NONLINEAR EFFECTS IN TRAVELING WAVE LASER AMPLIFIERS

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ABSTRACT

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Using semiclassical radiation theory, a formalism similar to that used by Lamb in his "Theory of an Optical Maser" is developed for studying the amplification of vector traveling waves in a laser-type medium. The effect of the medium on the waves is given in terms of space (or time) dependent field amplitudes and phases and a nonlinear index of refraction. With particular emphasis on typical gaseous media, the effects of Doppler broadening are treated in detail for arbitrary ratios of natural to Doppler linewidths. Polarization and propagation vectors in various directions are considered, and the nonlinear effects are found to make an isotropic medium effectively anisotropic.

Lowest order nonlinear effects (due to a polarization cubic in the field amplitudes) are studied extensively, and the frequency dependence of several of these processes is presented in graphical form. In particular, the introduction of fields at new frequencies and polarization effects are considered. The characteristics of these nonlinear processes peculiar to Doppler broadened lines are discussed, and the processes are interpreted in terms of saturation and coherent modulation of the population inversion density.

Strong nonlinear effects are considered in a more approximate way and are found to consist of saturation of the various linear and nonlinear processes previously considered. These strong nonlinear effects should occur at low enough intensities to be easily observed in practice on a CW basis. With the present formalism, the analytical results of Gordon, White and Rigden regarding gain saturation in laser amplifiers are obtained, and the extension is made to include frequencies away from line center and the effects of multiple spectral components. Again, the introduction of fields at new frequencies is considered in detail. These results are also discussed in terms of saturation and coherent modulation of the populations and "hole burning".

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CHAPTER ONE

INTRODUCTION

In the following chapters we shall study the amplification of traveling electromagnetic waves by a laser medium, i.e., a medium with an inverted population with respect to some two levels (1). We are particularly interested in studying the characteristics of typical gaseous media (2), where the atoms have a distribution of velocities.

It is well-known that the inverted population of a laser medium leads to "inverted absorption" or gain, so that an incident wave with spectral components within the transition linewidth is amplified exponentially with distance. Since this amplification is brought about by induced transitions of the atoms, the average number of atoms in the laser levels will change when a field is applied. It is this saturation and other nonlinear processes in which we are primarily interested, although we shall also obtain the linear amplification for comparison. Our purpose is to find what nonlinear processes are typically present, and to study and understand their characteristics and how they affect the linear result.

To study these effects, we shall use semiclassical radiation theory, as formulated by Kramers (3). This involves calculating the dipole moment induced in each atom by the field and using the expected value of this dipole moment in Maxwell's equations to find the reaction of the atoms on the field. This procedure provides a fundamental basis for the theory, and allows us to calculate explicitly the amplitude and frequency dependence of the various processes, in terms of basic atomic parameters. Rate equations are not adequate for our purposes, since they cannot predict coherent, field-dependent effects. Our results are derived in terms of space or time dependent field amplitudes and phases, and field-dependent gain per unit length and index of refraction functions. There is no external cavity or other assumed means of mode suppression or discrimination, so that we are able to study the effect of closely spaced spectral components in the presence of dissipation processes.

Armstrong, Bloembergen, Ducuing and Pershan (4) treated the nonlinear interaction of traveling waves in dielectric media using a similar formulation and the method of selecting particular Fourier components of the response. Dissipation in the atoms and Doppler broadening were not considered, so that the theory applied to parametric effects in stationary atoms. Although vector waves were used, no polarization effects were studied.

In extensive calculations on various nonlinear processes, Bloembergen and Shen (5) included the effects of atomic dissipation and considered the reaction of the medium on cavity modes. Only stationary atoms were studied, and polarization effects were not considered.

Schulz-DuBois and others have made detailed calculations on traveling wave masers (6), including saturation and some coherent effects (7-9). In particular, with waves incident at frequencies ω_1 and ω_2, an output at $2\omega_1 - \omega_2$ and $2\omega_2 - \omega_1$ is predicted. In the case of microwave masers, this effect was found to be quite small and observable only under extreme conditions (9); and the same conclusion was extrapolated to the optical range. In the following, we shall find that this and even higher order effects should be observable under CW operating conditions in typical gaseous laser amplifiers.

In considering the effects of saturation on gaseous absorption, Tang and Statz studied some polarization effects and noted that the atomic populations would be modulated (10). However, they did not consider any effects at frequencies other than those of the incident fields. Different relaxation rates of the upper and lower laser levels were not considered, nor were higher order perturbations on the cavity modes. Their simple treatment of Doppler broadening is not sufficient for our purposes.

Lamb has used the semiclassical radiation theory to make a detailed study of some characteristics of laser oscillators, including nonlinear effects (11,12). Since scalar cavity mode fields were used, polarization effects could not be studied. The treatment of Doppler broadening is limited to Doppler widths very large compared to the natural linewidth, and frequency spacings small compared to the Doppler width. Strong saturation is considered briefly, although not with respect to higher order nonlinear effects.

Haken and Sauermann (13,14) have considered similar effects, using similar techniques. In addition, they consider the polarization dependence of the lowest order saturation process.

Using a different approach, based on the Kramers-Kronig relations as well as semiclassical radiation theory, Bennett has studied some effects of saturation in laser oscillators (2,15). Doppler broadening was considered only to the extent that it resulted in an inhomogeneously broadened gain curve. Bennett's ideas of "hole burning" are very useful in the physical analysis of nonlinear processes in gaseous laser devices, and we shall extend and use them in our work.

Gordon, White and Rigden (16), using a method based on rate equations, have studied strong saturation effects in laser amplifiers and found good agreement with their experimental results. Only a single wave at line center is treated, although the extension to the case of a single wave at other frequencies is straightforward using the same approach. Using the method to be outlined in the following chapters, we shall obtain the same analytical results and extend them to cover more complicated situations, with multiple waves.

There are several other references which deal with related ideas and calculations, primarily in applications of the semiclassical radiation theory to various problems, including nonlinear effects. We shall refer to some of these during the discussion of our work. For a more complete listing and discussion of work pertaining to lasers, the review book by Birnbaum (1) is available.

The following work uses an approach which is generally similar to that taken by Lamb (12). The equations of motion for the atoms and vector fields are derived in Chapter 2. The technique and form of solutions of these equations is presented in Chapter 3. In Chapter 4 the formalism is applied to linear amplification. Lowest order nonlinear effects are studied in detail in Chapter 5, featuring the frequency dependence of several nonlinear effects including Doppler broadening.

These results, which are strictly good only for relatively small field strengths, are extended in Chapter 6, where some strong nonlinear effects are studied and related to the perturbation expansion solutions. Finally, in Chapter 7, we discuss the results, and applications and extensions of the theory.

Except for some brief discussion on effects of amplification of spontaneous emission, noise properties of laser amplifiers are not considered in the following work. Also, we do not treat the effects of collisions, harmonic generation or boundary conditions.

CHAPTER TWO

EQUATIONS OF MOTION FOR THE ATOMS AND FIELDS

2.1 Introduction

The purpose of this chapter is to introduce the quantities used to represent the medium and fields, and to find the equations of motion for the atom-field system. For sufficiently slowly varying fields, we can effectively deal with two separate systems, atoms and fields, and obtain the solution for the complete system by requiring that the separate solutions be consistent. The atoms are treated in section 2.2, the fields in section 2.3.

2.2 Microscopic Equations of Motion for the Atoms

In this section we derive the equations governing the behavior of each of the atoms comprising the medium. The characteristics of the atoms and their interaction with the electromagnetic field are discussed in 2.2.1, and the equations of motion for a single atom are derived in 2.2.2.

2.2.1 Description of the Atoms

Taking the electronic charge to be $-e$, the Hamiltonian for an electron in the atom in the presence of an electromagnetic field is, in the Coulomb gage (17)

$$H = \frac{1}{2m}(\underline{p} + e\underline{A})^2 + V \quad , \qquad 2.2.1\text{-}1$$

where V is the potential energy of the electron in the absence of an applied field, $\underline{A}$ is the vector potential of the applied field, $\underline{p}$ is the momentum operator for the electron and m is the electronic mass. Since $\nabla \cdot \underline{A} = 0$ in the Coulomb gage, $\vec{p} = -i\hbar\nabla$ commutes with $\underline{A}$ and 2.2.1-1 can be expanded as

$$H = \frac{p^2}{2m} + V + \frac{e\underline{p} \cdot \underline{A}}{m} + \frac{e^2A^2}{2m} \quad . \qquad 2.2.1\text{-}2$$

The part of H independent of $\underline{A}$ determines the eigenstates of the atomic system in the absence of an electromagnetic field. We designate this part

$$H_o = \frac{p^2}{2m} + V \quad . \qquad 2.2.1\text{-}3$$

The remainder represents the interaction between the field and atomic system and is designated as

$$H' = \frac{e\underline{p} \cdot \underline{A}}{m} + \frac{e^2A^2}{2m} \quad . \qquad 2.2.1\text{-}4$$

The time-independent eigenstates of the noninteracting atom are the solutions of the Schrodinger equation

$$H_o/\phi) = E/\phi) \quad , \qquad 2.2.1\text{-}5$$

where E is the energy eigenvalue. For the problems we will consider, we need consider explicitly only two of these eigenstates, namely those two which have a transition frequency which is resonant with the frequencies of the electromagnetic field. Let us call these two orthonormal solutions of 2.2.1-5 $|a)$ and $|b)$ with energy eigenvalues

$$E_a = \hbar\omega_a > E_b = \hbar\omega_b$$

$|a)$ and $|b)$ will ordinarily both represent excited states of the atom and are assumed to have well-defined, opposite parity, i.e., one is an even and the other is an odd function. The time dependent eigenstates, which are solutions of the time dependent Schrodinger equation

$$i\hbar \frac{\partial\psi_{a,b}}{\partial t} = H_o\psi_{a,b} \qquad 2.2.1\text{-}6$$

are then

$$\psi_a = |a)\ e^{-i\omega_a t} \quad \text{and} \qquad 2.2.1\text{-}7a$$

$$\psi_b = |b)\ e^{-i\omega_b t} \qquad 2.2.1\text{-}7b$$

For our purposes, all other solutions of 2.2.1-5 are of importance only in establishing the unperturbed decay rates γ_a, γ_b of the states $|a)$ and $|b)$, such that for an atom in state $|a)$ at time $t = 0$, the probability of being in state $|a)$ at time $t > 0$ is $e^{-\gamma_a t}$. Ideally, these decay rates are determined by the "zero-point" interaction of the atom with electromagnetic fields (18), but for practical purposes the effective decay rates are also dependent on the atom's surroundings, for example on pressure and radiation trapping effects (2). The decay rates are here introduced phenomenologically into the atomic equations of motion, in order to account for dissipation effects.

