_1E_2 \cos \Delta_{12}}{E_o^2}$$

in the denominator is replaced by a more complicated one that includes an angular dependence. This angular dependence must be such that at $\theta = 90^\circ$ the results for perpendicular polarization, equations 6.68-6.71, are obtained. Thus for an arbitrary angle between the polarization vectors we replace the above quantity in equations 6.72, 6.73 by

$$\frac{E_1^2 + C_1(\theta)\, E_2^2 + C_2(\theta) 2E_1E_2 \cos \Delta_{12}}{E_o^2} , \qquad (6.74)$$

where $C_1(\pi/2) = 1$ and $C_2(\pi/2) = 0$. An extension of Close's experiment on combination tone generation to arbitrary angles θ

should help to decide the nature of the functions $C_1(\theta)$ and $C_2(\theta)$.

Finally, for the case of large magnetic fields $\omega_{+-} \gg \gamma_{ab}$ and small frequency separation $\Delta\nu \ll \gamma_a, \gamma_b$ all coherence and mutual saturation effects between opposite circularly polarized components are negligible. Both saturation and combination tone generation are caused by independent action of the same circularly polarized components (E_{1+}, E_{2+} and E_{1-}, E_{2-}) on their respective transitions. For this case equations 6.72 and 6.73 still apply but in a somewhat modified form. For the Doppler broadened case 6.73 becomes

$$\frac{\nu P(z,t)}{2\epsilon_o c} = \frac{\alpha}{\pi} \int_{-\infty}^{+\infty} \frac{e^{-\xi^2} d\xi}{a^2\left(1 + \dfrac{E_1^2+E_2^2+2E_1E_2\cos\Delta_{12}}{2E_o^2}\right) + (y+\xi)^2}$$

$$\times\left\{\left[a-i(y-\eta)\right]E_1 e^{i(k_1 z-\nu_1 t+\varphi_1)} + \left[a-i(y+\eta)\right]E_2 e^{i(k_2 z-\nu_2 t+\varphi_2)}\right\} , \tag{6.75}$$

independent of the orientation of the linearly polarized signals. For $\nu_1 - \nu_2 = 0$, i.e. single linearly polarized wave, this reduces to equation 5.57 of chapter five.

CHAPTER SEVEN

THE GENERAL $J_a \rightarrow J_b$ TRANSITION

7.1 Introduction

In the previous chapters we have found the nonlinear characteristics of a laser medium using a $J = 1 \rightarrow J = 0$ atomic model. It has already been indicated that this model is somewhat special since it involves, for axial magnetic fields, only two transitions that have a common lower level. In this chapter we extend some of the previous results to a general $J_a \rightarrow J_b$ transition. Only the iterative solutions will be examined in detail.

For axial magnetic fields the sublevels with $\Delta M = 0$ are not connected by the perturbation of the fields and any transition can be broken up into two independent sets as shown on Figure 21. Any two adjacent transitions have a common level and all but the most "outside" levels have two opposite circularly polarized transitions originating or terminating on them. The basic simplicity of the level scheme makes the handling of some problems much easier than it would appear at first thought.

7.2 The Iterative Results for Two Opposite Circularly Polarized Signals

The density matrix equation 2.19 is valid for any level system and can be written out in long form for a given transition. If this is done it is seen that a much more complicated set of

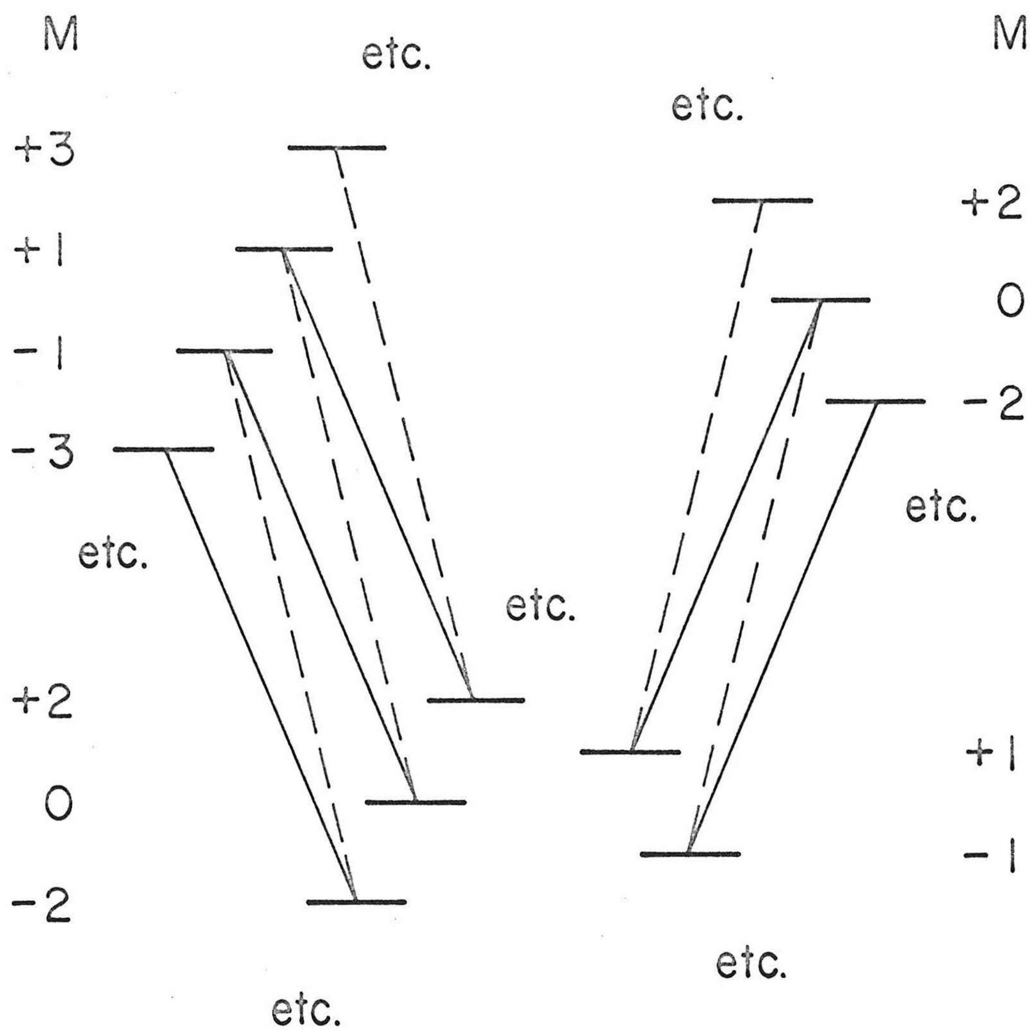

Figure 21.

differential equations than for $J = 1 \rightarrow J = 0$ transition is obtained from which simple integral equations can no longer be derived. Instead of attempting to convert the density matrix equations into integral equations we shall write down the first and third order solutions directly by applying the techniques and results of the previous chapters.

For two opposite circularly polarized input waves (in the same direction) the first and third order induced polarization is

$$P_-^{(1)+(3)}(v) = -i \frac{p_o^2}{\gamma_{ab}\hbar} \frac{1}{2J_b+1} \Big\{ E_- \sum_M \big(\langle J_a, 1, M, -1 | J_b, M-1 \rangle \big)^2 N^{a,b}_{M,M-1}$$

$$+ E_+ \sum_M \langle J_a, 1, M-2, +1 | J_b, M-1 \rangle \langle J_a, 1, M, -1 | J_b, M-1 \rangle \rho^{a,a}_{M,M-2} e^{-i\Delta}$$

$$+ E_+ \sum_M \langle J_a, 1, M, +1 | J_b, M+1 \rangle \langle J_a, 1, M, -1 | J_b, M-1 \rangle$$

$$\times \rho^{b,b}_{M+1,M-1} e^{-i\Delta} \Big\} \frac{\gamma_{ab}}{\gamma_{ab} + i(\omega^{a,b}_{M,M-1} - \nu_- + kv)} , \qquad (7.1)$$

where p_o is the reduced matrix element $\langle a \| p \| b \rangle$, the $\langle J_a, 1, m, -1 | J_b, m' \rangle$'s are the Clebch-Gordan coefficients, and the summation runs over all M values of the upper level a. The $\rho^{a,a}_{m,m'}$ and $\rho^{b,b}_{m,m'}$ are density matrix elements and are self explanatory, while $N^{a,b}_{m,m'}$ are population inversion densities between the m_a and m'_b sublevels. These last quantities are to the zeroeth and second order approximation (the superscript indicating this has been omitted) and are given by

$$N^{a,b}_{M,M-1} = N_o W(v) - N_o W(v) \frac{p_o^2}{\hbar^2 \gamma_a \gamma_b} \frac{1}{2J_b+1}$$

$$\times \Big\{ \Big(\langle J_a, 1, M, -1 | J_b, M-1 \rangle \Big)^2 E_-^2 \frac{1}{2} \Big[\frac{\gamma_{ab}}{\gamma_{ab} + i\big(\omega^{a,b}_{M,M-1} - \nu_- + kv\big)} + \text{c.c.} \Big]$$

$$+ \frac{\gamma_b}{2\gamma_{ab}} \Big(\langle J_a, 1, M, +1 | J_b, M+1 \rangle \Big)^2 E_+^2 \frac{1}{2} \Big[\frac{\gamma_{ab}}{\gamma_{ab} + i\big(\omega^{a,b}_{M,M+1} - \nu_+ + kv\big)} + \text{c.c.} \Big]$$

$$+ \frac{\gamma_b}{2\gamma_{ab}} \Big(\langle J_a, 1, M-2, +1 | J_b, M-1 \rangle \Big)^2 E_+^2 \frac{1}{2} \Big[\frac{\gamma_{ab}}{\gamma_{ab} + i\big(\omega^{a,b}_{M-2,M-1} - \nu_+ + kv\big)} + \text{c.c.} \Big] \Big\} . \tag{7.2}$$

$$\rho^{a,a}_{M,M-2} = -N_o W(v) \frac{p_o^2}{\hbar^2 \gamma_a \gamma_b} \frac{1}{2J_b+1} E_- E_+ \langle J_a, 1, M, -1 | J_b, M-1 \rangle \langle J_a, 1, M-2, +1 | J_b, M-1 \rangle$$

$$\times \frac{1}{2} \Big[\frac{\gamma_{ab}}{\gamma_{ab} + i\big(\omega^{a,b}_{M,M-1} - \nu_- + kv\big)} + \frac{\gamma_{ab}}{\gamma_{ab} - i\big(\omega^{a,b}_{M,M-1} - \nu_+ + kv\big)} \Big] \frac{\gamma_a}{\gamma_a + i\big(\omega^{a,a}_{M,M-2} - (\nu_- - \nu_+)\big)} e^{i\Delta} \tag{7.3}$$

$$\rho^{b,b}_{M+1,M-1} = -N_o W(v) \frac{p_o^2}{\hbar^2 \gamma_a \gamma_b} \frac{1}{2J_b+1}$$

$$\times \frac{\gamma_b}{2\gamma_{ab}} E_- E_+ \langle J_a, 1, M, -1 | J_b, M-1 \rangle \langle J_a, 1, M, +1 | J_b, M+1 \rangle$$

$$\times \frac{1}{2}\left[\frac{\gamma_{ab}}{\gamma_{ab} + i\left(\omega^{a,b}_{M,M-1} - \nu_- + kv\right)} + \frac{\gamma_{ab}}{\gamma_{ab} - i\left(\omega^{a,b}_{M,M+1} - \nu_+ + kv\right)}\right] \frac{\gamma_b}{\gamma_b + i\left(\omega^{b,b}_{M+1,M-1} - (\nu_- - \nu_+)\right)} e^{i\Delta}. \tag{7.4}$$

Equal excitation of the magnetic sublevels has been assumed and k_+ and k_- has been set equal in the denominators.