It is well known that the effects of the term in H' proportional to A^2 are very small compared to the effects due to the term proportional to A. To see this, let us consider matrix elements of these terms between the states $|a)$ and $|b)$. In order to deal easily with the first term, we need to express the operator $\underline{p}$ differently. Using the operator commutation relation $[r_i, p_j] \equiv r_i p_j - p_j r_i = i\hbar\delta_{ij}$, we easily show that (assuming $\underline{r}$ commutes with V)

$$\underline{p} = \frac{m}{i\hbar}[\underline{r}, H_o] \quad . \tag{2.2.1-8}$$

Thus we have

$$\frac{e\underline{p} \cdot \underline{A}}{m} = \frac{e\underline{A}}{i\hbar} \cdot [\underline{r}, H_o] \quad . \tag{2.2.1-9}$$

Since $|a)$ and $|b)$ are assumed to have opposite parity, only off-diagonal matrix elements of 2.2.1-9 exist, and we have

$$\begin{aligned}(a| \frac{e\underline{p} \cdot \underline{A}}{m} |b) &= (a| \frac{e\underline{A}}{i\hbar} \cdot (\underline{r}\, H_o - H_o \underline{r})|b) \\ &= ie\omega_o \underline{A} \cdot \underline{r}_{ab} = -i\omega_o \underline{A} \cdot \underline{P}_o \quad ,\end{aligned} \tag{2.2.1-10}$$

where $\omega_o \equiv \omega_a - \omega_b$ and $\underline{P}_o \equiv -e(a|\underline{r}|b)$ is the matrix element of the dipole moment operator between the states $|a)$ and $|b)$, and is assumed for convenience to be real. For an electric field $\underline{E} = -\partial A/\partial t$ sinusoidally varying with frequency ω_o and of magnitude 10^2 v/m , and assuming $P_o - 10^{-29}$ MKS , we have

$$|(a| \frac{e\underline{p} \cdot \underline{A}}{m} |b)| \simeq 10^{-27} \text{ joule.}$$

The term $e^2A^2/2m$ clearly has only diagonal matrix elements,

$$(a| \frac{e^2A^2}{2m} |a) = \frac{e^2A^2}{2m} = \frac{e^2E^2}{2m\omega_o^2} \simeq 10^{-34} \text{ joule}$$

for the above field at a frequency $\omega_o = 10^{15}$, corresponding to about 1 ev photons. Thus the matrix elements, and therefore the effects, of the quadratic term are negligible compared to those of the linear term, even for fields several orders of magntidue larger than 10^2 v/m. In the following we will see that fields of the order of magnitude 10^2 v/m can cause strong saturation effects in many systems. Thus we need not consider the A^2 term further, and we can redefine

$$H' \equiv \frac{e\underline{p} \cdot \underline{A}}{m} = \frac{e\underline{A}}{i\hbar} \cdot [\underline{r}, H_o] \quad , \qquad 2.2.1\text{-}11$$

with

$$H'_{ab} \equiv (a|H'|b) = -i\omega_o \underline{A} \cdot \underline{P}_o \quad . \qquad 2.2.1\text{-}12$$

2.2.2 The Atomic Equations of Motion

We now proceed to derive the equations of motion for atoms subjected to an electric field. We need the solution of the time-dependent Schrodinger equation

$$i\hbar \frac{\partial\psi}{\partial t} = H\psi \quad . \qquad 2.2.2\text{-}1$$

We use the technique of time-dependent perturbation theory (19), and expand ψ as

$$\psi = A(t)\,\psi_a + B(t)\,\psi_b \quad , \qquad 2.2.2\text{-}2$$

where ψ_a and ψ_b are given by equations 2.2.1-7. As mentioned previously, 2.2.2-2 is not a complete expansion, and $|A|^2 + |B|^2$ may be less than unity; however, the only importance of other states for this problem will be included by the phenomenological introduction below of effective decay rates γ_a and γ_b .

Substituting 2.2.2-2 into 2.2.2-1, we have

$$\dot{A}|a)\, e^{-i\omega_a t} + \dot{B}|b)\, e^{-i\omega_b t} = \frac{AH'}{i\hbar}|a)\, e^{-i\omega_a t} + \frac{BH'}{i\hbar}|b)\, e^{-i\omega_b t} \quad . \qquad 2.2.2\text{-}3$$

Forming matrix elements from the left with $|a)$ and $|b)$, we find successively

$$\dot{A}\, e^{-i\omega_a t} = \frac{B}{i\hbar} H'_{ab}\, e^{-i\omega_b t} \quad \text{and} \qquad 2.2.2\text{-}4$$

$$\dot{B}\, e^{-i\omega_b t} = \frac{A}{i\hbar} H'^{*}_{ab}\, e^{-i\omega_a t} \quad . \qquad 2.2.2\text{-}5$$

In order to explicitly introduce the decay rates it is conveneient to define new variables a and b by

$$a = A\, e^{-i\omega_a t} \;, \quad b = B\, e^{-i\omega_b t} \;, \qquad 2.2.2\text{-}6$$

so that

$$\psi = a|a) + b|b) \quad . \qquad 2.2.2\text{-}7$$

With this transformation, equations 2.2.2-4 and 5 become

$$\dot{a} = -i\omega_a a + \frac{bH'_{ab}}{i\hbar} \quad \text{and} \qquad 2.2.2\text{-}8$$

$$\dot{b} = -i\omega_b b + \frac{aH'^{*}_{ab}}{i\hbar} \quad . \qquad 2.2.2\text{-}9$$

Decay rates are now explicitly introduced by having the frequencies ω_a and ω_b of the eigenstates $|a)$ and $|b)$ become complex:

$$\omega_{a,b} \to \omega_{a,b} - i\,\frac{\gamma_{a,b}}{2} \quad . \qquad 2.2.2\text{-}10$$

Equations 2.2.2-8 and 9 now become

$$\dot{a} = -i\omega_a a + \frac{bH'_{ab}}{i\hbar} - \frac{\gamma_a}{2}\,a \quad \text{and} \qquad 2.2.2\text{-}11$$

$$\dot{b} = -i\omega_b b + \frac{aH'^{*}_{ab}}{i\hbar} - \frac{\gamma_b}{2}\,b \quad , \qquad 2.2.2\text{-}12$$

which give the proper decay rates for the unperturbed atomic states. It should be noted that we have not included the natural decay from $|a)$ to $|b)$. Thus we neglect any processes which arise due to these transitions. In a careful study of these processes, Buczek (20) has shown that this is valid provided $\gamma_b \gg \gamma_a$. Thus our results will hold strictly only for this case, which is also one of the requirements for a large population inversion between laser states and thus of much practical interest.

The equations of motion for the electric field, derived from Maxwell's equations in section 2.3, require the macroscopic polarization produced by the atoms in response to the incident field. For this quantity, we take the sum over all atoms per unit volume of the microscopic dipole moment, defined as the expected value of the dipole moment operator, $-e\underline{r}$, in the state ψ :

$$\underline{P} = (\psi| \; -e\underline{r} \; |\psi) \quad . \qquad 2.2.2\text{-}13$$

Substituting from equation 2.2.2-7, we find

$$\underline{P} = -e\underline{r}_{ab}\,(a^*b + b^*a) = \underline{P}_o(a^*b + ab^*) \quad , \qquad 2.2.2\text{-}14$$

where the orthonormality of the states $|a)$ and $|b)$ has been used. Thus the quantity required from a solution of the atomic equations of motion is the quadratic combination ab^* , and we are led to consider the equations of motion for the quadratic combinations

$$\begin{aligned} ab^* &\equiv \rho_{ab} \\ aa^* &\equiv \rho_{aa} \\ bb^* &\equiv \rho_{bb} \quad . \end{aligned} \qquad 2.2.2\text{-}15$$

Using equations 2.2.2-10 and 11, we have

$$\begin{aligned} \dot{\rho}_{ab} &= \dot{a}b^* + a\dot{b}^* \\ &= -(\gamma + i\omega_o)\,\rho_{ab} - (\rho_{aa} - \rho_{bb})\,\frac{H'_{ab}}{i\hbar} \quad , \end{aligned} \qquad 2.2.2\text{-}16$$

where we have used the definitions 2.2.2-15 and defined

$$\gamma \equiv \frac{\gamma_a + \gamma_b}{2} \quad . \qquad 2.2.2\text{-}17$$

Similarly we find

$$\dot{\rho}_{aa} = -\gamma_a\rho_{aa} + \frac{\rho^*_{ab}H'_{ab}}{i\hbar} - \frac{\rho_{ab}H'^*_{ab}}{i\hbar} \quad , \qquad 2.2.2\text{-}18$$

and

$$\dot{\rho}_{bb} = -\gamma_b\rho_{bb} - \frac{\rho^*_{ab}H'_{ab}}{i\hbar} - \frac{\rho_{ab}H'^*_{ab}}{i\hbar} \quad . \qquad 2.2.2\text{-}19$$

Finally, using 2.2.1-12, the atomic equations of motion become

$$\mathring{\rho}_{ab} = -(\gamma + i\omega_o)\, \rho_{ab} + (\rho_{aa} - \rho_{bb})\, \frac{\omega_o \underline{A} \cdot \underline{P}_o}{\hbar} \qquad 2.2.2\text{-}20$$

$$\mathring{\rho}_{aa} = -\gamma_a \rho_{aa} - (\rho_{ab} + \rho^*_{ab})\, \frac{\omega_o \underline{A} \cdot \underline{P}_o}{\hbar} \qquad 2.2.2\text{-}21$$

$$\mathring{\rho}_{bb} = -\gamma_b \rho_{bb} + (\rho_{ab} + \rho^*_{ab})\, \frac{\omega_o \underline{A} \cdot \underline{P}_o}{\hbar} \qquad 2.2.2\text{-}22$$

Different solutions of these equations will be considered in Chapter 3, after the equations of motion for the electric field have been derived in the next section.

2.3 Equations of Motion for the Electromagnetic Field

2.3.1 Introduction, Description of the Field

In this section we derive the equations of motion for the electromagnetic field, using Maxwell's equations and assuming modes varying harmonically in time and space with a "slow" spatial variation of amplitude. We consider first a "dilute" medium, in the sense that the deviation of the index of refraction from unity is small. Media where this is not true will then be considered briefly.