After combining equations 7.1-7.4 the integration over the velocity distributions can be performed for the final result. All the integrals have been previously encountered and the result can be written down by inspection. Since in general the g factors of the levels a,b are different, the magnetic field splitting of the upper and lower levels are not the same. The transition frequency $\omega^{a,b}_{M,M-1}$ can be written as

$$\omega^{a,b}_{M,M-1} = \omega_o + \omega^b_{+-}/2 + M(\omega^a_{+-} - \omega^b_{+-})/2 = \omega_o + \omega^a_{+-}/2 + (M-1)(\omega^a_{+-} - \omega^b_{+-})/2 \ , \tag{7.5a}$$

where $\omega^k_{+-}/2$ is short for $\omega^{k,k}_{+1,-1}/2$ and is the Zeeman shift of the k level. Similarly,

$$\omega^{a,b}_{M,M+1} = \omega_o - \omega^b_{+-}/2 + M(\omega^a_{+-} - \omega^b_{+-})/2 \quad , \quad \text{and} \tag{7.5b}$$

$$\omega^{a,b}_{M-2,M+1} = \omega_o - \omega^b_{+-}/2 + (M-2)(\omega^a_{+-} - \omega^b_{+-})/2 = \omega_o - \omega^a_{+-}/2 + (M-1)(\omega^a_{+-} - \omega^b_{+-})/2 \ . \tag{7.5c}$$

Defining $y^a = \omega^a_{+-}/2ku$, $y^b = \omega^b/2ku$ and $\delta = (\omega^a_{+-} - \omega^b_{+-})/2ku$ and otherwise using the same symbols as in chapter five the induced polariation becomes

$$\frac{\nu}{2\epsilon_o c} P^{(1)+(3)} = i\alpha E_- \left\{ \sum_M \left(\langle J_a, 1, M, -1 | J_b, M-1 \rangle \right)^2 \frac{3}{2J_b+1} w^*(x_o + y^b + M\delta - \zeta + ia) \right.$$

$$- \frac{E_-^2}{2E_o^2} \sum_M \left(\langle J_a, 1, M, -1 | J_b, M-1 \rangle \right)^4 \frac{9}{(2J_b+1)^2} F_{1M}(y^a, y^b, \zeta)$$

$$- \frac{E_+^2}{2E_o^2} \sum_M \left(\langle J_a, 1, M, -1 | J_b, M-1 \rangle \right)^2 \left(\langle J_a, 1, M, +1 | J_b, M+1 \rangle \right)^2$$

$$\times \frac{9}{(2J_b+1)^2} \left[F^b_{2M}(y^a, y^b, \zeta) + F^b_{3M}(y^a, y^b, \zeta) \right]$$

$$- \frac{E_+^2}{2E_o^2} \sum_M \Big(\langle J_a,1,M,-1|J_b,M-1\rangle \Big)^2 \Big(\langle J_a,1,M-2,+1|J_b,M-1\rangle \Big)^2 \frac{9}{(2J_b+1)^2}$$

$$\times \Big[F_{2M}^a(y^a,y^b,\zeta) + F_{3M}^a(y^a,y^b,\zeta) \Big] \Big\} \quad , \tag{7.6}$$

where

$$F_{1M}(y^a,y^b,\zeta) = a\Big[\frac{2}{\sqrt{\pi}} - 2\Big(a+i(x_o+y^b+M\delta-\zeta)\Big)w^*(x_o+y^b+M\delta-\zeta+ia)$$

$$+ \frac{1}{a} \,\mathrm{Re}\, w^*(x_o+y^b+M\delta-\zeta+ia) \quad , \tag{7.7}$$

$$F_{2M}^a(y^a,y^b,\zeta) = A\Big[\frac{i}{2(y^a-\zeta)}\Big(w^*(x_o+y^a+(M-1)\delta-\zeta+ia) - w^*(x_o-y^a+(M-1)\delta+\zeta+ia)\Big)$$

$$+ \frac{1}{2[a+i(y^a-\zeta)]}\Big(w^*(x_o+y^a+(M-1)\delta-\zeta+ia) + w(x_o-y^a+(M-1)\delta+\zeta+ia)\Big)\Big] \quad , \tag{7.8}$$

$$F_{3M}^a(y^a,y^b,\zeta) \frac{AB}{A+i(y^a-\zeta)}\Big(\frac{2}{\sqrt{\pi}} - 2\Big(a+i(x_o+y^a+(M-1)\delta-\zeta)\Big)w^*\Big(x_o+y^a+(M-1)\delta-\zeta+ia\Big)$$

$$+ \frac{1}{2[a+i(y^a-\zeta)]}\Big(w^*(x_o+y^a+(M-1)\delta-\zeta+ia) + w\Big(x_o-y^a+(M-1)\delta+\zeta+ia\Big)\Big)\Big] \, , \tag{7.9}$$

$$F_{2M}^{b}(y^{a},y^{b},\zeta) = B\left[\frac{i}{2(y^{b}-\zeta)}\left(w^{*}(x_{o}+y^{b}+M\delta-\zeta+ia)-w^{*}(x_{o}-y^{b}+M\delta+\zeta+ia)\right)\right.$$

$$\left.+\frac{1}{2[a+i(y^{b}-\zeta)]}\left(w^{*}(x_{o}+y^{b}+M\delta-\zeta+ia)+w(x_{o}-y^{b}+M\delta+\zeta+ia)\right)\right] \quad , \qquad (7.10)$$

$$F_{3M}^{b}(y^{a},y^{b},\zeta) = \frac{AB}{B+i(y^{b}-\zeta)}\left(\frac{2}{\sqrt{\pi}} -2\left(a+i(x_{o}+y^{b}+M\delta-\zeta)\right)w^{*}(x_{o}+y^{b}+M\delta-\zeta+ia)\right.$$

$$\left.+\frac{1}{2[a+i(y^{b}-\zeta)]}\left(w^{*}(x_{o}+y^{b}+M\delta-\zeta+ia)+w(x_{o}-y^{b}+M\delta+\zeta+ia)\right)\right] \quad . \qquad (7.11)$$

It is seen that these functions are very closely related to $F_{1,2,3}$ of chapter five. F_{1M} describes the self saturation of the (-) transition originating on the M sublevel of the upper level a . The interaction functions $F_{2M}^{a} + F_{3M}^{a}$ and $F_{2M}^{b} + F_{3M}^{b}$ each are due to a three-level subsystem of the type encountered in the $J = 1 \rightarrow J = 0$ model. As in chapter five F_{2M}^{k} and F_{3M}^{k} describe the common level mutual saturation and the contribution of the double quantum interaction respectively. The superscript a indicates that the interaction is due to two opposite circularly polarized transitions from the

M and M - 2 sublevels of a to the common M - 1 sublevel of b, while the superscript b designates interactions caused by two opposite circular transitions originating on a common M sublevel of a and terminating on the M - 1 and M + 1 sublevels of b. It should be noted that the frequency behavior of F_{1M} depends only on the difference of the input frequency and the transition frequency originating on M_a, while the characteristics of the interaction functions are determined primarily <u>by the frequency separation of the two sublevels involved</u> ($\omega^{a,a}_{M,M-2}$ and $\omega^{b,b}_{M+1,M-1}$ for the a and b superscripts respectively). This property will be made use of in section 7.4 where we consider the effects on nuclear spin. For the present $\omega^{a,a}_{M,M-2}$ and $\omega^{b,b}_{M+1,M-1}$ are constants equal to twice the Zeeman shifts of the upper and lower levels respectively. The rules for obtaining F^k_{iM} from F_i are stated below since the same rules can be used to obtain other results that will not be written out explicitly.

$F_1 \rightarrow F_{1M}$: Replace x_o by $x_o + M\delta$ and y by y^b

<u>OR</u>, equivalently

x_o by $x_o + (M-1)\delta$ and y by y^a

$F_2 \rightarrow F^a_{2M}$

Replace x_o by $x_o+(M-1)\delta$ and y by y^a

$F_3 \rightarrow F^a_{3M}$

$F_2 \rightarrow F^b_{2M}$

Replace x_o by $x_o+M\delta$ and y by y^b

$F_3 \rightarrow F^b_{3M}$ Interchange A and B

It can be seen that if $y^a = y^b$ and therefore $\delta = 0$ $F_{1M} = F_1$ $F^a_{2M} = F^a_2$ and $F^a_{3M} = F_3$; in addition the linear part becomes $i\alpha E_- w^*(x_o-\zeta+ia)$, also identical to the $J = 1 \rightarrow J = 0$ result.

To obtain the corresponding equations for P_+ : in all the functions (equations 7.7-7.11) replace y^a, y^b, δ, and M by $-y^a, -y^b, -\delta$ and $-M$. For strong Doppler broadening the expressions, like those in chapter five, simplify considerably. The interaction term only becomes

$$\frac{\nu}{2\epsilon_o c} P^{(3)}_{-(E_+^2)} = i\alpha \frac{E_+^2}{2E_o^2} \left\{ \sum_M C^a_M \frac{A}{A-i(y^a-\zeta)} e^{-[x_o+(M-1)\delta]^2} + \sum_M C^b_M \frac{B}{B-i(y^b-\zeta)} e^{-[x_o+M\delta]^2} \right\} . \qquad (7.12)$$

where

$$C^a_M = \left(\langle J_a,1,M-1|J_b,M-1\rangle\langle J_a,1,M-2,+1|J_b,M-1\rangle\right)^2 \frac{9}{(2J_b+1)^2} ,$$

$$C^b_M = \left(\langle J_a,1,M,-1|J_b,M-1\rangle\langle J_a,1,M,+1|J_b,M+1\rangle\right) \frac{9}{(2J_b+1)^2} ,$$

and the self saturation term simplifies to

$$\frac{\nu}{2\epsilon_o c} P^{(3)}_{-(E_-^2)} = i\alpha \frac{E_-^2}{2E_o^2} \sum_M B_M \, e^{-[x_o + y^b + M\delta - \zeta]^2} \,, \qquad (7.13)$$

where $B_M = (\langle J_a, 1, M, -1 | J_b, M-1 \rangle)^4$.

The case of zero and small magnetic fields ω^a_{+-} , $\omega^b_{+-} \ll \gamma_a \gamma_b$ differs considerably from that of intermediate and large magnetic fields and will be examined first.

7.2.1 Zero and Small Magnetic Fields

For no magnetic field on the medium the levels are completely degenerate $y^b = y^a = \delta = 0$ and the strength of the interaction depends on the frequency separation of the signals. The interaction region is determined by the two Lorentians $A[A-i\zeta]^{-1}$ and $B[B-i\zeta]^{-1}$, the effect of one field on the other diminishing as $\Delta\nu$ becomes much larger than γ_a , γ_b . The self saturation, as expected, shows a Doppler variation only with the detuning of the field from the corresponding line center. The coupling strength also depends on the constants B_M , C^a_M , and C^b_M which are products of Clebsch-Gordan coefficients. At the limit of small frequency separation (or zero separation, which is of course the case of a single linearly polarized field) the interaction and self saturation terms become respectively

$$\sum_M (C^a_M + C^b_M) e^{-x_o^2} \quad \text{and} \quad \sum_M B_M \, e^{-x_o^2} \ .$$

On Table 1 are shown the values of $\sum_M C_M^a$, C_M^b, $C_M^a + C_M^b$ and B_M for various transitions. For small or zero frequency separation and no magnetic field the coupling between the circularly polarized waves is strong when $\sum_M (C_M^a + C_M^b) > \sum_M B_M$. It is seen that, unlike in the $J = 1 \rightarrow J = 0$ model, strong coupling between opposite circularly polarized waves can occur. With the exception of the $J = 1 \rightarrow J = 1$ transition, the coupling is strong whenever $\Delta J = 0$ and weak when $\Delta J = 1$. A single linearly polarized signal is therefore unstable in a laser amplifier for which $\Delta J = 0$ for if either of the circular components becomes slightly larger than the other the difference tends to grow. This instability of a single linearly polarized signal has been recently predicted by Heer and Graft (22). The instability is greater if there is a <u>small</u> magnetic field on the medium (such that $\omega_{+-}^a \ll \gamma_a$, $\omega_{+-}^b \ll \gamma_b$) since detuning the signal from line center causes a slight unbalance between the linear gains, hence in the amplitudes of the opposite circular components which then gets magnified by the strong coupling. For zero magnetic field and nonzero frequency separation, since for $\Delta J = 0$ $\sum_M C_M^a = \sum_M C_M^b$, the region of strong coupling is determined by the condition

$$\frac{\gamma_a^2}{\gamma_a^2+(\Delta\nu)^2} + \frac{\gamma_b^2}{\gamma_b^2+(\Delta\nu)^2} > \frac{\sum_M C_M^a}{\sum_M B_M} \quad . \tag{7.14}$$

For $\Delta\nu = (\gamma_a\gamma_b)^{1/2}$ the L.H.S. is equal to 1 and for all the transitions in the table we have passed into the weak coupling region. For

J_a	$\frac{1}{2}$	1	1	$\frac{3}{2}$	$\frac{3}{2}$	2	2	$\frac{5}{2}$	$\frac{5}{2}$	3	3	$\frac{7}{2}$	$\frac{7}{2}$	4	4	J_b
J_b	$\frac{1}{2}$	0	1	$\frac{1}{2}$	$\frac{3}{2}$	1	2	$\frac{3}{2}$	$\frac{5}{2}$	2	3	$\frac{5}{2}$	$\frac{7}{2}$	3	4	J_a
$\sum_M C^a_M$	0	1	$\frac{1}{4}$	$\frac{3}{8}$	$\frac{12}{50}$	$\frac{21}{100}$	$\frac{21}{100}$	$\frac{28}{200}$	$\frac{32}{175}$	$\frac{18}{175}$	$\frac{9}{56}$	$\frac{18}{224}$	$\frac{12}{84}$	$\frac{11}{168}$	$\frac{77}{600}$	$\sum_M C^b_M$
$\sum_M C^b_M$	0	0	$\frac{1}{4}$	0	$\frac{12}{50}$	$\frac{1}{100}$	$\frac{21}{100}$	$\frac{3}{200}$	$\frac{32}{175}$	$\frac{3}{175}$	$\frac{9}{56}$	$\frac{3}{224}$	$\frac{12}{84}$	$\frac{3}{168}$	$\frac{77}{600}$	$\sum_M C^a_M$
$\sum_M (C^a_M + C^b_M)$	0	1	$\frac{1}{2}$	$\frac{3}{8}$	$\frac{24}{50}$	$\frac{22}{100}$	$\frac{42}{100}$	$\frac{31}{200}$	$\frac{64}{175}$	$\frac{21}{175}$	$\frac{18}{56}$	$\frac{21}{224}$	$\frac{24}{84}$	$\frac{14}{168}$	$\frac{154}{600}$	$\sum_M (C^a_M + C^b_M)$
$\sum_M B_M$	1	1	$\frac{1}{2}$	$\frac{5}{8}$	$\frac{17}{50}$	$\frac{46}{100}$	$\frac{26}{100}$	$\frac{73}{200}$	$\frac{37}{175}$	$\frac{53}{175}$	$\frac{10}{56}$	$\frac{58}{224}$	$\frac{13}{84}$	$\frac{38}{168}$	$\frac{82}{600}$	$\sum_M B_M$
$\lvert\sum_M B_M - (C^a_M + C^b_M)\rvert$	1	0	0	$\frac{2}{8}$	$\frac{7}{50}$	$\frac{24}{100}$	$\frac{16}{100}$	$\frac{42}{200}$	$\frac{27}{175}$	$\frac{32}{175}$	$\frac{8}{56}$	$\frac{37}{224}$	$\frac{9}{84}$	$\frac{24}{168}$	$\frac{72}{600}$	$\lvert\sum_M B_M - (C^a_M + C^b_M)\rvert$

Table 1.

The Clebsch-Gordan coefficient combinations determining interaction strength and combination tone generation a (small) nonzero magnetic field and small $\Delta\nu$ equation 7.12 can be used to compute the regions of strong and weak coupling for any given transition.

7.2.2 Large Magnetic Fields

By large in this case we mean that the Zeeman separation of the sublevels is much larger than the decay rates, i.e., $y^a, y^b \gg A, B$. It is evident from equation 7.12 and 7.13 that if the g factors of the upper and lower levels are equal ($y^a = y^b = y$, $\delta = 0$) the results are the same as for the zero magnetic field case except that the frequency variable on which the interaction depends is $y - \zeta$, same as for the simple $J = 1 \rightarrow J = 0$ transition. The interaction has one resonance at $\omega_{+-} = \Delta\nu$ ($y = \zeta$) at which point the coupling is weak for $\Delta J = 1$ and strong for $\Delta J = 0$; for $\Delta J = 1$ the transition to weak coupling occurs at $y - \zeta = (AB)^{1/2}$. Usually, however, the g factors are not equal and the two resonances $A[A-i(y^a-\zeta)]^{-1}$ and $B[B-i(y^b-\zeta)]^{-1}$ do not coincide. There are two peaks in the interaction, corresponding to the points where the frequency separation of the input signals equals the Zeeman splitting of the upper and lower levels respectively. The net nonlinear gain is minimum at these points and a dip in the output of the amplifier can be observed. Since $\sum_M C_M^a$ and $\sum_M C_M^b$ are both individually smaller than $\sum_M B_M$ the coupling is always weak and both of the opposite circularly polarized signals are amplified in a stable manner. Although the equations are more complicated the basic situation remains the same for weak Doppler or even purely natural broadening.

7.2.3 Single Linearly Polarized Input Field

We want also to examine briefly the special case of a single linearly polarized input field. For the $J = 1 \rightarrow J = 0$ transition this was done in some detail and it is of interest to know the validity of the results for the more general transitions. As it was mentioned above, for $\Delta J = 1$, $J > 1$ an unstable situation exists and it is meaningful to discuss this case separately only for $\Delta J = 0$ which is stable, the polarization of the field remaining linear. It is readily seen from equations 7.6-7.12 that the results are basically similar to that discussed in chapter five. The difference is that the "dip" at zero magnetic field is dependent on <u>both</u> γ_a and γ_b. Equations 5.32-5.35 are still valid but with

$$F(y) = \frac{1}{4} \sum_M \left[F_{1M} + \left(F^a_{2M} + F^a_{3M} \right) + \left(F^b_{2M} + F^b_{3M} \right) \right] \quad , \quad \text{with } \zeta = 0 \ .$$

The "dip" and the anomalous Faraday rotation are due to the interaction functions $F^k_{2M} + F^k_{3M}$. For strong Doppler broadening these become

$$\sum_M \left[C^a_M \frac{\gamma_a}{\gamma_a - i\omega^a_{+-}} + C^b_M \frac{\gamma_b}{\gamma_b - i\omega^b_{+-}} \right] \ .$$

For γ_a and γ_b about equal there is no essential difference from the $J = 1 \rightarrow J = 0$ case. If, however $\gamma_a \ll \gamma_b$ or vice versa the "dip" and the anomalous rotation can take on the shape shown by the sketch of Figure 22.

For large magnetic field, just as in chapter five, all the interaction functions are negligibly small and, since the F_{1M} functions are similar to F_1 , the results are the same as for the J = 1 → J = 0 model; the graph of the incremental Faraday rotation vs. field intensity is essentially identical to that shown on Figure 13.

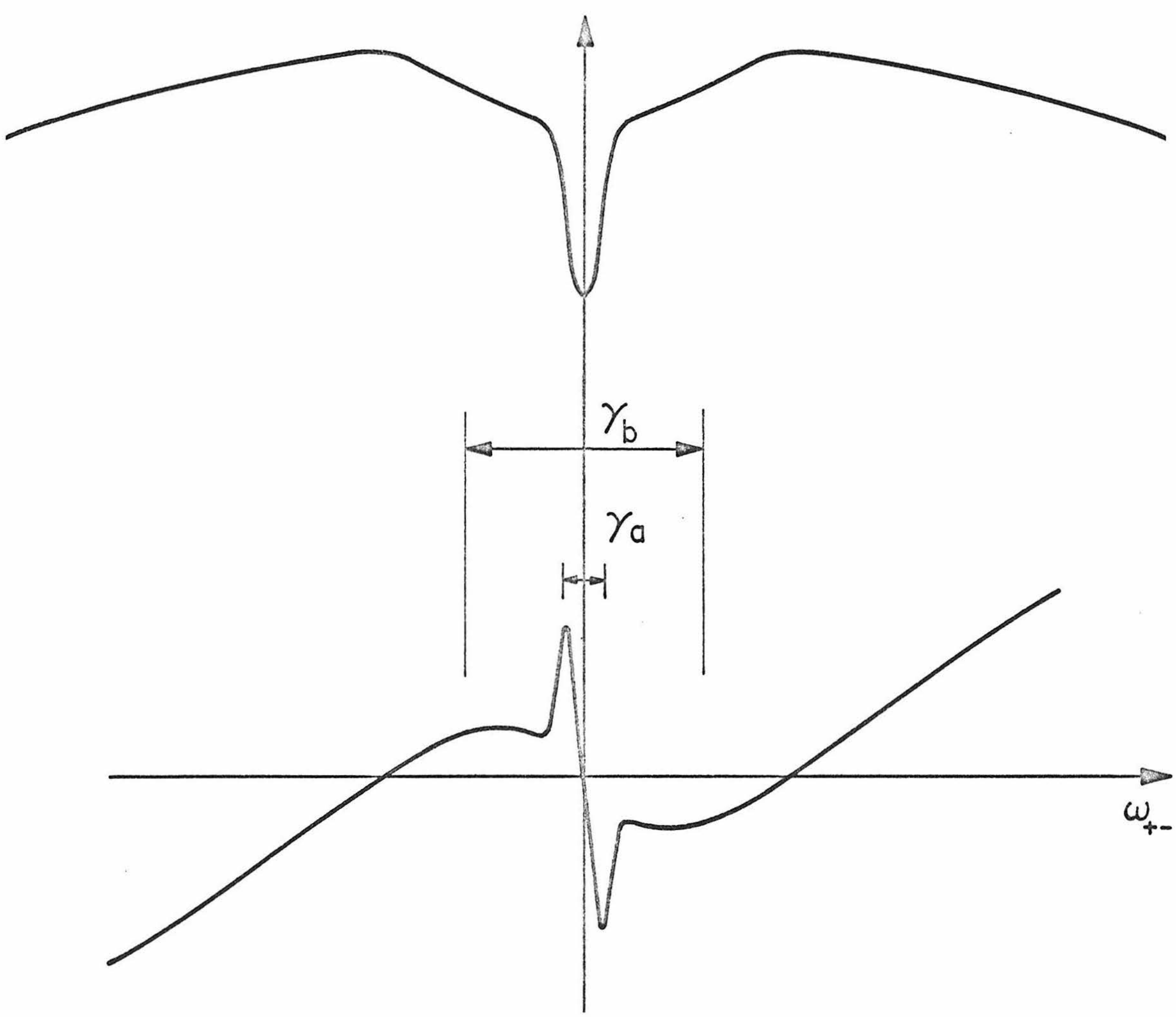

Figure 22.