For considering the behavior of waves in a dispersive medium, we need to use characteristic modes. For traveling waves, this of course means a Fourier decomposition of the field, which has to be put in a form suitable for considering nonlinear effects. If we define the incident field as that field which would be present in the absence of any material medium, the incident field is very generally expressed as a four-dimensional Fourier transform:

$$\underline{E}(\underline{r},t) = \int d^3k \int_{-\infty}^{\infty} d\omega \; \underline{E}(\underline{k},\omega) \; e^{i(\underline{k} \cdot \underline{r} - \omega t)} \quad , \qquad 2.3.1\text{-}1$$

where

$$\underline{E}(\underline{k},\omega) = \frac{1}{(2\pi)^4} \int d^3r \int_{-\infty}^{\infty} dt \; \underline{E}(\underline{r},t) \; e^{-i(\underline{k} \cdot \underline{r} - \omega t)} \qquad 2.3.1\text{-}2$$

The integrals are assumed to exist, including the possibility of generalized functions (21), especially the Dirac delta function. The field must obey some equations of motion, in our case these are the coupled Maxwell's equations and equations of motion for the atoms. In any case, these equations furnish a "dispersion relation" i.e., a relation between $\underline{k}$ and ω, and then the integral 2.3.1-1 becomes an integral over ω or over $\underline{k}$ space, depending on the order in which the integrals are taken. As written in 2.3.1-1, the ω integral is first and the dispersion relation is written as

$$\omega = \omega(\underline{k}) \quad , \qquad 2.3.1\text{-}3$$

which we assume to be a single-valued function, so that the ω integral just results in replacing ω by $\omega(\underline{k})$. For our purposes, however, we would like to invert the order of integration from that in 2.3.1-1, and write the dispersion relation as

$$\underline{k} = \underline{k}(\omega) \quad , \qquad 2.3.1\text{-}4$$

leaving an integral over modes with well-defined frequency. The disadvantage of course is that the relation 2.3.1-4 is in general not single-valued. As an example, for free space we have

$$\omega = \omega(\underline{k}) = kc \quad , \qquad 2.3.1\text{-}5$$

where c is the free space velocity of light, so that for a given ω, only the magnitude of $\underline{k}$ is fixed. For this approach, we might express the field in the form

$$\underline{E}(\underline{r},t) = \int_{-\infty}^{\infty} d\omega \, \underline{E}(\omega) \, e^{i(\underline{k} \cdot \underline{r} - \omega t)} \quad , \qquad 2.3.1\text{-}6$$

where $\underline{k} = \underline{k}(\omega)$ and we assume that the $\underline{k}$ integral over all $\underline{k}$'s associated with the given ω has been carried out.

There are two difficulties with this way of expressing the field, when we wish to study nonlinear effects. First, we need to express the field in terms of real modes; and second, this form is at the very least difficult to work with when superposition fails with the appearance of nonlinearities. The effect of superposition is that any combination of input frequencies results in the same output frequencies, and there is no interaction between fields at different frequencies. Nonlinearities produce some transformation of the input spectrum into an output spectrum which in general is not one to one. In addition, it will become evident later that the medium is in a sense made anisotropic by the nonlinearity. For example, the introduction of a field at a third frequency due to the nonlinear interaction of fields at two other frequencies depends on the directions of propagation of the two original fields. Rather than attempt any general discussion of such effects, we will usually assume that the incident field is composed of a number of plane waves, and treat each case explicitly. With this assumption, 2.3.1-6 becomes

$$\underline{E}(\underline{r},t) = \sum_{\omega} \underline{E}_{\omega}\, e^{i(\underline{k} \cdot \underline{r} - \omega t)} \quad . \qquad 2.3.1\text{-}7$$

To write this incident field in terms of real modes, we use the fact that $\underline{E}(\underline{r},t)$ is real to write

$$\underline{E}(\underline{r},t) = \frac{1}{2} \sum_{\omega} \left[\underline{E}_{\omega}\, e^{i(\underline{k} \cdot \underline{r} - \omega t)} + \underline{E}^{*}_{\omega}\, e^{-i(\underline{k} \cdot \underline{r} - \omega t)} \right] \qquad 2.3.1\text{-}7$$

If we write $\underline{E}_{\omega} = \underline{e}_{\omega}\, E_{\omega} = \underline{e}_{\omega}\, |E_{\omega}|\, e^{i\varphi_{\omega}}$, 2.3.1-7 becomes

$$\underline{E}(\underline{r},t) = \sum_{\omega} \underline{e}_{\omega}\, |E_{\omega}| \cos(\underline{k} \cdot \underline{r} - \omega t + \varphi_{\omega}) \quad . \qquad 2.3.1\text{-}8$$

We can generalize 2.3.1-8 slightly by redefining

$$E_{\omega} \equiv |E_{\omega}|,\ \varphi_{\omega} \equiv \varphi_{\omega} \qquad \text{if } \varphi < \pi \quad ,$$

$$E_{\omega} \equiv -|E_{\omega}|,\ \varphi_{\omega} \equiv \varphi_{\omega} - \pi \text{ if } \varphi \geq \pi \qquad 2.3.1\text{-}9$$

$$\underline{E}_{\omega} \equiv \underline{e}_{\omega}\, E_{\omega} \, ,$$

so that we can have positive or negative fields, with $0 \leq \varphi < \pi$. With these definitions the incident field becomes

$$\underline{E}(\underline{r},t) = \sum_{\omega} \underline{E}_{\omega} \cos(\underline{k} \cdot \underline{r} - \omega t + \varphi_{\omega}) \quad , \qquad 2.3.1\text{-}10$$

where $\underline{k} = \underline{k}(\omega)$, and in general two polarizations and more than one $\underline{k}$ may be present for a given ω .

The form 2.3.1-10 for the incident field could have been written down immediately, as is usually done. The purpose of the above discussion is to clarify the difficulties encountered when dealing with more general incident fields.

2.3.2 The Field Equations of Motion for a Dilute Medium

We wish to find the equations obeyed by a field (in the medium) which we assume to have the form

$$\underline{E}(\underline{r},t) = \sum_{\omega} \underline{E}_{\omega}(\underline{r}) \cos(\underline{k} \cdot \underline{r} - \omega t + \varphi_{\omega}) \quad , \qquad 2.3.2\text{-}1$$

where, in general

$$\underline{k} = \underline{k}(\omega,\underline{r}) \quad \text{and} \quad \varphi_{\omega} = \varphi_{\omega}(\underline{r}) \quad . \qquad 2.3.2\text{-}2$$

Assuming the medium to be nonmagnetic and to have no free charge, we can write Maxwell's equations in the form

$$\nabla \times \underline{E}(\underline{r},t) = - \frac{\partial \underline{B}(\underline{r},t)}{\partial t} \qquad 2.3.2\text{-}3$$

$$\epsilon_o c^2 \nabla \times \underline{B}(\underline{r},t) = \frac{\partial \underline{D}(\underline{r},t)}{\partial t} = \epsilon_o \frac{\partial \underline{E}(\underline{r},t)}{\partial t} + \frac{\partial \underline{P}(\underline{r},t)}{\partial t} \qquad 2.3.2\text{-}4$$

$$\nabla \cdot \underline{E}(\underline{r},t) = - \frac{1}{\epsilon_o} \nabla \cdot \underline{P}(\underline{r},t) \qquad 2.3.2\text{-}5$$

$$\nabla \cdot \underline{B}(\underline{r},t) = 0 \quad . \qquad 2.3.2\text{-}6$$

Taking the curl of 2.3.2-3 and substituting in 2.3.2-4, we have

$$\nabla \times \nabla \times \underline{E}(\underline{r},t) = - \frac{1}{c^2} \frac{\partial^2 \underline{E}(\underline{r},t)}{\partial t^2} - \frac{1}{\epsilon_o c^2} \frac{\partial^2 \underline{P}(\underline{r},t)}{\partial t^2} \quad . \qquad 2.3.2\text{-}7$$

Upon using the vector identity

$$\nabla \times \nabla \times \underline{E} = - \nabla^2 \underline{E} + \nabla(\nabla \cdot \underline{E}) \qquad 2.3.2\text{-}8$$

and 2.3.2-5, 2.3.2-7 becomes

$$-\nabla^2 \underline{E}(\underline{r},t) + \frac{1}{c^2} \frac{\partial^2 \underline{E}(\underline{r},t)}{\partial t^2} = \frac{1}{\epsilon^o} \nabla(\nabla \cdot \underline{P}(\underline{r},t) - \frac{1}{\epsilon_o c^2} \frac{\partial^2 \underline{P}(\underline{r},t)}{\partial t^2} \qquad 2.3.2\text{-}9$$

Using the form 2.3.2-1 for the field, we find

$$\nabla^2 E_j(\underline{r},t) = \sum_{\omega}\left\{\left[\nabla^2 E_{\omega j} - (\underline{k} + \nabla\varphi_{\omega})^2 E_{\omega j}\right] \cos(\underline{k} \cdot \underline{r} - \omega t + \varphi_{\omega}) + \left[- 2\nabla E_{\omega j} \cdot (\underline{k} + \nabla\varphi_{\omega})\right] \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_{\omega})\right\} ,$$

2.3.2-10

where $j = x,y,z$. Space derivatives of $\underline{k}$ have been neglected as being small. This assumption is justified below. We also use the assumption of slow spatial variation to neglect the terms

$$\nabla^2 E_{\omega j} , (\nabla\varphi_{\omega})^2, \nabla E_{\omega j} \cdot \nabla\varphi_{\omega}, \nabla^2\varphi_{\omega}$$

in 2.3.2-10. For this to be valid, we must have, for example,

$$\nabla^2 E_{\omega j} \ll \underline{k} \cdot \nabla E_{\omega j} = k\, \underline{e}_{\omega} \cdot \nabla E_{\omega j} \quad . \qquad 2.3.2\text{-}11$$

We will show later that for a plane wave propagating in the z direction in a linear medium, $E_{\omega x}(z) = E_{\omega x}(o)\, e^{\alpha z}$. Using $k \cong \frac{\omega}{c}$, 2.3.2-11 becomes

$$\alpha \ll \frac{\omega}{c} \approx 10^6 \text{ meter}^{-1} \qquad 2.3.2\text{-}12$$

for optical frequencies, which is very strongly satisfied by observed α's , which are of order unity. Thus to a very good approximation we can neglect higher order spatial derivatives.

Using the form

$$\underline{P}(\underline{r},t) = \sum_{\omega}\left[\underline{P}_{\omega c}(\underline{r}) \cos(\underline{k} \cdot \underline{r} - \omega t + \varphi_{\omega}) + \underline{P}_{\omega s}(\underline{r}) \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_{\omega})\right]$$

2.3.2-13

for the polarization and carrying out the indicated derivatives, we see that the lowest order terms in $\frac{1}{\epsilon_o} \nabla(\nabla \cdot \underline{P})$ are terms like

$$-\frac{1}{\epsilon_o}\left\{\left[\underline{k}(\nabla \cdot \underline{P}_{\omega c})\right] \cos(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega) + \left[(\underline{k} \cdot \nabla)\underline{P}_{\omega c}\right] \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega)\right.$$
$$\left. + \left[\underline{k} \times (\nabla \times \underline{P}_{\omega c})\right] \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega) + \underline{k}(\underline{k} \cdot \underline{P}_{\omega c}) \cos(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega)\right\}$$
2.3.2-14

Since for transverse plane waves $\underline{k} \cdot \underline{P} = 0$ to at least first order, all of these terms are of higher than first order and one thus neglected. With these approximations, we can write 2.3.2-9 in the form

$$\sum_\omega \left[(k^2 - \frac{\omega^2}{c^2} + 2\underline{k} \cdot \nabla\varphi_\omega) E_{\omega j} \cos(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega) \right.$$
$$+ 2\nabla E_{\omega j} \cdot \underline{k} \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega)$$
$$\left. - \frac{\omega^2}{\epsilon_o c^2} (P_{\omega cj} \cos(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega) + P_{\omega sj} \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega)) \right] = 0 \quad ,$$
2.3.2-15

for $j = x,y,z$.