7.2.4 Extension of Other Results to the $J_a \to J_b$ Transition

In chapter five we have also examined the case of two fields of the same circular polarization (same helicity) that are travelling in opposite directions. It was seen that the common level mutual saturation is still important but the contribution of the multiple quantum interactions is negligible even for weak Doppler broadening, although for completely homogeneous broadening (stationary atoms) the results are identical to the case of two opposite circular fields in the same direction. It is easy to extend these results also for the general $J_a \to J_b$ transition. All that is required is to replace $F_{20}^{a,b}$ and $F_{30}^{a,b}$ by new functions $F_{20M}^{a,b}$ and $F_{30M}^{a,b}$ that are derivable from F_{20} and F_{30} by exactly the same rules as $F_{2M}^{a,b}$, from F_2, F_3 . It is not necessary therefore to give them here.

Results valid for strong fields are considerably more difficult to obtain due to the complexity of the complete density matrix equations. On the basis of previous results we expect that the effects predicted by the iterative results will remain qualitatively the same.

7.3 The Iterative Results for Two Linearly Polarized Signals

The weak field results for two linearly polarized input fields can also be obtained directly. Instead of writing out the zeroeth and second order terms in the population inversion densities and in $\rho^{a,a}_{m,m'}$, $\rho^{b,b}_{m,m'}$ we give the results directly since no new computation is involved, all the Doppler broadening integrals have already been encountered.

7.3.1 Saturation Effects

The circularly polarized component P_{1-} is given by

$$\frac{\nu}{2\epsilon_o c}P_{1-} = i\alpha E_{1-} \left\{\sum_M \left(\langle J_a,1,M,-1|J_b,M-1\rangle\right)^2 w^*(x_o+y^b+M\delta-\zeta+ia)\right.$$

$$- \frac{E_{1-}^2}{2E_o^2}\sum_M B_M H_{1M} - \frac{E_{1+}^2}{2E_o^2}\left[\sum_M \left(H^a_{2M}C^a_M + H^b_{2M}C^b_M\right) + \sum_M \left(H^a_{3M}C^a_M + H^b_{3M}C^b_M\right)\right]$$

$$- \frac{E_{2-}^2}{2E_o^2}\left[\sum_M H_{4M}B_M + \sum_M H_{5M}B_M\right] - \frac{E_{2+}^2}{2E_o^2}\left[\sum_M \left(H^a_{6M}C^a_M + H^b_{6M}C^b_M\right)\right.$$

$$\left.\left. + \sum_M\left(H^a_{7M}C^a_M + H^b_{7M}C^b_M\right)\right]\right\} - i\alpha\,\frac{E_{1+}E_{2-}E_{2+}}{2E_o^2}\left[\sum_M\left(H^a_{8M}C^a_M + H^b_{8M}C^b_M\right)\right.$$

$$\left. + \sum_M\left(H^a_{9M}C^a_M + H^b_{9M}C^b_M\right)\right] \quad , \tag{7.15}$$

where the rules for obtaining H_{1M} , H_{4M} and H_{5M} are the same as that for obtaining F_{1M} , while the rules for all the others are the same as those for obtaining $F_{2M}^{a,b}$, $F_{3M}^{a,b}$. The other polarization components may be found by the rules stated in chapter six.

The induced polarization in linearly polarized form can be obtained in the same manner as was done in chapter six. The case of zero magnetic field and strong Doppler broadening will be examined in detail.

7.3.1A Zero Magnetic Field

If there is no magnetic field on the medium $H_{2M}^{a} + H_{3M}^{a} = H_{2M}^{b} + H_{3M}^{b} = H_{1M}$. If in addition strong Doppler broadening exists each of the functions become very simple

$$H_{1M} \rightarrow e^{-x_o^2}$$

$$H_{4M} + H_{5M} \rightarrow \left(\frac{A}{A-i\eta} + \frac{B}{B-i\eta}\right)e^{-x_o^2}$$

$$H_{6M}^{a} + H_{7M}^{a} \rightarrow \frac{A}{A-i\eta}e^{-x_o^2} \leftarrow H_{8M}^{b} + H_{9M}^{b}$$

$$H_{6M}^{b} + H_{7M}^{b} \rightarrow \frac{B}{B-i\eta}e^{-x_o^2} \leftarrow H_{8M}^{a} + H_{9M}^{a} \quad ,$$

and the third order induced polarization at frequency ν_1 along the direction of the input field E_1 is

$$\frac{\nu}{2\epsilon_o c} P_{1p}^{(3)} = i\alpha E_1 \left\{ -\frac{E_1^2}{4E_o^2}\left[\sum_M \left(B_M + C_M^a + C_M^b\right)\right] + \frac{E_2^2}{4E_o^2}\left[\sum_M B_M \left(\frac{A}{A-i\eta} + \frac{B}{B-i\eta}\right)\right.\right.$$

$$\left.\left. + \sum_M C_M^a \frac{A}{A-i\eta} + \sum_M C_M^b \frac{B}{B-i\eta} + \left(\sum_M C_M^a \frac{B}{B-i\eta} + \sum_M C_M^b \frac{A}{A-i\eta}\right)\cos 2\theta\right]\right\} e^{-x_o^2} . \tag{7.16}$$

The equation for $\frac{\nu}{2\epsilon_o c} P_{2p}$ (at frequency ν_2, parallel to E_2) is the same but with $\eta \to -\eta$. For $J_a = 1$ $J_b = 0$ this reduces to the result of chapter six (equation 6.33).

At the limit of small frequency separation ($\eta \ll A,B$) both $A(A-i\eta)^{-1}$ and $B(B-i\eta)^{-1}$ approach unity and the interaction is determined by the relative magnitude of the quantities

$$\beta_1 = \sum_M \left[B_M + C_M^a + C_M^b\right] e^{-x_o^2} \text{ and } \theta_{12} = \sum_M \left[2B_M + C_M^a + C_M^b + (C_M^a + C_M^b)\cos 2\theta\right] e^{-x_o^2} .$$

For parallel polarization of the input signals, $\theta = 0$, the coupling is always strong $\theta_{12} = 2\beta_1$, same as for the simple level system of chapter six. For perpendicular polarization of the fields, however, the interaction depends on the J values of the levels. Since for $\Delta J = 1$ $\sum_M B_M > \sum_M (C_M^a + C_M^b)$ the coupling is strong for this case, while for $\Delta J = 0$ the coupling is weak. This is the reverse of the situation for opposite circularly polarized fields where the coupling was strong for $\Delta J = 0$ and weak for $\Delta J = 1$.

For arbitrary frequency separation the region of strong coupling can be determined from equation 9.16 for given J values of the levels and angle θ . We note that for $\theta = 0$

$$\theta_{12} = \sum_M (B_M + C_M^a + C_M^b)\left(\frac{A^2}{A^2+\eta^2} + \frac{B^2}{B^2+\eta^2}\right)e^{-x_o^2} ,$$

$$= \sum_M (B_M + C_M^a + C_M^b)\left(\frac{\gamma_a^2}{\gamma_a^2+(\Delta\nu)^2} + \frac{\gamma_b^2}{\gamma_b^2+(\Delta\nu)^2}\right)e^{-(\omega_o-\nu_o)^2/(ku)^2} \qquad (7.17)$$

which has the same frequency dependence as we have found for the $J = 1 \rightarrow J = 0$ transition. For any other angle, however, the frequency dependence is different and equation 9.16 must be used for the more complicated transitions. For $\theta = 90^\circ$ we may note that since for $\Delta J = 0$ $C_M^a = C_M^b$ the coupling always remains weak.

The change in the angle between the polarization vectors is given by

$$\frac{d\theta}{dz} = \alpha\, \frac{E_1^2+E_2^2}{4E_o^2} \mathrm{Re} \sum_M \left[C_M^a (H_{8M}^a + H_{9M}^a) + C_M^b (H_{8M}^b + H_{9M}^b) \right] , \qquad (7.18)$$

which for strong Doppler broadening becomes

$$\frac{d\theta}{dz} = \alpha\, \frac{E_1^2+E_2^2}{4E_o^2} \sum_M \left[C_M^a \frac{B^2}{B^2+\eta^2} + C_M^b \frac{A^2}{A^2+\eta^2} \right] \sin 2\theta$$

$$= \alpha\, \frac{E_1^2+E_2^2}{4E_o^2} \sum_M \left[C_M^a \frac{\gamma_b^2}{\gamma_b^2+(\Delta\nu)^2} + C_M^b \frac{\gamma_a^2}{\gamma_a^2+(\Delta\nu)^2} \right] \sin 2\theta \quad . \qquad (7.19)$$

This is the extension of equation 6.40 to arbitrary transitions. It is often the case in a laser system that $\gamma_a \ll \gamma_b$. For such a case practically achievable minimum $\Delta\nu$ is often much larger than γ_a but much smaller than γ_b. It is seen that in such systems the rotation is largest if $C_M^a \gg C_M^b$ and the most suitable laser transitions for the observation of the effect are those with $J_a = J_b + 1$ rather than those with $J_a = J_b - 1$.

It should be noted at this point that deriving equation 7.16 we have assumed, as in chapter six, that $E_{j+} = E_{j-}$. That this assumption is not really valid because there exists an induced anisotropy in the medium was pointed out in chapter six. For $\Delta J = 0$ there exists additionally a strong coupling between opposite circularly polarized components which causes further instability. Unlike the previous one this latter instability remains even for the cases of parallel and perpendicular polarization of the fields. We can then conclude that no truly stable situation exists in a laser amplifier with J_a or $J_b > 1$ for two linearly polarized signals with close frequency spacing, although highly stable situations exists for a single linearly or two opposite circularly polarized waves.

7.3.1B Nonzero Magnetic Fields

As in chapter six, only small frequency separation is of interest since only then do the fields remain linearly polarized, although the general equation 7.15 is suitable for handling arbitrary frequency separation and arbitrary elliptical polarization of the fields. For large magnetic fields all the functions except H_{1M} H_{4M} and H_{5M} are negligible. Since these all have the same multiplier B_M the results are identical to those of chapter six, that is the coupling is uniformly strong and the Faraday rotation is such that while for strong Doppler broadening the mean of the rotation of the two fields remains constant at its linear value, the effect of saturation is to cause the angle between the two fields to change. Equations 6.44-6.49 apply except the nonlinear part is multiplied by $\sum_M B_M$. For small magnetic fields results similar to those given on Figure 20 are expected, with additional strong coupling regions possible when the Zeeman separation of the upper or lower levels is equal to the frequency difference of the input fields. Equations 6.46, and 6.47 apply but include all the H functions with the proper multipliers.

7.3.2 Combination Tone Generation

For the circularly polarized components of the polarization induced at the new frequency $2\nu_1 - \nu_2$

$$\frac{\nu}{2\epsilon_o c} P_{o-} = i\alpha \left\{ \frac{E_{1-} E_{2-}^2}{E_o^2} \sum_M B_M U_{1M} + \frac{E_{1+} E_{2+} E_{2-}}{E_o^2} \left[\sum_M C_M^a U_{2M}^a + C_M^b U_{2M}^b \right] e^{i2\theta} \right\} , \tag{7.20}$$

The interesting significance of this equation is that since $\sum_M B_M$ in general is not equal to $\sum_M (C_M^a + C_M^b)$ combination tones are produced even for perpendicularly polarized fields. For $\theta = \pi/2$ the multiplying factor equals $\sum_M B_M - \sum_M (C_M^a + C_M^b)$ whose value is also given in the table. This quantity equals zero for the $J = 0 \rightleftarrows J = 1$ transitions. This is in accordance with the discussion of chapter four where it was suggested that only for the above simple cases can the degenerate levels be combined to a new set giving perpendicularly polarized transitions. From equation 7.21 it is seen that for a given transition the combination tone generation is strongest for parallel and weakest for perpendicular polarization of the fields. The frequency dependence, however, is identical. It should also be noted that for $\Delta J = 0$, because of the strong coupling between the opposite circular components of either of the fields, the E_{j+} will not remain equal to E_{j-} .

7.4 The Effects of Nuclear Magnetic Moment

From the results of this chapter it is evident that the effects of nuclear spin on the nonlinear interactions cannot be ignored. Fortunately, it is possible to do this with only minor modifications. The effect of a nonzero nuclear spin on the level structure is to split a given energy level according to the law of addition of angular momenta. In the usual notation the nuclear spin is designated I and the total angular momentum is denoted by F , with F taking on integer values from J - I (or I - J) to J + I. The amount of splitting, which is usually of the order of 10^3Mc, does not concern us here.

In zero magnetic field the equations developed previously for levels with total angular momentum J_a and J_b are still correct for a given hyperfine transition, with F and M_F used in place of J and M_J. If the Doppler width is smaller than the hyperfine splitting, only one transition needs to be considered. If on the other hand the Doppler width is larger than the splitting and the gain curves overlap, the nonlinear polarization must be computed separately for each transition and the contributions added.

In a weak magnetic field the hyperfine Zeeman effect takes place with equal separation of the magnetic sublevels M_F. All the formulae are correct but with ω^a_{+-}, ω^b_{+-} replaced by $\omega_{+-}(F_a)$ and $\omega_{+-}(F_b)$, where

$$\frac{1}{2}\,\omega_{+-}(F_j) = \frac{1}{2}\,\frac{eB}{m}\,g(F_j)M_F\ ,$$

and

$$g(F) = g(J)\left[F(F+1) + J(J+1) - I(I+1)\right]\left[2F(F+1)\right]^{-1}\ .$$

The resonances in the nonlinear polarization occur, as before, where the Zeeman separation of a pair of levels with $\Delta M_F = 2$ equals signal frequency separation.

In magnetic fields that are large enough that the precession of F about the field is faster than that of J and I about F a "hyperfine Paschen-Back effect" takes place. (On account of the weakness of the coupling between J and I this occurs at much lower field strength than the ordinary Paschen-Back effect which we do not consider). Figure 23 shows this schematically for a simple case: J = 1, I = 1/2 .

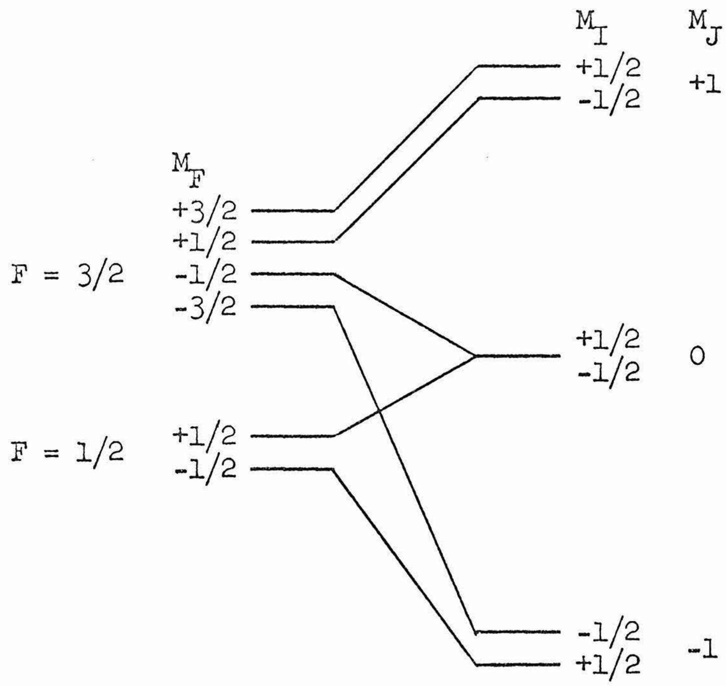

Figure 23.

The ordinary Zeeman effect now occurs, governed by the values of M_J. For each value of M_J, however, there are 2I+1 equidistant hyperfine levels. These are due to the interaction of I and J, and the levels shift is $K\,M_J\,M_I$, where K is the constant determining the field-free hyperfine splitting. The good quantum numbers are J, M_J, M_I and the transitions occur with the selection rule $\Delta M_I = 0$ (and of course $\Delta M_J = \pm 1$). The hyperfine splitting not being the same in the upper and lower levels, each of the Zeeman transitions is split into 2I + 1 components. Although this splitting may be small compared to the Doppler width its effects cannot be ignored since the width of the resonances in the nonlinear polarization is the decay rates of the levels which is usually smaller still. For a given value of M_I the equations of this chapter are modified by replacing y^a and y^b by the corresponding quantities that are now different for each value of M_J. Thus, for example, $y^a \rightarrow y^a(M,M-2;M_I) = \omega^{a,a}_{M,M-2}(M_I)/2ku$, the frequency difference between the M_J and M_J-2 sublevels of a, for a given value of M_I. The "detuning" quantities $x_o+(M-1)\delta$ and $x_o+M\delta$ (i.e., the difference between the mean frequency of the signals and the mean frequency of the two opposite circular transitions that are involved) are likewise changed to $x_o+(M-1)\delta+(M-1)\delta'$ and $x_o+M\delta+M\delta'$, where $\delta' = (K^a - K^b)(M-1)M_I/2ku$, to account for the hyperfine shift of the transitions.

For intermediate magnetic fields the situation is still the same, the resonances in the interaction are determined by the separation of two sublevels with $\Delta M_J = 2$. The only additional difference is that the mean frequency of two opposite circularly polarized transitions varies in a complicated manner with the magnetic field and the "detuning" is consequently also a rather complex function for which simple formulae cannot be given.

7.5 Weak and Strong Coupling in a Laser Amplifier

Let us now digress briefly and discuss the consequences of the phenomena we have called weak and strong coupling in a laser amplifier. We have briefly touched the subject in chapter five. All the quantitative results have now been derived, and we have encountered both weak and strong coupling. Some equations valid for strong fields are also available and it is appropriate to examine the physical consequences in more detail. The nonlinear gain equations for weak fields are always of the form

$$\frac{dE_1}{dz} = \alpha_1 E_1 - \beta_1 E_1^3 - \theta_{12} E_1 E_2^2 \tag{7.22a}$$

$$\frac{dE_2}{dz} = \alpha_2 E_2 - \beta_2 E_2^3 - \theta_{21} E_2 E_1^2 \quad . \tag{7.22b}$$

When $\theta_{12}\theta_{21} > \beta_1\beta_2$ (strong coupling), and if the intensities are unequal the gain of the weaker signal is decreased more than that of the stronger one by the nonlinear effects, with the result that the stronger signal is amplified more. For weak coupling $(\theta_{12}\theta_{21} < \beta_1\beta_2)$ the reverse takes place, the weaker signal having higher gain. Strong coupling thus tends to amplify any unbalance between two signals, while weak coupling has the effect of equalizing them. While the effective gain in equations 7.22a,b appears to go to zero and become negative for high enough fields (regardless of coupling strength) this is not so since these equations become invalid before such intensity is reached. This was already pointed out in chapter five. In the same chapter we have obtained some results for opposite circularly polarized signals that are valid for arbitrarily strong fields and have seen that for critical coupling $(\theta_{21} = \theta_{12} = \beta_1 = \beta_2)$ the valid gain equations for a Doppler broadened transition are

$$\frac{dE_{1,2}}{dz} = \frac{\alpha\, E_{1,2}}{(1+E_1^2/E_o^2+E_2^2/E_o^2)^{1/2}} \quad . \tag{7.23}$$

The gain always remains positive and both fields are amplified equally. While no strong field equations valid for weak coupling were derived we can assume that for that case the gain is of the form

$$\frac{1}{E_1}\frac{dE_1}{dz} = \frac{\alpha_1}{(1+hE_1^2/E_o^2+ g\ E_1^2/E_o^2)^{1/2}} \quad , \tag{7.24a}$$

$$\frac{1}{E_2}\frac{dE_2}{dz} = \frac{\alpha_2}{(1+hE_2^2/E_o^2+ g\ E_1^2/E_o^2)^{1/2}} \quad , \tag{7.24b}$$

where $h > g$. Expanded to first order in E_1^2/E_o^2 , E_2^2/E_o^2 these equations become the same as 7.22a,b with $\theta_{12}\theta_{21} < \beta_1\beta_2$. The effect of the condition $h > g$ is the same as that of $\theta_{12}\theta_{21} < \beta_1\beta_2$, that is if the linear gains are equal or nearly equal to fields tend to equalize. We might say a highly stable two field operation results.

No results valid for strong fields were obtained for the more complicated transitions of this chapter, where the results show that strong coupling can exist for opposite circularly polarized signals. Equations 7.24a,b with $g > h$ when expanded to first order in E_1^2/E_o^2, E_2^2/E_o^2 give equations similar to 7.22a,b and with $\theta_{12}\theta_{21} > \beta_1\beta_2$. We could assume therefore that for opposite circularly polarized fields and $J_a = J_b$ the equations valid for arbitrary intensities possibly are of this form. If this is indeed the case the effect is as follows. Both signals continue to be amplified but the actual gain of a weaker signal is smaller than that of the stronger one, the unbalance between them growing continuously. While no quenching of the weaker signal takes place since the gains remain positive, this is an unstable situation in which any initial unbalance is magnified.