For 2.3.2-15 to be satisfied in general, we must have the coefficients of the sine and cosine functions separately equal to zero, for each ω. Thus we have

$$\left[k^2 - \frac{\omega^2}{c^2} + 2\underline{k} \cdot \nabla\varphi_\omega\right] E_{\omega j} = \frac{\omega^2}{\epsilon_o c^2} P_{\omega cj} \qquad 2.3.2\text{-}16$$

and

$$\underline{k} \cdot \nabla E_{\omega j} = \frac{\omega^2}{2\epsilon_o c^2} P_{\omega sj} \quad , \qquad 2.3.2\text{-}17$$

for $j = x,y,z$. In free space, 2.3.2-16 gives $k = \frac{\omega}{c}$, so that $\underline{k}$ can be dependent on space only in a first order correction term, and the neglect of space derivatives of $\underline{k}$ in obtaining 2.3.2-10 is justified. Since 2.3.2-16 and 17 are meant to include only first order terms, we should replace $\underline{k}$ by $\frac{\omega}{c} \underline{e}_{\omega}$ and

$$k^2 - \frac{\omega^2}{c^2} = (k + \frac{\omega}{c})\ (k - \frac{\omega}{c})$$

by $2\frac{\omega}{c}(k - \frac{\omega}{c})$. Defining an index of refraction by

$$k(\omega) = n(\omega)\,\frac{\omega}{c} \quad , \qquad 2.3.2\text{-}18$$

we have

$$k^2 - \frac{\omega^2}{c^2} = \frac{2\omega^2}{c^2}\,[n(\omega) - 1] \qquad 2.3.2\text{-}19$$

to first order.

Then 2.3.2-16 and 17 become

$$[n(\omega) - 1 + \frac{c}{\omega}\,\underline{e}_{\omega} \cdot \nabla\varphi_{\omega}]\ E_{\omega j} = \frac{1}{2\epsilon_o}\,P_{\omega cj} \qquad 2.3.2\text{-}20$$

and

$$\underline{e}_{\omega} \cdot \nabla E_{\omega j} = \frac{\omega}{2\epsilon_o c}\,P_{\omega sj} \quad , \qquad 2.3.2\text{-}21$$

for $j = x,y,z$. These are the final equations of motion for the field 2.3.2-1 in the dilute medium, and along with the atomic equations of motion 2.2.2-20 through 22 form the basis of our treatment of the field-matter interaction.

Equations 2.3.2-20 and 21 show the well known result that an in-phase component of the induced polarization reacts on the field to change its propagation constant (phase), while an out-of-phase component of the polarization changes the amplitude of the wave. If there is no induced polarization, we of course find that the amplitude of the wave is constant, and its propagation vector has the magnitude $\frac{\omega}{c}$.

The use of the external field in the atomic equations of motion and the assumption of slow variation require that $|n(\omega) - 1| \ll 1$, i.e., that we have a "dilute" medium. In section 2.3.4 this restriction is modified to allow a linear interaction with the medium in addition to the effects of the transition under consideration.

2.3.3 On the Difference Between Traveling Wave and Cavity Modes

It is interesting to derive 2.3.2-20 and 21 by assuming modes whose amplitudes are varying slowly in time (compared to the frequencies ω), i.e., a field in the medium of the form

$$\underline{E}(\underline{r},t) = \sum_{\omega} \underline{E}_{\omega}(t) \cos(\underline{k} \cdot \underline{r} - \omega t + \varphi_{\omega}) \quad , \qquad 2.3.3\text{-}1$$

where in general $\underline{k} = \underline{k}(\omega,t)$ and $\varphi_{\omega} = \varphi_{\omega}(t)$. The polarization is given by a similar expression with sine as well as cosine terms. Approximations exactly similar to those made in deriving equations 2.3.2-20 and 21 are made, and 2.3.2-9 leads to the equations of motion

$$\left[n(\omega) - 1 + \frac{1}{\omega}\frac{\partial\varphi}{\partial t}\right] E_{\omega j} = \frac{1}{2\epsilon_o} P_{\omega c j} \qquad 2.3.3\text{-}2$$

and

$$\frac{\partial E_{\omega j}}{\partial t} = \frac{\omega}{2\epsilon_o} P_{\omega s j} \quad . \qquad 2.3.3\text{-}3$$

We can get space-dependent mode amplitudes by using the fact that t and z/c are basically equivalent for a plane wave propagating in the z direction, i.e., by making the change

$$\frac{\partial}{\partial t} \rightarrow c \frac{\partial}{\partial z} \qquad 2.3.3\text{-}4$$

in the equations of motion 2.3.3-2 and 3. We could alternatively substitute z/c for t in the solutions for the mode amplitudes as a function of time. If the change 2.3.3-4 is made in the equations of motion, the latter become

$$\left[n(\omega) - 1 + \frac{c}{\omega} \frac{\partial \varphi}{\partial z} \right] E_{\omega j} = \frac{1}{2\epsilon_o} P_{\omega c j} \qquad 2.3.3\text{-}5$$

and

$$\frac{\partial E_{\omega j}}{\partial z} = \frac{\omega}{2\epsilon_o c} P_{\omega s j} \quad . \qquad 2.3.3\text{-}6$$

We can allow for more general directions of propagation by noting that $\partial/\partial z$ is the gradient in the direction of propagation. In general, then, we should replace $\partial/\partial z$ in 2.3.3-5 and 6 by $\underline{e}_{\omega} \cdot \nabla$, which gives the previous equations of motion 2.3.2-20 and 21.

The assumption of modes whose amplitudes depend on time is similar to treating the wave interactions as a cavity problem, where the "cavity" is all of space. However, in a true cavity the magnitude of $\underline{k}$ is fixed by boundary conditions and the effect of a phase shift due to the medium appears as a change in the eigenfrequency of the cavity. Also, the cavity modes are standing waves rather than traveling waves. It might appear possible to construct proper cavity modes by using a superposition of two of the modes 2.3.3-1 with oppositely-directed $\underline{k}$'s to give a standing wave, and changing ω so that $\frac{\omega}{c}n(\omega)$ equals the value of $\underline{k}$ required by boundary conditions*. Lamb (12) has indicated that a difficulty of this approach is that the nonlinearities associated with a standing wave mode are in general different from those for a traveling wave mode, and that the behavior of a laser oscillator should therefore be studied using cavity modes. Although it is certainly true that the nonlinear interactions are in general different for a standing wave mode and a traveling wave mode, it is also true that they are different for one traveling wave mode and two traveling wave modes, as indicated in section 2.3.1. The following theory should show that the behavior of any field which can be formed as a superposition of traveling wave modes can be correctly studied using those modes. It follows that cavity modes (at least those expressable as a superposition of the traveling wave modes of 2.3.3-1) can in principle be studied using traveling wave modes, but the reverse is not true. In this sense the traveling wave mode approach is more fundamental.

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* This is essentially the technique used by Bennett to study hole burning effects in a laser oscillator (15), except his mode amplitudes were space dependent rather than time dependent.

2.3.4 The Field Equations of Motion for Interaction with a "Linear" Medium

In this section we derive the equations obeyed by the field in a medium whose polarization can be divided into two parts: a part linear in the field and a part due to the transition of interest, containing in general linear and nonlinear contributions, and assumed small. For such a medium, the displacement vector may be written as

$$\underline{D}(\underline{r},t) = \epsilon_o\, \underline{E}(\underline{r},t) + \underline{P}_1(\underline{r},t) + \underline{P}(\underline{r},t) \quad , \qquad 2.3.4\text{-}1$$

where $\underline{P}_1(\underline{r},t)$ is the linear polarization. In the absence of $\underline{P}$, $\underline{P}_1$ gives rise to an effective ϵ for the medium. For a dispersive medium we can write

$$\underline{D}(\underline{r},t) = \epsilon_o\, \underline{E}(\underline{r},t) + \underline{P}_1(\underline{r},t) = \int_0^\infty \epsilon(\tau)\, \underline{E}(\underline{r},t-\tau)d\tau \qquad 2.3.4\text{-}1$$

where we have assumed an isotropic, homogeneous medium. In the presence of $\underline{P}_1$, the Maxwell's equation 2.3.2-4 thus becomes

$$\epsilon_o c^2 \nabla \times \underline{B}(\underline{r},t) = \frac{\partial}{\partial t}\int_0^\infty \epsilon(\tau)\, \underline{E}(\underline{r},t-\tau)dt + \frac{\partial \underline{P}(\underline{r},t)}{\partial t} \quad . \qquad 2.3.4\text{-}2$$

Introducing the forms 2.3.2-1 for the field and 2.3.2-13 for $\underline{P}$, we proceed as before to find

$$\sum_\omega \Big[(k^2 + 2\underline{k}\cdot\nabla\varphi_\omega)\, E_{\omega j} \cos(\underline{k}\cdot\underline{r} - \omega t + \varphi_\omega)$$

$$+ 2\underline{k}\cdot\nabla E_{\omega j} \sin(\underline{k}\cdot\underline{r} - \omega t + \varphi_\omega)\Big]$$

$$= \sum_{\omega} \frac{1}{\epsilon_o c^2} \left\{ - \frac{\partial^2}{\partial t^2} \int_0^\infty \epsilon(\tau)\, \underline{E}_\omega \cos(\underline{k} \cdot \underline{r} - \omega(t - \tau) + \varphi_\omega) d\tau \right.$$

$$\left. + \omega^2 \left[\underline{P}_{\omega cj} \cos(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega) + \underline{P}_{\omega sj} \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega) \right] \right\} .$$

2.3.4-3

Taking the time derivatives inside the integral, the first term in the curly brackets becomes

$$\omega^2 \int_0^\infty \epsilon(\tau)\, \underline{E}_\omega \cos(\underline{k} \cdot \underline{r} - \omega(t - \tau) + \varphi_\omega) d\tau$$

$$= \omega^2 \epsilon'(\omega)\, \underline{E}_\omega \cos(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega) \qquad 2.3.4\text{-}4$$

$$+ \omega^2 \epsilon''(\omega)\, \underline{E}_\omega \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega) \quad ,$$

where we have expanded the cosine in the integral and defined

$$\epsilon'(\omega) = \int_0^\infty \epsilon(\tau) \cos \omega\tau \; d\tau$$

$$\epsilon''(\omega) = \int_0^\infty \epsilon(\tau) \sin \omega\tau \; d\tau \qquad 2.3.4\text{-}5$$