In all the results derived we have assumed that no losses exist in the medium. This is why there is always a positive gain regardless of how strong the field intensities are. In a laser oscillator, which has weak field equations similar to 7.22a,b except with time derivative in place of the space derivative, there are diffraction losses. These can be accounted for by the subtraction of a fixed "linear" loss (11), thus replacing $\alpha_{1,2}$ by $\alpha_{1,2} - \varkappa$. It is readily seen that the result is that for strong coupling the gain of the weaker mode can become negative (even while the iterative results are valid) and it is quenched by the stronger one. This highly unstable situation was discussed in detail by Lamb (5). There are many practical cases when losses exist in a laser amplifier. Intensity dependent losses of course introduce further complications and have to be treated by new nonlinear equations. An actual laser signal is most commonly a quasi plane wave of limited transverse extent with a slight divergence. It will now be shown that a small divergence of the laser beam can be treated as a "linear" loss. Let us assume that the signal emerges from an aperture of radius a with a divergence θ (typical θ is roughly 1 sec) and enters the amplifying medium at a distance R away. Then

$$\frac{1}{I}\frac{dI}{dz} = \frac{dA}{dz}\frac{1}{A} = -\frac{2\pi\ \theta(a_o+R\theta)}{\pi(a_o+R\theta)^2} = \frac{-\theta}{a_o+R\theta} \quad , \tag{7.25}$$

since $dA = \pi[a_o+(R+dz)\theta]^2-\pi[a_o+R\theta]^2 = 2\pi\theta(a_o+R\theta)$.

If $(R + z)\theta \ll a_o$, $\frac{1}{E}\frac{dE}{dz} = -\frac{1}{2}\frac{\theta}{a_o} = -\kappa$.

Rewriting equations 7.24a,b (with $\alpha_1 = \alpha_2$) as

$$\frac{1}{E_1}\frac{dE_1}{dz} = \frac{\alpha}{(1+hE_1^2/E_o^2+gE_2^2/E_o^2)^{1/2}} - \kappa \qquad (7.26a)$$

$$\frac{1}{E_2}\frac{dE_2}{dz} = \frac{\alpha}{(1+hE_2^2/E_o^2+gE_1^2/E_o^2)^{1/2}} - \kappa \quad , \qquad (7.26b)$$

it can be readily ascertained that if $g > h$ not only is any inequality of the two signals magnified by the higher gain of the stronger field but the weaker signal can eventually be quenched, while the stronger one eventually reaches a maximum intensity.

The case of linearly polarized fields is somewhat more complicated since there exists, except in the special case of perpendicularly polarized fields in a $J = 1 \rightarrow J = 0$ or $J = 1 \rightarrow J = 1$ laser, combination tone generation as well. Examination of Close's strong field solutions given in chapter six shows that the two aspects of the interaction are "mixed up" in the complete solutions and there is no simple way of separating out the saturation part. Numerical methods can be used to determine the behavior of the gain for unequal input intensities. Without doing this it is suggested that qualitatively the effects of strong coupling are similar to the cases already discussed. The additional effect of combination tone generation, being a parametric transfer of energy from the input to the sideband frequencies, amounts to intensity dependent losses.

7.6 Summary and Conclusions

In this chapter we have generalized the results of the previous chapters to arbitrary $J_a \rightarrow J_b$ transitions. While qualitatively the results were found to be often similar, several new results different from those for the $J = 1 \rightarrow J = 0$ case also emerged. These can be summarized as follows.

The interaction between the two waves depends on the frequency separation, the magnetic field and on the J values of the levels. For opposite circular polarization of the signals in zero or weak magnetic fields and for small frequency separation the coupling is strong for $\Delta J = 0$ and weak for $\Delta J = 1$. For nonzero magnetic fields the coupling is always weak unless the g factors of the levels are equal, in which case $\Delta\nu - \omega_{+-}$ takes the place of $\Delta\nu$ and otherwise the zero magnetic field results apply. For perpendicular linear polarizations (and small $\Delta\nu$) in zero magnetic field the coupling is strong for $\Delta J = 1$ and weak for $\Delta J = 0$. The transitions when one of the J values is unity are exceptions, the coupling being weak for either opposite circular or perpendicular linear polarizations in zero magnetic field although strong coupling regions can exist in the later case in weak magnetic fields.

Combination tone generation was found to occur even in zero magnetic field for perpendicular linear polarization of the input fields in all but the above named special cases. There is of course no combination tone generation for opposite circular waves, regardless of the J values of the levels. When comparisons are appropriate our results are in agreement with those of other works on the subject (22, 25,26) .

CHAPTER EIGHT

SUMMARY AND CONCLUSIONS

In this chapter we first give a brief summary of the results obtained in section 8.1. Section 8.2 contains some applications of the theory, while in Section 8.3 we briefly discuss the relationship to some other nonlinear processes. Possible experiments are suggested in Section 8.4 and some useful extensions are discussed briefly in Section 8.5.

8.1 Summary

In the preceding chapters various nonlinear effects were studied in a gas laser amplifier which may have an axial, D.C. magnetic field. Among these were calculations of the nonlinear intensity dependent gain, the strength of the interaction (coupling) between two or more input waves, nonlinear phase shift and Faraday rotation, induced anisotropy of the medium and the generation of new frequencies. Both stationary and Maxwellian velocity distribution of the excited atoms were considered and the effects of intermediate Doppler broadening were included. Although the majority of the results were obtained by calculating only the lowest order term in a perturbational expansion , some results valid for arbitrarily strong fields were also found. While these are not sufficiently general to be always applicable they do give an idea of the limitations of the weak field results and of how the solutions behave when the field intensities are high. Besides being functions of the magnetic field, and of the frequency separation and tuning of the input signals, all the nonlinear effects were found to depend greatly on the polarization states of the optical waves and on the J values of the laser levels.

Perhaps the single most significant result of the preceding chapters is the importance of the coherent double quantum interactions that can take place in a multilevel laser amplifier. These processes greatly influence all the nonlinear effects and under favorable conditions are as strong or stronger in their influence on the behavior of the laser as the normal saturation of the population inversions. The instabilities found in two field operation of the laser amplifier are largely due to the coherences between the sublevels caused by the double quantum processes, and the interesting narrow resonances that were found to occur when the Zeeman separation of the levels equals the frequency difference of the input fields are also attributable to these Raman type interactions.

8.2 Applications of the Theory

Aside from calculating the frequency, amplitude, magnetic field and polarization dependence of various processes in the nonlinear interaction of optical frequency waves the above results have several other interesting applications.

The existence of the narrow resonances in the interaction of two fields when the level separation equals the frequency difference of the interacting waves makes it possible to use this effect for the study of level structures. Since the output of the laser amplifier has a dip where the interaction goes through a resonance, the hyperfine structure of transitions between levels, at least one of which has a slow natural decay rate, can be probed by applying a variable magnetic field to the medium and thus tuning the levels. A theory emphasizing this application for both amplifiers and oscillators, with scalar electromagnetic fields, has been very recently published by Schloessberg and Javan (26) who have also reported results of a study of the hyperfine level structure of a number of isotopes of Xenon utilizing a laser oscillating on the 3.37μ line (39).

With only minor modifications the theory is applicable to multilevel structures where laser action is possible in several transitions having some common levels. Xenon, for example has a number of laser transitions in the 2.02μ - 5.57μ range several of which start or terminate on a common level. In an oscillator set up some of the possible laser lines oscillate while others do not. To properly study the competition between oscillating or about to osscillate transitions it is necessary to take into account the effects of possible double quantum interactions. Rate equations are inadequate and a somewhat modified theory similar to ours should be used.

While the theory is not well suited for studying the behavior of cavity modes in a laser oscillator it should prove useful for such studies too under certain circumstances. For example the modes of a laser oscillator with mirrors of different reflectivities will not be true cavity modes, but will have travelling wave components. Since oppositely running waves were also examined the theory would be useful in studying the effects of such a situation. Nonlinear interactions in a closed path ring laser also must be studied in terms of travelling waves. Calculations of polarization and magnetic field effects should be highly interesting for this type of device.

8.3 Relationship to Other Nonlinear Effects

There is a close relationship between the nonlinear effects examined in this work and certain other effects recently observed or treated theoretically. We have already mentioned briefly optical frequency mixing, treated in some detail by Javan and Szöke (45). In this effect the coherent double quantum interaction of two strong fields produces an induced polarization at the difference frequency. If the upper levels are the magnetic sublevels of a transition the difference frequency radiation is magnetic dipole type. The calculations of this induced polarization are similar to those performed in the preceeding chapters, the difference being that (using our terminology) the density matrix element ρ_{+-}, rather than $\rho_{\pm b}$

is of interest. In a perturbational expansion the second, fourth, etc., orders produce contributions to the mixing frequency polarization. The above authors also obtained results valid for strong fields; these solutions can be related to the expressions for ρ_{+-} obtained in the course of our calculations of ρ_{+b} valid for intense fields. It is interesting to inquire as to what are the possible effects of the polarization states of the input fields, of somewhat more complex level structures, or the effect of multiple spectral components in each of the input fields. It is expected that the latter will cause modulation of the population densities in the manner seen in our work and induce sidebands on the difference frequency field as well.

The importance of the double quantum interactions in the nonlinear effects treated by the theory suggests a close relationship to Stimulated Raman Scattering as well. This effect received a great deal of experimental and theoretical attention of late (49) . Most of the theoretical work is different in spirit and approach from our work. Javan, however, treated the stimulated amplification of the Stokes and anti-Stokes frequencies by a density matrix approach similar to ours, using scalar electromagnetic fields (50). To make the connection between his work and ours it is necessary to assume different excitation conditions in the derivation of the integral equations. It is interesting to note that for a case similar to that discussed in chapter five, with opposite circularly polarized fields and correspondingly polarized transitions, there can be no parametric amplification of the anti-Stokes frequency for the same reason that no combination tone generation was found. The case of two linearly or elliptically polarized fields relates more closely to SRS since the total field interacts with both transitions and parametric generation of new frequencies as well as transfer of energy between the two input fields can take place.

8.4 Possible Experiments

As indicated at times in the course of the discussions it should be quite feasible to observe and measure many of the nonlinear effects calculated in the preceeding chapters. The following experiments are suggested.

(1) Strong Coupling. It should be possible to observe the instabilities that arise from the strong coupling of waves of various polarization, and the variation of this effect with frequency separation and with magnetic field. The simplest experiment is to detect strong coupling, in zero magnetic field, between opposite circularly polarized components of a single linearly polarized signal as indicated by equation 7.12.

In this experiment a single mode Brewster angle laser oscillating on a $\Delta J = 0$ $(J > 1)$ transition will provide the signal which will be fed into a high gain test amplifier. The output signal will be elliptical and the degree of ellipticity will be detected by a variable angle polarizer-analyzer followed by a detector. To measure the variation with frequency separation, the signal source can be replaced by a single mode laser with plane windows to which an axial magnetic field is applied. This configuration provides two opposite circularly polarized waves of variable (small) frequency separation (19,20). Suitable transitions are available in the Xe laser.

(2) Combination Tone Generation. For parallel polarized input fields and no magnetic field this has already been observed in a laser amplifier by Close (30). The polarization and magnetic field effects described in chapter five and six can be investigated by simple extensions of his experiment. For example the sudden appearance of combination tones between perpendicularly polarized signals in a $J = 1 \rightarrow J = 0$ laser with magnetic field (Figure 20) can be detected in a setup identical to that of Close except for the addition of a magnetic field to the test amplifier.

(3) The Effects of Double Quantum Transitions and of Strong Saturation on the Faraday Rotation. Order of magnitude calculations in chapter five show that the anomalous rotation at low magnetic fields shown on Figure 15 should be observable using a high gain laser amplifier. The experimental arrangement will be similar to that described in (1) with a single mode laser providing a linearly polarized signal. The saturation of the Faraday rotation in the normal region, as described by equation 5.63 and shown on Figure 13, necessitates the addition of one or more power amplifiers to bring the signal level into the highly saturated region. A transition with a fairly narrow Doppler width is desirable for the latter experiment, while a strongly Doppler broadened line is preferable for the former.