Thus we obtain the equations of motion as

$$\left(k^2 - \frac{\omega^2 \epsilon'(\omega)}{\epsilon_o c^2} + 2\underline{k} \cdot \nabla\varphi_\omega \right) E_{\omega j} = \frac{\omega^2}{\epsilon_o c^2} P_{\omega cj} \qquad 2.3.4\text{-}6$$

$$2\underline{k} \cdot \nabla E_{\omega j} = \frac{\omega^2 \epsilon''(\omega)}{\epsilon_o c^2} E_{\omega j} + \frac{\omega^2}{\epsilon_o c^2} P_{\omega s j} \quad . \qquad 2.3.4\text{-}7$$

For $P_{\omega c j} = P_{\omega s j} = 0$ we find

$$k^2 = \frac{\omega^2 \epsilon'(\omega)}{\epsilon_o c^2} \equiv \frac{\omega^2 n_o^2(\omega)}{c^2} \quad . \qquad 2.3.4\text{-}8$$

For $P_{\omega c j}, P_{\omega s j} \neq 0$, we define the index of refraction by

$$k^2 = \frac{n^2(\omega)\omega^2}{c^2} \qquad 2.3.4\text{-}9$$

and write $n(\omega) = n_o(\omega)u(\omega)$. Assuming $|u(\omega) - 1| \ll 1$, we proceed as before and find the final equations of motion

$$\left(u - 1 + \frac{c}{n_o(\omega)\omega} \underline{e}_\omega \cdot \nabla \varphi_\omega\right) E_{\omega j} = \frac{1}{2\epsilon_o n_o^2(\omega)} P_{\omega c j} \qquad 2.3.4\text{-}10$$

$$\underline{e}_\omega \cdot \nabla E_{\omega j} = \frac{\omega \epsilon''(\omega)}{2\epsilon_o c n_o(\omega)} E_{\omega j} + \frac{\omega}{2n_o(\omega)\epsilon_o c} P_{\omega s j} \quad . \qquad 2.3.4\text{-}11$$

If we define $L(\omega)$ by

$$\frac{-\omega \epsilon''(\omega)}{\epsilon_o c n_o(\omega)} = \frac{1}{L(\omega)} \quad , \qquad 2.3.4\text{-}12$$

2.3.4-11 becomes

$$\underline{e}_\omega \cdot \nabla E_{\omega j} = -\frac{E_{\omega j}}{2L(\omega)} + \frac{\omega}{2n_o(\omega)\epsilon_o c} P_{\omega s j} \quad . \qquad 2.3.4\text{-}13$$

For the time varying modes of section 2.3.3, the corresponding equations are

$$\left(u - 1 + \frac{1}{n_o(\omega)\omega}\frac{\partial\varphi_\omega}{\partial t}\right)E_{\omega j} = \frac{1}{2\epsilon_o n_o^2(\omega)} P_{\omega cj} \qquad 2.3.4\text{-}14$$

$$\frac{\partial E_{\omega j}}{\partial t} = -\frac{\omega E_{\omega j}}{2Q_m(\omega)} + \frac{\omega}{2n_o(\omega)\epsilon_o} P_{\omega sj} \quad , \qquad 2.3.4\text{-}15$$

where the "material Q" is defined as

$$Q_m(\omega) = -\frac{\epsilon_o n_o(\omega)}{\epsilon''(\omega)} \quad . \qquad 2.3.4\text{-}16$$

If cavity losses need to be included the "cavity Q", Q_c , may be introduced, and $Q_m(\omega)$ in 2.3.4-15 replaced by the total Q, Q_T, where

$$\frac{1}{Q_T} = \frac{1}{Q_m(\omega)} + \frac{1}{Q_c} \quad . \qquad 2.3.4\text{-}17$$

In the following chapters, we will neglect any effects of reflection at the boundaries of finite media, although these can be important if n_o is appreciably different from unity. We will instead be concerned with the interaction of the fields with the medium and with each other via the medium. If necessary, the effects of boundary conditions can be included by using the techniques of Bloembergen and Pershan (22). The assumption that the single transition of interest causes only a small change in the linear index of refraction $n_o(\omega)$ could be removed by using the more general techniques indicated by Armstrong, et al (4), but this will not be attempted in the following.

The effects of $n_o(\omega)$ and $L(\omega)$ are to give stronger phase matching conditions and thresholds for various processes to be observed, respectively. These effects do not change the nature of the processes involved. Since we are primarily interested in gaseous media where the effects are small, they will generally be ignored. For other media, they should be included, and this can be done quite easily.

CHAPTER THREE

SOLUTIONS OF THE ATOMIC EQUATIONS OF MOTION

In this chapter we discuss how the equations of motion are solved for studying the behavior of fields in a CW amplifying medium. Excitation of the medium is introduced, and the effects of atomic motion are studied. The treatment generally follows that of Lamb (11,12), adapted for fields composed of vector traveling waves. The basic assumption is that the fields perturbing the atom remain effectively constant in amplitude during the time required for the atom to decay to lower levels. This means that these field amplitudes must change slowly compared to the decay rates γ_a, γ_b, a requirement easily met in practice.

In section 3.1 the atomic equations of motion are expressed in terms of the field 2.3.2-1, and the field free solution for a single atom is shown. Excitation of the medium and steady-state solutions are considered in section 3.2, and the form of the solutions is given in section 3.3.

3.1 The Equations for a Single Atom

Corresponding to the form 2.3.2-1 for the electric field, we write the vector potential $\underline{A}(\underline{r},t)$ in the form

$$\underline{A}(\underline{r},t) = \sum_{\omega} \underline{A}_{\omega}(\underline{r}) \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_{\omega}) \quad . \qquad 3.1\text{-}1$$

Using $\underline{E} = - \partial \underline{A}/\partial t$, and comparing 3.1-1 to 2.3.2-1, we can write this as

$$\underline{A}(\underline{r},t) = \sum_{\omega} \frac{\underline{E}_{\omega}(\underline{r})}{\omega} \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_{\omega}) \quad . \qquad 3.1\text{-}2$$

Inserting this form for $\underline{A}$ into the equations of motion 2.2.2-20 to 22, the latter become

$$\dot{\rho}_{aa} = - \gamma_a \rho_{aa} - (\rho_{ab} + \rho^*_{ab}) \frac{\omega_o}{\hbar} \sum_{\omega} \frac{\underline{P}_o \cdot \underline{E}_{\omega}(\underline{r})}{\omega} \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_{\omega}) \qquad 3.1\text{-}3$$

$$\dot{\rho}_{bb} = - \gamma_b \rho_{bb} + (\rho_{ab} + \rho^*_{ab}) \frac{\omega_o}{\hbar} \sum_{\omega} \frac{\underline{P}_o \cdot \underline{E}_{\omega}(\underline{r})}{\omega} \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_{\omega}) \qquad 3.1\text{-}4$$

$$\dot{\rho}_{ab} = - (\gamma + i\omega_o)\, \rho_{ab} + (\rho_{aa} - \rho_{bb}) \frac{\omega_o}{\hbar} \sum_{\omega} \frac{\underline{P}_o \cdot \underline{E}_{\omega}(\underline{r})}{\omega} \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_{\omega}) \, . \qquad 3.1\text{-}5$$

The quantities ρ_{aa}, ρ_{bb}, and ρ_{ab} are functions of time and the atomic position $\underline{r}$, and for given fields the solutions are defined by the initial conditions.

We are interested primarily in gaseous media, and so must take into account the motion of the atoms. This can be done by noting that an atom with a velocity $\underline{v}$, initially at $\underline{r}_o$ at time t_o, has the position $\underline{r} = \underline{r}_o + \underline{v}(t - t_o)$ at time t. The period $(t - t_o)$ of interest is of order $1/\gamma_a$ or $1/\gamma_b$, which is rather short for states giving transitions at optical frequencies. This leads to two simplifying assumptions, viz. that the velocity $\underline{v}$ of the atom does not change during the time required for the atom to decay, and that the amplitude of the field perturbing the atom remains constant during this time. Thus we neglect the effects of collisions which change the velocity as well as those which perturb the states $|a)$ and $|b)$. The validity of these assumptions can be checked as follows: The time between collisions is given roughly by the ratio of the mean free path length to the average thermal velocity. For the He-Ne laser, these quantities are about 5×10^{-4}m and 6×10^{2} m/sec, respectively, making the ratio about 10^{-6} sec., compared to typical values for $1/\gamma_a$, $1/\gamma_b$ of smaller than about 10^{-7} sec. Thus the assumption of fixed velocity should be fairly good. The distance traveled by a moving excited atom before it decays is roughly the product of the average thermal velocity times $1/\gamma_a$, $1/\gamma_b$, or less than about 10^{-4} m. For a small signal gain of 80 db/meter, this corresponds to about a 0.1% increase in the field strength which can be justifiably neglected. For stationary atoms the above assumptions are of course superfluous for our formulation of the problem.

The simplest initial condition placed on a single atom is that it is either in the state $|a)$ or the state $|b)$ at time t_o, and no other possibilities will be considered in the following. For the initial conditions $\rho_{aa}(t_o) = 1$, $\rho_{bb}(t_o) = 1$ and with no fields present, 3.1-3 and 4 give the solutions for $t > t_o$

$$\rho_{aa} = e^{-\gamma_a(t - t_o)}, \quad \rho_{bb} = e^{-\gamma_b(t - t_o)},$$

respectively. For the simplest case of a single, monochromatic field and a stationary atom, equations 3.1-3 to 5 can be solved in the standard way (20) by neglecting the off-resonance exponential in the sine expansion, differentiating 3.1-3 and 4 again, using 3.1-5, and solving the resulting second order equations for ρ_{aa} and ρ_{bb} . The resulting solutions show the oscillation between the states $|a)$ and $|b)$, with

$$\gamma_b \int_{t_o}^{\infty} \rho_{bb}(t)\, dt$$

being the probability that a photon is emitted by the atom due to stimulated emission (15), if the initial condition is $\rho_{aa}(t_o) = 1$. In dealing with the coherent interactions of the fields and medium, this approach is not adequate. The present approach in terms of Maxwell's equations and the induced polarization is then a useful one, where we solve 2.4.1-3 to 5 for ρ_{ab} and thus the induced microscopic dipole moment

$$\underline{P} = P_o\ (\rho_{ab} + \rho^*_{ab}) \quad , \qquad 3.1\text{-}6$$

which, when summed over all atoms in a unit volume, is used with 2.3.2-20 and 21 (or more generally 2.3.4-10 and 13) to give the behavior of the fields. This process will be accomplished by summing the results over the excitation times of the atoms and using an assumption of quasi-steady state conditions. This approach will be outlined in the next two sections.

3.2 Excitation and the Quasi-Steady State Solution

For a given incident field, the motion of the atoms will reach a quasi-steady state if the excitation process is uniform. By a quasi-steady state we mean a state in which the populations of atoms in the various levels are changing slowly enough so that the quantity ρ_{ab} (and thus the polarization) is essentially a sum of harmonically varying components, in the same sense as the field. As was the case for the field, this is not a severe restriction at optical frequencies. In fact, it will be seen later that the effects of any such rapid population variations are strongly "quenched".