(4) The Rotation of Two Linearly Polarized Fields in Zero Magnetic Field. This effect was described in section 6.3.3A where it was also shown that rotations as high as 30° could be expected in an amplifier with gain of 30 db or more. For observation of this anisotropic effect a source providing two fields, polarized linearly with an arbitrary angle between them, with variable, stable, small frequency separation is needed. To achieve this, a single mode laser with plane windows gives an output of two opposite circularly polarized waves whose separation is a function of the magnetic field (18, 19). This output will be converted into the desired two linearly polarized waves by means of quarter wave plates and polarizers in a suitable optical configuration. A sensitive detector, tuned to the beat frequency, preceeded by a variable polarizer-analyzer, will be used to measure the angle between the polarization vectors in the output. As an alternate signal source, providing signals with frequency separation tunable over a wider range, a single mode laser coupled with a Debye-Sears modulator can be utilized.

(5) Uses of the theory to probe the level structure of a medium have already been described. Such an experiment has been carried out by Schlossberg and Javan (39). In addition, several of the nonlinear effects described in the previous chapters depend critically on the lifetimes of the levels and on the saturation field E_o^2. The latter in turn depends on the matrix element $|\langle a\|p\|b\rangle|^2$. These quantities can be determined by several of the experiments suggested above. In addition it might be possible to measure excitation rates since the

product of the gain parameter α and the saturation field intensity E_o^2 depends only on the decay rates and on the excitation rates.

Following is a partial list of some known laser transitions that are suitable for some or all of the above suggested experiments. Only neutral laser transitions capable of CW oscillations are listed. Obviously, both high gain and low saturation power are the best for observations of nonlinear effects.

Wavelength and Substance		J values	Saturation parameter (E_o^2)	Gain
2.65μ	He-Xe or pure Xe	1 → 0	Not known	Fairly high
3.99μ	He-Xe	0 → 1	Not known	High
1.52μ	He-Ne	1 → 0	Not known	Low (6%/m) Suitable for oscillators only
2.03μ	He-Xe or pure Xe	1 → 1	Not known but probably low	Fairly high
3.39μ	He-Ne	1 → 2	Low (1mw/cm^2)	Very high (40 db/m)
3.68μ	He-Xe	2 → 2	Probably low	High
3.51μ	He-Xe	3 → 2	Very low	High
5.57μ	He-Xe or pure Xe	4 → 3	Probably low	Very high

7.5 Extensions

It would be fairly easy to extend the theory to deal with cavity modes. A number of papers have appeared recently dealing with polarization and magnetic field effects in laser oscillators (14, 15, 18 - 25). The case of a nonzero magnetic field, generalized J values and multimode operation, however, has not been fully investigated.

Another interesting extension would be to deal with non-axial magnetic fields along the lines of this work. Particularly interesting would be the interaction of the σ and π modes in an amplifier with transverse magnetic field. Some work has been done on this problem also recently (20, 22) but further investigation is possible, particularly on combination tones.

A very fruitful extension would be to find solutions valid for arbitrarily strong fields, especially for the cases where there is strong coupling and/or combination tone generation.

Finally, the theory should be extended to include the effects of collisions. Recent experimental observations have shown (38) that collisions often play an important role in the interaction between waves of various polarization by mixing and perturbing the levels in a time shorter than the life times.

APPENDIX I

EVALUATION OF THE DOPPLER BROADENING INTEGRALS

In this appendix we evaluate the first and third order Doppler integrals that are encountered in the calculations of the induced polarization. Rather than calculating separately each of the expressions for the various cases, we perform the integration for the first (two opposite circularly polarized fields in the same directions), and thereafter only those terms that are different from previously encountered ones will be considered.

I.A Doppler Integrals for Opposite Circular Fields in the Same Direction

The complete expression for the induced polarization is given by equation 5.10 . The first integral is a well known one, it is the integral representation for the complimentary error function of complex argument (6 , 46). (The various terms are indicated by their order number in the equation).

$$(1) = \frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{e^{-\xi}d\xi}{a+i(x_-+\xi)} = w^*(x_-+ia) \quad , \tag{I.1}$$

where $w(z) = \exp(-z^2)\mathrm{erfc}(-iz) = \frac{2}{\sqrt{\pi}}\exp(-z^2)\int_z^{\infty}\exp(-t^2)dt$.

This function has the properties:

$$w(-x+iy) = w^*(x+iy) \tag{I.2}$$

$$w'(z) = -2z\, w(z) + 2i/\sqrt{\pi} \quad . \tag{I.3}$$

Also, evidently $w(0+iy)$ is real and for large $|z|$ the asymptodic form gives

$$w(z) \simeq \frac{i}{\sqrt{\pi}} \left(\frac{1}{z} + \frac{1}{2z^3}\right) \quad . \tag{I.4}$$

For a more complete description of the properties of w the references should be consulted.

The second term can be simply evaluated as the derivative of the same function

$$(2) = \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{e^{-\xi^2} d\xi}{[a+i(x_-+\xi)]^2} = -\frac{\partial}{\partial a} \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{e^{-\xi^2} d\xi}{a+i(x_-+\xi)} = -\frac{\partial}{\partial a} w^*(x_-+ia) \; . \tag{I.5}$$

Since $w'(z) = \frac{\partial}{\partial x} w = -i \frac{\partial}{\partial a} w$. Then,

$$(2) = \frac{2}{\sqrt{\pi}} - 2(a-ix_-)w^*(x_-+ia) \quad . \tag{I.6}$$

The other terms can be broken up into partial fractions and thus reduced to integrals of the type (1) .

$$(3) = \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{e^{-\xi^2} d\xi}{a+i(x_-+\xi)} \frac{1}{a-i(x_-+\xi)} = \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{1}{2a} \frac{e^{-\xi^2} d\xi}{a+i(x_-+\xi)} + \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{1}{2a} \frac{e^{-\xi^2} d\xi}{a+i(-x_--\xi)} \; . \tag{I.7}$$

If in the second integral we let $\xi \rightarrow -\xi$ and use the property given by (I.2) ,

$$(3) = \frac{1}{a}\,\text{Re}\; w^*(x_-+ia) \quad . \tag{I.8}$$

$$(4) = \frac{1}{\pi}\int_{-\infty}^{+\infty} \frac{e^{-\xi^2}d\xi}{a+i(x_-+\xi)}\,\frac{1}{a+i(x_++\xi)} = \frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{i}{x_--x_+}\left[\frac{1}{a+i(x_-+\xi)} - \frac{1}{a+i(x_++\xi)}\right]e^{-\xi^2}d\xi$$

$$= \frac{i}{x_--x_+}\left[w^*(x_-+ia) - w^*(x_++ia)\right] \quad . \tag{I.9}$$

$$(5) = \frac{1}{\pi}\int_{-\infty}^{+\infty} \frac{e^{-\xi^2}d\xi}{a+i(x_-+\xi)}\,\frac{1}{a-i(x_++\xi)} = \frac{1}{\pi}\,\frac{1}{2a+i(x_--x_+)}$$

$$\times\left[\int_{-\infty}^{+\infty}\frac{e^{-\xi^2}d\xi}{a+i(x_-+\xi)} + \int_{-\infty}^{+\infty}\frac{e^{-\xi^2}d\xi}{a+i(-x_+-\xi)}\right] \quad . \tag{I.10}$$

Again we let $\xi \to -\xi$ in the second integral and obtain

$$(5) = \frac{1}{2a+i(x_-+x_+)}\left[w^*(x_-+ia)+w(x_++ia)\right] \quad . \tag{I.11}$$

$(6) \equiv (2)$

$(7) \equiv (3)$

Equations 5.11-5.12 follow, since $x_- = x_o+y-\zeta$ and $x_+ = x_o-y+\zeta$.

I.B Other Doppler Integrals

The following integral not yet evaluated, is encountered in the strong field solutions for a single linearly polarized wave.

$$\frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{a-i(y+\xi)}{b'^2+(y+\xi)^2}e^{-\xi^2} = \frac{1}{\pi}\int_{-\infty}^{+\infty}\left[\frac{a+b'}{2b'}\,\frac{1}{b'+i(y+\xi)} + \frac{a-b'}{2b'}\,\frac{1}{b'+i(-y-\xi)}\right]e^{-\xi^2}d\xi \; .$$

Letting $\xi \to -\xi$ in the second term this equals

$$\frac{1}{2b'}\left[(a+b')w^*(y+ib')+(a-b')w(y+ib')\right] \quad . \tag{I.12}$$

In the solutions for opposite circularly polarized fields in opposite directions, the new terms in equation 5.10, after setting $k_+ = -k$ as specified, using the techniques of I.A , are

$$(4) = \frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{e^{-\xi^2}d\xi}{a+i(x_-+\xi)}\,\frac{1}{a+i(x_+-\xi)}$$

$$= \frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{1}{2a+i(x_-+x_+)}\left[\frac{1}{a+i(x_-+\xi)}+\frac{1}{a+i(x_+-\xi)}\right]e^{-\xi^2}d\xi$$

$$= \frac{1}{2a+i(x_-+x_+)}\left[w^*(x_-+ia)+w^*(x_++ia)\right] \quad . \tag{I.13}$$

$$(5) = \frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{e^{-\xi^2}d\xi}{a+i(x_-+\xi)}\,\frac{1}{a-i(x_+-\xi)} =$$

$$= \frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{i}{x_-+x_+}\left[\frac{1}{a+i(x_-+\xi)}-\frac{1}{a+i(-x_++\xi)}\right]e^{-\xi^2}d\xi$$

$$= \frac{i}{x_-+x_+}\left[w^*(x_-+ia)-w(x_++ia)\right] \quad . \tag{I.14}$$

$$(6) = \frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{e^{-\xi}d\xi}{A+i(y-\zeta+\xi)}\,\frac{1}{[a+i(x_-+\xi)]^2} = \frac{1}{\pi}\int_{-\infty}^{+\infty}\left\{\frac{1}{[a-A+i(x_--y+\zeta)]^2}\left[\frac{1}{A+i(y-\zeta+\xi)}\right.\right.$$

$$\left.\left.-\frac{1}{a+i(x_-+\xi)}\right]-\frac{1}{a-A+i(x_--y+\zeta)}\,\frac{1}{[a+i(x_-+\xi)]^2}\right\}e^{-\xi^2}d\xi$$

$$= \frac{1}{[B+i(x_- - y+\zeta)]^2}\left[w^*(y-\zeta+iA) - w^*(x_- + ia)\right] - \frac{1}{B+i(x_- - y+\zeta)}$$

$$\times\left[\frac{2}{\sqrt{\pi}} - 2(a+ix_-)w^*(x_-+ia)\right] \quad . \tag{I.15}$$

$$(7) = \frac{1}{\pi}\int_{-\infty}^{+\infty} \frac{e^{-\xi^2}d\xi}{A+i(y-\zeta+\xi)}\,\frac{1}{a+i(x_-+\xi)}\,\frac{1}{a-i(x_+-\xi)}$$

$$= \frac{1}{\pi}\int_{-\infty}^{+\infty}\left\{\frac{1}{[a-A+i(x_- - y+\zeta)][a-A-i(x_+ + y-\zeta)]}\,\frac{1}{A+i(y-\zeta+\xi)}\right.$$

$$\left. - \frac{i}{x_- + x_+}\left[\frac{1}{a-A+i(x_- - y+\zeta)}\,\frac{1}{a+i(x_-+\xi)} - \frac{1}{a-A-i(x_+ + y-\zeta)}\,\frac{1}{a+i(-x_+ + \xi)}\right]\right\} e^{-\xi^2} d\xi$$

$$= \frac{1}{B+i(x_- - y+\zeta)}\,\frac{1}{B-i(x_+ + y-\zeta)}\, w^*(y-\zeta+iA)$$

$$- \frac{i}{x_- + x_+}\left[\frac{1}{B+i(x_- - y+\zeta)}\, w^*(x_- + ia) - \frac{1}{B-i(x_+ + y-\zeta)}\, w(x_+ + ia)\right] \quad . \tag{I.16}$$

Substituting $x_\pm = x_o \mp y \pm \zeta$ equations 5.66 - 5.67 follow.

Finally in chapter six for the case of two linearly polarized signals if we let $\omega_{+b} - \nu_1 = x_{11}$, $\omega_{+b} - \nu_2 = x_{12}$, $\omega_{-b} - \nu_1 = x_{21}$ and $\omega_{-b} - \nu_2 = x_{22}$ it is seen that none of the integrals to be evaluated are new but can be obtained from the ones evaluated previously by simple changes of variables. For example

$$(4) = \frac{1}{\pi}\int_{-\infty}^{+\infty} \frac{e^{-\xi^2}d\xi}{a+i(x_{11}+\xi)}\,\frac{1}{a+i(x_{12}+\xi)} \quad \text{is identical to}$$

(4) in I.A, if in the latter $x_- \rightarrow x_{11}$ $x_+ \rightarrow x_{12}$; and so on for all the other terms. Using the identities

$$x_{11} = x_o + y - \eta \qquad x_{21} = x_o - y - \eta$$

$$x_{12} = x_o + y + \eta \qquad x_{22} = x_o - y + \eta \quad ,$$

equations 6.12-6.18 are obtained. The same holds for all the Doppler integrals of chapter seven.

APPENDIX II

RATE EQUATION CALCULATIONS OF THE INTERACTION OF TWO OPPOSITE CIRCULARLY POLARIZED WAVES IN A THREE LEVEL SYSTEM

The purpose of this Appendix is to show that rate equation calculations for the three level system of chapter five with two opposite circularly polarized signals produce results that include the common level mutual saturation but not the double quantum interaction, which cannot be obtained using this method. The approach and nomenclature will be similar to those of Gordon, White and Rigden (10), with modifications were necessary. The rate equations for those excited atoms that are moving with velocity v can be written

$$\dot{n}_-(v,z) = S_a(v) - n_-(v,z)\left[A_a+B'_{-b}(v,\nu_+)I_+(\nu_+,z)\right]+n_b(v,z)B'_{-b}(v,\nu_+)I_+(\nu_+,z), \tag{II.1}$$

$$\dot{n}_+(v,z) = S_a(v)-n_-(v,z)\left[A_a+B'_{+b}(v,\nu_-)I_-(\nu_-,z)\right]+n_b(v,z)B'_{+b}(v,\nu_-)I_-(\nu_-,z), \tag{II.2}$$

$$\begin{aligned}\dot{n}_b(v,z) = {} & S_b(v)+n_+(v,z)\left[A_{+b}+B'_{+b}(v,\nu_-)I_-(\nu_-,z)\right]\\ & +n_-(v,z)\left[A_{-b}+B'_{-b}(v,\nu_+)I_+(\nu_+,z)\right]\\ & -n_b\left[A_b+B'_{+b}(v,\nu_-)I_-(\nu_- z)+B'_{-b}(v,\nu_+)I_+(\nu_+,z)\right] .\end{aligned} \tag{II.3}$$

Where $n_-(v,z)$, $n_+(v,z)$ and $n_b(v,z)$ are the volume density of atoms in the two upper and the one lower laser levels, moving with velocity v at plane z along the amplifier. The intensities of the two circularly polarized waves are given by $I_\pm(\nu_\pm, z)$ and $S_a(v)$ is the pumping rate to either of the two upper levels while $S_b(v)$ is the pumping rate to the lower level. The $A_{a,b}$'s are the Einstein A coefficients or the spontaneous decay rates of the levels a and b respectively, while the spontaneous decay rate from levels a to level b is denoted by $A_{\pm b}$. The rate of stimulated emissions for atoms with Doppler velocity v due to radiation at ν is given

$$B'_{\pm b}(v,\nu_\mp) = B'_{b\pm}(v,\nu_\mp) = B_{\pm b}\,\frac{(2/\pi)\Delta\nu_n}{(\Delta\nu_n)^2+4(\omega_{\pm b}+\nu v/c-\nu_\mp)^2} \tag{II.4}$$

in which $B_{\pm b}$ are the appropriate Einstein B coefficients (50). The normalization is such that

$$\int_0^\infty B'_{\pm b}(v,\nu_\mp)\,d\nu_\mp = B_{\pm b} \quad .$$

The gains can be written as

$$\frac{dI_\mp(\nu,z)}{dz} = \frac{\hbar\nu I_\mp(\nu,z)}{4\pi}\int_{-\infty}^{+\infty} B'_{\pm b}(v,\nu_\mp)\left[n_\pm(v,z)-n_b(v,z)\right]dv \quad . \tag{II.5}$$

For the steady state condition, $\dot{n}_\pm = \dot{n}_b = 0$, we can rewrite equations II.1-II.3 as follows

$$
\begin{bmatrix}
B'_{-b}I_-/4\pi + A_a & 0 & -B'_{-b}I_-/4\pi \\
0 & B'_{+b}I_+/4\pi + A_a & -B'_{+b}I_+/4\pi \\
-B'_{-b}I_-/4\pi - A_{-b} & -B'_{+b}I_+/4\pi - A_{+b} & B'_{+b}I_+/4\pi + B'_{-b}I_-/4\pi + A_b
\end{bmatrix}
\begin{bmatrix} n_- \\ n_+ \\ n_b \end{bmatrix}
=
\begin{bmatrix} \lambda_a \\ \lambda_a \\ \lambda_b \end{bmatrix}
\tag{II.6}
$$

$$
n_- - n_b = \frac{\begin{vmatrix} S_a & \cdot & \cdot \\ S_a & \cdot & \cdot \\ S_b & \cdot & \cdot \end{vmatrix} - \begin{vmatrix} \cdot & \cdot & S_a \\ \cdot & \cdot & S_a \\ \cdot & \cdot & S_b \end{vmatrix}}{|\,\mathrm{Det.}\,|}
$$

$$
= \frac{S_a[(A_b-(A_{-b}+A_{+b}))B'_{+b}I_+/4\pi + A_a(A_b-(A_{-b}+A_{+b}))] - S_b[A_a B'_{+b}I_+/4\pi + A_a^2]}{A_a^2 A_b + (A_b+2A_a-A_{+b}-A_{-b})(B'_{-b}I_-/4\pi)(B'_{+b}I_+/4\pi) + A_a(A_a+A_b-A_{-b})B'_{-b}I_-/4\pi + A_a(A_a+A_b-A_{+b})B'_{+b}I_+/4\pi}
\tag{II.7}
$$

If we neglect A_{-b} and A_{+b} as we do in the density matrix approach we get

$$B'_{-b}(n_- - n_b) = \frac{\frac{1}{\sqrt{\pi}u} e^{-v^2/u^2} \left[1 + \frac{B'_{+b}I_+}{4\pi} \frac{A_b}{A_a A_b}\right] \left[\frac{S_{ao}}{A_a} - \frac{S_{bo}}{A_b}\right] B'_{-b}}{1 + \frac{A_a + A_b}{A_a A_b}\left[\frac{B'_{-b}I_-}{4\pi} + \frac{B'_{+b}I_+}{4\pi}\right] + \left[\frac{(A_a+A_b)^2}{A_a^2 A_b^2} - \frac{A_a^2}{A_a^2 B_b^2}\right] \frac{B'_{-b}I_-}{4\pi} \frac{B'_{+b}I_+}{4\pi}} \quad . \tag{II.8}$$

Where S_a and S_b have been taken to have the Maxwellian velocity profile

$$S_i = S_{io} \frac{1}{\sqrt{\pi}u} e^{-v^2/u^2} \tag{II.9}$$

Defining $I_o^{-1} = \frac{A_a + A_b}{A_a A_b} \frac{B_{\pm b}}{4\pi} \frac{2}{\pi \Delta\nu_n}$ we get, since $\Delta\nu_n \simeq 2\gamma_{ab} \simeq A_a + A_b$

$$\frac{1}{I}\frac{dI}{dz} = h\nu \frac{B_{+b}}{4\pi}\left(\frac{S_{ao}}{A_a} - \frac{S_{bo}}{A_b}\right) \frac{1}{\sqrt{\pi}u}$$

$$\times \int_{-\infty}^{+\infty} \frac{e^{-v^2/u^2}\left[\gamma_{ab}^2 + \gamma_{ab}A_b I_+/2I_o + (\omega_{-b} - \nu_+ + iv)^2\right]\gamma_{ab}}{\left[\gamma_{ab}^2(1+I_+/I_o) + (\omega_{-b} - \nu_+ + kv)^2\right]\left[\gamma_{ab}^2(1+I_-/I_o)(\omega_{+b} - \nu_- + kv)^2\right] - A^2\gamma_{ab}^2 I_+ I_-/4I_o} \tag{II.10}$$

where γ_{ab} has been substituted for $\frac{A_a + A_b}{2}$.

Letting $2\alpha = h\nu \frac{B_{+b}}{4\pi} \frac{\sqrt{\pi}}{ku}$ we obtain after dividing through by $(ku)^2$

$$\frac{1}{I_-}\frac{dI}{dz} = 2\alpha \int_{-\infty}^{+\infty} \frac{a\left[a^2(1+(B/a)(I_+/I_o)) + (x_+ + \xi)^2\right] e^{-\xi^2} d\xi}{\left[a^2(1+I_-/I_o) + (x_- + \xi)^2\right]\left[a^2(1+I_+/I_o) + (x_+ + \xi)^2\right] - A^2 a^2 I_+ I_-/4I_o} \, , \tag{II.11}$$

where $a = \frac{\gamma_{ab}}{ku} = \frac{A_a+A_b}{2ku}$, $A = \frac{A_a}{2ku} = \frac{\gamma_a}{2ku}$, $B = \frac{A_b}{2ku} = \frac{\gamma_b}{2ku}$.

For $\nu_+ = \nu_- = \nu = \frac{\omega_{+b}+\omega_{-b}}{2}$, i.e., $x_+ = -y$, $x_- = +y$, $I_+ = I_- = I/2$ which is the single, center tuned linearly polarized wave case the above equation is identical with the gain part of equation 5.55 which was derived for large magnetic fields neglecting the coherence effects which are very small for that case $(\rho_{+-} \approx 0)$. Thus the rate equation approach which ignores the double quantum interactions is valid only when ρ_{+-} is negligible. For zero magnetic field $(y = 0)$ the double quantum interactions make up the difference between equation II.11 and the correct expression 5.52 . Note that if $A_b \to 0$ and $A_a/2 \approx \gamma_{ab}$, i.e., $A = a$, then II.11 does give the right result. This is consistent with the fact that for $A_b \ll A_a$ the double quantum interactions are negligible as it was observed in chapter five.

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