The excitation process is conveniently defined by the number of atoms excited to the states $|a)$ and $|b)$ per unit time, per unit volume, per unit velocity interval. This number is assumed to be a function only of velocity and not of position or time. We further assume that the velocity distribution of excited atoms is the same for both states $|a)$ and $|b)$. The excitation is thus defined by

$$\lambda_{a,b} \, W(\underline{v}) \quad , \qquad 3.2\text{-}1$$

where $\lambda_{a,b}$ is the total number of atoms excited to $|a)$, $|b)$ per sec. per m^3, and $W(\underline{v})$ is the velocity distribution function, normalized to unity by

$$\iiint_{-\infty}^{\infty} W(\underline{v}) \, dv_x dv_y dv_z = 1 \quad . \qquad 3.2\text{-}2$$

The quasi-steady-state solution at a time t is found by integrating over all previous excitation times the solutions due to individual excitations of the states $|a)$ and $|b)$. For example, if we denote the solution of 3.1-3 for atoms excited to state $|a)$ at time t_o and position $\underline{r}_o$ with velocity $\underline{v}$ as

$$\rho_{aa}^{(a)}(\underline{r},t,\underline{v},r_o,t_o) \quad , \tag{3.2-3}$$

then we can only have contributions to the quasi-steady state solution

$$\rho_{aa}(\underline{r},t,\underline{v}) \tag{3.2-4}$$

for $\underline{r}_o$ and $\underline{v}$ such that $\underline{r} = \underline{r}_o + \underline{v}(t - t_o)$. Thus the contribution of solutions 3.2-3 to 3.2-4 is given by the integral

$$\rho_{aa}^{(a)}(\underline{r},t,\underline{v}) = \lambda_a \, W(\underline{v}) \int_{-\infty}^{t} dt_o \; \rho_{aa}^{(a)}(\underline{r} = \underline{r}_o + \underline{v}(t - t_o),\ \underline{v},t,t_o) \quad , \tag{3.2-5}$$

where $\rho_{aa}^{(a)}(\underline{r},t,\underline{v})$ denotes the contribution to 3.2-4 of atoms excited to state $|a)$. This contribution has been obtained by summing over all atoms per unit volume at $\underline{r}_o$, giving the factor $\lambda_a \, W(\underline{v})$, and then integrating over the possible values of t_o which can contribute. Similarly, we find

$$\rho_{bb}^{(a)}(\underline{r},t,\underline{v}) = \lambda_a \, W(\underline{v}) \int_{-\infty}^{t} dt_o \; \rho_{bb}^{(a)}(\underline{r} = \underline{r}_o + \underline{v}(t - t_o),\ \underline{v},t,t_o) \quad , \tag{3.2-6}$$

and

$$\rho_{ab}^{(a)}(\underline{r},t,\underline{v}) = \lambda_a \, W(\underline{v}) \int_{-\infty}^{t} dt_o \; \rho_{ab}^{(a)}(\underline{r} = \underline{r}_o + \underline{v}(t - t_o),\ \underline{v},t,t_o) \quad , \tag{3.2-7}$$

with similar equations for the contributions due to atoms excited to the state $|b)$.

If we can calculate 3.2-5 to 7 and the corresponding contributions from excitation of the $|b)$ states, we can calculate $\rho_{ab}(\underline{r},t)$ and thus the polarization by integrating over the velocity distribution. However, these equations in general cannot be solved exactly, and the next section gives an approach leading to various approximate solutions.

For the special case of stationary atoms, where $W(\underline{v}) = \delta(\underline{v})$, the three-dimensional Dirac delta function, the integration over velocities is trivial and can be carried out immediately by setting $\underline{v} = 0$ and $\underline{r} = \underline{r}_o$, giving the result

$$\rho_{aa}^{(a)}(\underline{r},t) = \lambda_a \int_{-\infty}^{t} dt_o \; \rho_{aa}^{(a)}(\underline{r},t,t_o) \quad , \qquad 3.2\text{-}8$$

with similar equations for the other quantities of interest. If we now calculate the time derivative of 3.2-8, we have

$$\dot{\rho}_{aa}^{(a)}(\underline{r},t) = \lambda_a \int_{-\infty}^{t} dt_o \left[\dot{\rho}_{aa}^{(a)}(\underline{r},t,t_o) + \lambda_a \; \rho_{aa}^{(a)}(\underline{r},t_o,t_o)\right] \quad . \qquad 3.2\text{-}9$$

Using 3.1-3 and the fact that $\rho_{aa}^{(a)}(\underline{r},t_o,t_o) = 1$ by definition, this becomes

$$\dot{\rho}_{aa}^{(a)}(\underline{r},t) = \lambda_a - \gamma_a\lambda_a \int_{-\infty}^{t} dt_o \; \rho_{aa}^{(a)}(\underline{r},t,t_o)$$
$$- \lambda_a \int_{-\infty}^{t} dt_o(\rho_{ab}^{(a)}(\underline{r},t,t_o) + \rho_{ab}^{*(a)}(\underline{r},t,t_o)) \qquad 3.2\text{-}10$$
$$\times \frac{\omega_o}{\hbar} \sum_{\omega} \frac{\underline{E}_\omega \cdot \underline{P}_o}{\omega} \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega) \quad .$$

Using 3.2-8 and the corresponding equation for $\rho_{ab}^{(a)}(\underline{r},t)$, 3.2.10 becomes

$$\dot{\rho}_{aa}^{(a)}(\underline{r},t) = \lambda_a - \gamma_a \rho_{aa}^{(a)}(\underline{r},t) - (\rho_{ab}^{(a)}(\underline{r},t) + \rho_{ab}^{*(a)}(\underline{r},t))$$

$$\times \frac{\omega_o}{\hbar} \sum_{\omega} \frac{\underline{E}_\omega \cdot \underline{P}_o}{\omega} \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega) \quad . \qquad 3.2\text{-}11$$

Thus we have an equation of motion for the quantity $\rho_{aa}^{(a)}(\underline{r},t)$. By changing subscripts and using

$$\rho_{bb}^{(a)}(\underline{r},t_o,t_o) = \rho_{ab}^{(a)}(\underline{r},t_o,t_o) = \rho_{aa}^{(b)}(\underline{r},t_o,t_o)$$

$$= \rho_{ab}^{(b)}(\underline{r},t_o,t_o) = 0 \; ; \quad \rho_{bb}^{(b)}(\underline{r},t_o,t_o) = 1 \quad , \qquad 3.2\text{-}11$$

and

$$\rho_{aa}(\underline{r},t) = \rho_{aa}^{(a)}(\underline{r},t) + \rho_{aa}^{(b)}(\underline{r},t) \quad , \qquad 3.2\text{-}12$$

etc., the full set of equations of motion is found directly to be:

$$\dot{\rho}_{aa}(\underline{r},t) = \lambda_a - \gamma_a \rho_{aa}(\underline{r},t) - (\rho_{ab}(\underline{r},t) + \rho_{ab}^{*}(\underline{r},t))$$

$$\times \frac{\omega_o}{\hbar} \sum_{\omega} \frac{\underline{E}_\omega \cdot \underline{P}_o}{\omega} \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega) \quad , \qquad 3.2\text{-}13$$

$$\dot{\rho}_{bb}(\underline{r},t) = \lambda_b - \gamma_b \rho_{bb}(\underline{r},t) + (\rho_{ab}(\underline{r},t) + \rho_{ab}^{*}(\underline{r},t))$$

$$\times \frac{\omega_o}{\hbar} \sum_{\omega} \frac{\underline{E}_\omega \cdot \underline{P}_o}{\omega} \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega) \quad , \qquad 3.2\text{-}14$$

and

$$\dot{\rho}_{ab}(\underline{r},t) = -(\gamma + i\omega_o)\,\rho_{ab}(\underline{r},t) + (\rho_{aa}(\underline{r},t) - \rho_{bb}(\underline{r},t))$$

$$\times \frac{\omega_o}{\hbar} \sum_{\omega} \frac{\underline{E}_\omega \cdot \underline{P}_o}{\omega} \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi_\omega) \quad . \qquad 3.2\text{-}15$$

These equations are analogous to ones used earlier by Lamb as the basis of a treatment of optical masers which neglected atomic motion (11). It should be noted that the quantities $\rho_{aa}(\underline{r},t)$ etc., in 3.2-13 to 15 have dimensions m^{-3} rather than being dimensionless as were ρ_{aa} etc., in 3.1-3 to 5. This is of course due to the fact that the former represent summations of the latter over all atoms within the unit volume. Thus $\rho_{aa}(\underline{r},t)$ and $\rho_{bb}(\underline{r},t)$ are the populations of the states $|a)$ and $|b)$ per unit volume at time t and position $\underline{r}$, and $\rho_{aa}(\underline{r},t) - \rho_{bb}(\underline{r},t)$ is the population inversion density for the transition between $|a)$ and $|b)$. Correspondingly,

$$\underline{P}(\underline{r},t) = \underline{P}_o(\rho_{ab}(\underline{r},t) + \rho^*_{ab}(\underline{r},t)) \qquad 3.2\text{-}16$$

is the dipole moment per unit volume, and is thus the macroscopic polarization appearing in the equations of motion for the field, 2.3.2-20 and 21. The quantities of 3.2-13 to 15 are those which are to satisfy the quasi-steady state solution that was introduced at the beginning of this section and which will be discussed in more detail in the next section and following chapters.

If we attempt the same approach for the case of moving atoms, we get from 3.2-6 using 3.1-3,

$$\dot{\rho}_{aa}^{(a)}(\underline{r},t,\underline{v}) = \lambda_a W(\underline{v}) \int_{-\infty}^{t} dt_o \Big[- \gamma_a \rho_{aa}^{(a)}(\underline{r} = \underline{r}_o + \underline{v}(t - t_o), \underline{v},t,t_o)$$

$$- (\rho_{ab}^{(a)}(\underline{r} = \underline{r}_o + \underline{v}(t - t_o), \underline{v},t,t_o) + \rho_{ab}^{*(a)}(\underline{r} = \underline{r}_o + \underline{v}(t - t_o), \underline{v},t,t_o)$$

$$\times \frac{\omega_o}{\hbar} \sum_{\omega} \frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega} \sin(\underline{k} \cdot \underline{r}_o + \underline{k} \cdot \underline{v}(t - t_o) - \omega t + \varphi) \Big] \quad .$$

3.2-17

Due to the presence of t_o in the sine factor of 3.2-17, we can no longer carry out the t_o integration to obtain differential equations similar to 3.2-13 to 15. One alternative is to explicitly calculate, in some approximation, the quantities like $\rho_{aa}^{(a)}(\underline{r} = \underline{r}_o + \underline{v}(t - t_o), \underline{v}, t, t_o)$ from 3.1-3 to 5, and then use 3.2-5, etc., to find $\rho_{aa}(\underline{r},t,\underline{v})$, etc. We would then integrate $\rho_{ab}(\underline{r},t,\underline{v})$ over the velocity distribution to obtain the macroscopic polarization 3.2-16. Lamb used this approach, with a small-field assumption allowing a perturbation expansion of the solutions, in his theory of an optical maser (12). In the next section, we derive an integral equation approach, from which the perturbation and other useful approximations follow easily.

3.3 The Form of Solutions for the Medium

In this section we derive an integral equation for the population inversion density and relate the latter to the polarization induced by the fields. We then briefly discuss the types of approximate solutions obtainable from this formalism.

The object here is to calculate the quantities $\rho_{aa}(\underline{r},t,\underline{v})$, $\rho_{bb}(\underline{r},t,\underline{v})$ and $\rho_{ab}(\underline{r},t,\underline{v})$, which were defined in the last section. For the field equations of motion we of course need only $\rho_{ab}(\underline{r},t,\underline{v})$, but it is convenient and physically interesting to express this in terms of the quantity

$$\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v}) \quad , \qquad 3.3\text{-}1$$

which is interpreted as the population inversion density at $\underline{r},t$, of atoms with velocity $\underline{v}$. When 3.3-1 is integrated over velocities, we obtain the total population inversion density at $\underline{r},t$. Integrating $\rho_{ab}(\underline{r},t,\underline{v})$ over velocities gives us 3.2-16, the macroscopic polarization induced by the fields.

Each of the above quantities will have contributions from atoms excited to the state $|a)$ and to the state $|b)$. These contributions are calculated separately and then added. The notation of the last section is followed. To simplify writing the equations, we will not explicitly write some of the arguments of various functions. For example, the contribution to $\rho_{aa}(\underline{r},t,\underline{v})$ of an atom excited to state $|a)$ at $\underline{r}_o$, t_o will be denoted by $\rho_{aa}^{(a)}(t,t_o)$ rather than $\rho_{aa}^{(a)}(\underline{r} = \underline{r}_o + \underline{v}(t - t_o),\ t,\underline{v},t_o)$. This will make the following clearer as well as simpler.

The quantities $\rho_{aa}^{(a)}(t,t_o)$, etc., are solutions of the differential equations 3.1-3 to 5. We consider first an atom excited to state $|a)$ at r_o,t_o, with velocity $\underline{v}$. Then the quantities of interest are

$$\rho_{aa}^{(a)}(t,t_o),\ \rho_{bb}^{(a)}(t,t_o),\ \rho_{ab}^{(a)}(t,t_o),$$

with the initial conditions

$$\rho_{aa}^{(a)}(t_o,t_o) = 1,\ \rho_{bb}^{(a)}(t_o,t_o) = \rho_{ab}^{(a)}(t_o,t_o) = 0 \quad . \qquad 3.3\text{-}2$$

For added simplicity, we temporarily drop the superscript (a) ; superscripts will be explicitly written later when identification of each contribution is necessary. If we define $\rho'_{aa}(t,t_o)$, $\rho'_{bb}(t,t_o)$ and $\rho'_{ab}(t,t_o)$ by

$$\rho_{aa}(t,t_o) = \rho'_{aa}(t,t_o)\, e^{-\gamma_a(t - t_o)} \quad , \qquad 3.3\text{-}3$$

$$\rho_{bb}(t,t_o) = \rho'_{bb}(t,t_o)\, e^{-\gamma_b(t - t_o)} \quad , \qquad 3.3\text{-}4$$

$$\rho_{ab}(t,t_o) = \rho'_{ab}(t,t_o)\, e^{-(\gamma + i\omega_o)(t - t_o)} \quad , \qquad 3.3\text{-}5$$

and substitute these expressions into 3.1-3 to 5, we find

$$\dot{\rho}'_{aa}(t,t_o) = -e^{\gamma_a(t - t_o)}(\rho_{ab}(t,t_o) + \rho^*_{ab}(t,t_o))\frac{\omega_o}{\hbar}$$
$$\times \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right) \sin(\underline{k}\cdot\underline{r}_o + \underline{k}\cdot\underline{v}(t - t_o) - \omega t + \varphi) \quad , \qquad 3.3\text{-}6$$

$$\dot{\rho}'_{bb}(t,t_o) = e^{\gamma_b(t - t_o)}(\rho_{ab}(t,t_o) + \rho^*_{ab}(t,t_o))\frac{\omega_o}{\hbar}$$

$$\times \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right) \sin(\underline{k} \cdot \underline{r}_o + \underline{k} \cdot \underline{r}(t - t_o) - \omega t + \varphi) \quad , \qquad 3.3\text{-}7$$

$$\dot{\rho}'_{ab}(t,t_o) = e^{(\gamma + i\omega_o)(t - t_o)}(\rho_{aa}(t,t_o) - \rho_{bb}(t,t_o)) \frac{\omega_o}{\hbar}$$

$$\times \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right) \sin(\underline{k} \cdot \underline{r}_o + \underline{k} \cdot \underline{v}(t - t_o) - \omega t + \varphi) \quad , \qquad 3.3\text{-}8$$

with the same initial conditions as those for the unprimed quantities, 3.3-2. The differential equations 3.3-6 to 8 can be formally integrated to give

$$\rho'_{aa}(t,t_o) = 1 - \int_{t_o}^{t} dt' \; e^{\gamma_a(t' - t_o)}(\rho_{ab}(t',t_o) + \rho^*_{ab}(t',t_o)) \frac{\omega_o}{\hbar}$$

$$\times \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right) \sin(\underline{k} \cdot \underline{r}_o + \underline{k} \cdot \underline{v}(t' - t_o) - \omega t' + \varphi) \quad , \qquad 3.3\text{-}9$$

$$\rho'_{bb}(t,t_o) = \int_{t_o}^{t} dt' \; e^{\gamma_b(t' - t_o)}(\rho_{ab}(t',t_o) + \rho^*_{ab}(t',t_o)) \frac{\omega_o}{\hbar}$$

$$\times \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right) \sin(\underline{k} \cdot \underline{r}_o + \underline{k} \cdot \underline{v}(t' - t_o) - \omega t' + \varphi) \quad , \qquad 3.3\text{-}10$$

$$\rho'_{ab}(t,t_o) = \int_{t_o}^{t} dt' \, e^{(\gamma + i\omega_o)(t' - t_o)} (\rho_{aa}(t',t_o) - \rho_{bb}(t',t_o)) \frac{\omega_o}{\hbar}$$

$$\times \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega} \right) \sin(\underline{k} \cdot \underline{r}_o + \underline{k} \cdot \underline{v}(t' - t_o) - \omega t' + \varphi) \quad . \qquad 3.3\text{-}11$$

Using 3.3-3 to 5, we obtain the unprimed quantities by multiplying with the appropriate exponential. Thus we have

$$\rho_{aa}(t,t_o) = e^{-\gamma_a(t - t_o)} - \int_{t_o}^{t} dt' \, e^{\gamma_a(t' - t)} (\rho_{ab}(t',t_o) + \rho^*_{ab}(t',t_o)) \frac{\omega_o}{\hbar}$$

$$\times \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega} \right) \sin(\underline{k} \cdot \underline{r}_o + \underline{k} \cdot \underline{v}(t' - t_o) - \omega t' + \varphi) \quad . \qquad 3.3\text{-}12$$

$$\rho_{bb}(t,t_o) = \int_{t_o}^{t} dt' \, e^{\gamma_b(t' - t)} (\rho_{ab}(t',t_o) + \rho^*_{ab}(t',t_o)) \frac{\omega_o}{\hbar}$$

$$\times \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega} \right) \sin(\underline{k} \cdot \underline{r}_o + \underline{k} \cdot \underline{v}(t' - t_o) - \omega t' + \varphi) \quad , \qquad 3.3\text{-}13$$

$$\rho_{ab}(t,t_o) = \int_{t_o}^{t} dt' \, e^{(\gamma + i\omega_o)(t' - t)} (\rho_{aa}(t',t_o) - \rho_{bb}(t',t_o)) \frac{\omega_o}{\hbar}$$

$$\times \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega} \right) \sin(\underline{k} \cdot \underline{r}_o + \underline{k} \cdot \underline{v}(t' - t_o) - \omega t + \varphi) \quad . \qquad 3.3\text{-}14$$

In the following calculations we will often have occasion to compare expressions like

$$\int_0^\infty dt\ e^{-[\gamma + i(\omega_o - \omega)]t} \quad \text{and} \int_0^\infty dt\ e^{-[\gamma + i(\omega_o + \omega)]t}$$

For $\gamma \ll \omega_o$, $\omega_o \approx \omega$, these are of relative order ω_o/γ, or about $10^{15}/10^8 - 10^{+7}$ for optical transitions. We will therefore always neglect "anti-resonant" terms like the second compared to "resonant" terms like the first.

We can subtract 3.3-13 from 3.3-12 to obtain an expression for $\rho_{aa}(t,t_o) - \rho_{bb}(t,t_o)$ in terms of $\rho_{ab}(t',t_o)$. Substituting $\rho_{ab}(t',t_o)$ from 3.3-14 into this expression gives an integral equation for $\rho_{aa}(t,t_o) - \rho_{bb}(t,t_o)$:

$$\rho_{aa}(t,t_o) - \rho_{bb}(t,t_o) = e^{-\gamma_a(t - t_o)} - \left(\frac{\omega_o}{2\hbar}\right)^2 \int_{t_o}^{t} dt' \int_{t_o}^{t'} dt''$$

$$\times \left(e^{\gamma_a(t'-t)} + e^{\gamma_b(t'-t)}\right)\left(e^{(\gamma+i\omega_o)(t''-t')} + e^{(\gamma-i\omega_o)(t''-t')}\right)$$

$$\times \left(\rho_{aa}(t'',t_o) - \rho_{bb}(t'',t_o)\right) \sum_{\omega,\omega'} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right)$$

$$\times \left[e^{i[(\underline{k} - \underline{k}')\cdot\underline{r}_o + \underline{k}\cdot\underline{v}(t'-t_o) - \underline{k}'\cdot\underline{v}(t''-t_o) - \omega t' + \omega' t'' + \varphi - \varphi']} + \text{c.c.}\right] \qquad 3.3\text{-}15$$

where "c.c." indicates the complex conjugate, and anti-resonant terms have been neglected. 3.3-15 is written for a position $\underline{r}$ at time t such that $\underline{r} = \underline{r}_o + \underline{v}(t - t_o)$. We can use this to write the final exponent in 3.3-15 as

$$\left[(\underline{k} - \underline{k}') \cdot \underline{r}_o + \underline{k} \cdot \underline{v}(t' - t_o) - \underline{k}' \cdot \underline{v}(t'' - t_o) - \omega t' + \omega' t'' + \varphi - \varphi'\right]$$

$$= \left[(\underline{k} - \underline{k}') \cdot \underline{r} - \underline{k} \cdot \underline{v}(t - t') + \underline{k}' \cdot \underline{v}(t - t'') - \omega t' + \omega' t'' + \varphi - \varphi'\right] .$$

3.3-16

If we sum 3.3-15 over all atoms per unit volume at $\underline{r}_o$, we obtain a factor $\lambda_a W(\underline{v})$ as previously discussed in section 3.2. By integrating over all possible excitation times t_o and using 3.2-5, we find

$$\rho_{aa}^{(a)}(\underline{r},t,\underline{v}) - \rho_{bb}^{(a)}(\underline{r},t,\underline{v}) = \frac{\lambda_a W(\underline{v})}{\gamma_a} - \lambda_a W(\underline{v}) \left(\frac{\omega_o}{2\hbar}\right)^2$$

$$\times \int_{-\infty}^{t} dt_o \int_{t_o}^{t} dt' \int_{t_o}^{t'} dt'' \left(e^{\gamma_a (t' - t)} + e^{\gamma_b (t' - t)}\right)\left(e^{(\gamma + i\omega_o)(t'' - t')} + e^{(\gamma - i\omega_o)(t'' - t')}\right)$$

$$\times \left(\rho_{aa}^{(a)}(t'',t_o) - \rho_{bb}^{(a)}(t'',t_o)\right) \sum_{\omega,\omega'} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right)$$

$$\times \left[e^{i[(\underline{k} - \underline{k}') \cdot \underline{r} - \underline{k} \cdot \underline{v}(t - t') + \underline{k}' \cdot \underline{v}(t - t'') - \omega t' + \omega' t'' + \varphi - \varphi']} + c.c.\right]$$

3.3-17

where the superscript (a) has been written again. By two interchanges of the order of integration,

$$\int_{-\infty}^{t} dt_o \int_{t_o}^{t} dt' \int_{t_o}^{t'} dt'' \quad \text{becomes} \quad \int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt'' \int_{-\infty}^{t''} dt_o \quad .$$

The t_o integration can be done by using 3.2-5 and 6 again, giving finally

$$\rho_{aa}^{(a)}(\underline{r},t,\underline{v}) - \rho_{bb}^{(a)}(\underline{r},t,\underline{v}) = \frac{\lambda_a W(\underline{v})}{\gamma_a} - \left(\frac{\omega_o}{2\hbar}\right)^2 \int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt''$$

$$\times \left(e^{\gamma_a(t'-t)} + e^{\gamma_b(t'-t)}\right)\left(e^{(\gamma + i\omega_o)(t''-t')} + e^{(\gamma - i\omega_o)(t''-t')}\right)$$

$$\times \left(\rho_{aa}^{(a)}(\underline{r},t'',\underline{v}) - \rho_{bb}^{(a)}(\underline{r},t'',\underline{v})\right) \sum_{\omega,\omega'} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right)$$

$$\times \left[e^{i\Delta + i[-k \cdot \underline{v}(t-t') + \underline{k}' \cdot \underline{v}(t-t'') - \omega(t'-t) + \omega'(t''-t)]} + c.c.\right] , \qquad 3.3\text{-}18$$

where

$$\Delta \equiv (\underline{k} - \underline{k}') \cdot \underline{r} - (\omega - \omega')t + \varphi - \varphi' \quad . \qquad 3.3\text{-}19$$

In an exactly similar way we find the contribution due to excitation of atoms to the state $|b)$:

$$\rho_{aa}^{(b)}(\underline{r},t,\underline{v}) - \rho_{bb}^{(b)}(\underline{r},t,\underline{v}) = -\frac{\lambda_b W(\underline{v})}{\gamma_b} - \left(\frac{\omega_o}{2\hbar}\right)^2 \int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt''$$

$$\times\left(e^{\gamma_a(t'-t)} + e^{\gamma_b(t'-t)}\right)\left(e^{(\gamma+i\omega_o)(t''-t')} + e^{(\gamma-i\omega_o)(t''-t')}\right)$$

$$\times\left(\rho_{aa}^{(b)}(\underline{r},t,\underline{v}) - \rho_{bb}^{(b)}(\underline{r},t,\underline{v})\right)\sum_{\omega,\omega'}\left(\frac{\underline{P}_o\cdot\underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o\cdot\underline{E}_{\omega'}}{\omega'}\right)$$

$$\times\left[e^{i\Delta + i[-\underline{k}\cdot\underline{r}(t-t') + \underline{k}'\cdot\underline{v}(t-t'') - \omega(t'-t) + \omega'(t''-t)]} + c.c.\right].$$

3.3-20

If we add 3.3-18 and 20 and define the new variables of integration $t_1 = t - t'$, $t_2 = t' - t''$, we obtain

$$\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v}) = NW(\underline{v}) - \left(\frac{\omega_o}{2\hbar}\right)^2 \int_0^\infty dt_1 \int_0^\infty dt_2$$

$$\times\left(e^{-\gamma_a t_1} + e^{-\gamma_b t_1}\right)\left(\rho_{aa}(\underline{r},t-t_1-t_2,\underline{v}) - \rho_{bb}(\underline{r},t-t_1-t_2,\underline{v})\right)$$

$$\times\sum_{\omega,\omega'}\left(\frac{\underline{P}_o\cdot\underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o\cdot\underline{E}_{\omega'}}{\omega'}\right)\left[e^{i\Delta + i(\omega-\omega'-(\underline{k}-\underline{k}')\cdot\underline{v})t_1}\right.$$

$$\left.\times\, e^{-[\gamma - i(\omega_o - \omega' + \underline{k}'\cdot\underline{v})]t_2} + c.c.\right],$$

3.3-21

where we have neglected antiresonant terms, defined

$$N \equiv \frac{\lambda_a}{\gamma_a} - \frac{\lambda_b}{\gamma_b} \qquad 3.3\text{-}22$$

and assumed $\underline{k} \cdot \underline{v} \ll \omega$.

3.3-21 is an integral equation for the population inversion density at $\underline{r},t$ with velocity $\underline{v}$. This is a physically interesting quantity, and some approximate solutions of 3.3-21 will be used in Chapter 6 for studying higher order nonlinear effects. We are more directly interested in the quantity $\rho_{ab}(\underline{r},t,\underline{v})$ and thus the macroscopic polarization. We can relate $\rho_{ab}(\underline{r},t,\underline{v})$ to the population inversion density by the same techniques used to derive the integral equation 3.3-21, and this will now be done. For our purposes it is better to do this than to find an integral equation for $\rho_{ab}(\underline{r},t,\underline{v})$, although the latter could also be done.

For the contribution to $\rho_{ab}(\underline{r},t,\underline{v})$ from atoms excited to state $|a)$, we must substitute 3.3-15 into 3.3-14, giving

$$\rho_{ab}^{(a)}(t,t_o) = \frac{\omega_o}{2i\hbar}\int_{t_o}^{t} dt' \, e^{(\gamma + i\omega_o)(t' - t)} \Bigg\{ e^{-\gamma_a(t' - t_o)}$$

$$\times \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right) \left[e^{i(\underline{k} \cdot \underline{r}_o + \underline{k} \cdot \underline{v}(t' - t_o) - \omega t' + \varphi)} - \text{c.c.}\right]$$

$$\left(\frac{\omega_o}{2\hbar}\right)^2 \int_{t_o}^{t'} dt'' \int_{t_o}^{t''} dt''' \left(e^{\gamma_a(t'' - t')} + e^{\gamma_b(t'' - t')}\right)$$

$$\times \left(e^{(\gamma + i\omega_o)(t''' - t'')} + e^{(\gamma - i\omega_o)(t''' - t'')} \right) \left(\rho_{aa}^{(a)}(t''', t_o) \right.$$

$$\left. - \rho_{bb}^{(a)}(t''', t_o) \right) \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega} \right) \left[e^{i(\underline{k} \cdot \underline{r}_o + \underline{k} \cdot \underline{v}(t' - t_o) - \omega t' + \varphi)} - c.c. \right]$$

$$\times \sum_{\omega', \omega''} \left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'} \right) \left(\frac{\underline{P}_o \cdot \underline{E}_{\omega''}}{\omega''} \right) \left[e^{i[(\underline{k}' - \underline{k}'') \cdot \underline{r}_o + \underline{k}' \cdot \underline{v}(t'' - t_o) - \underline{k}'' \cdot \underline{v}(t''' - t_o)]} \right.$$

$$\left. \left. \times e^{i[-\omega' t'' + \omega'' t''' + \varphi' - \varphi'']} + c.c. \right] \right\} . \qquad 3.3\text{-}23$$

A similar contribution is obtained due to excitation of atoms to state $|b)$, and the sum of these contributions gives $\rho_{ab}(t, t_o)$. We again use $\underline{r} = r_o + \underline{v}(t - t_o)$ to get rid of t_o in the exponentials, sum each contribution over all atoms per unit volume, and integrate over excitation times t_o. The order of integration can be changed to make the t_o integration first, and this integration can be carried out by using 3.2-5 to 7. With a change of variables to

$$t_1 = t - t', \; t_2 = t' - t'', \; t_3 = t'' - t''' ,$$

we then have

$$\rho_{ab}(\underline{r}, t, \underline{v}) = \frac{\omega_o}{2i\hbar} \int_0^\infty dt_1 \, e^{-(\gamma + i\omega_o)t_1} \left\{ NW(\underline{v}) \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega} \right) \right.$$

$$\times \left[e^{i[1] + i(\omega - \underline{k} \cdot \underline{v})t_1} - c.c. \right] - \left(\frac{\omega_o}{\hbar} \right)^2 \int_0^\infty dt_2 \int_0^\infty dt_3$$

$$\times\left(e^{-\gamma_a t_2} + e^{-\gamma_b t_2}\right)\left(e^{-(\gamma + i\omega_o)t_3} + e^{-(\gamma - i\omega_o)t_3}\right)$$

$$\times\left(\rho_{aa}(\underline{r}, t - t_1 - t_2 - t_3, \underline{v}) - \rho_{bb}(\underline{r}, t - t_1 - t_2 - t_3, \underline{v})\right)$$

$$\times \sum_{\omega}\left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left[e^{i[1] + i(\omega - \underline{k} \cdot \underline{v})t_1} - \text{c.c.}\right]\sum_{\omega',\omega''}\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega''}}{\omega''}\right)$$

$$\times\left[e^{i\Delta + i(\omega' - \underline{k}' \cdot \underline{v})(t_1 + t_2) - i(\omega'' - \underline{k}'' \cdot \underline{v})(t_1 + t_2 + t_3)} + \text{c.c.}\right]\Bigg\} \qquad 3.3\text{-}24$$

where

$$[1] = \underline{k} \cdot \underline{r} - \omega t + \varphi \qquad 3.3\text{-}25$$

and

$$\Delta = (\underline{k}' - \underline{k}'') \cdot \underline{r} - (\omega' - \omega'')t + \varphi' - \varphi'' \quad .$$

We see that 3.3-24 has the form

$$\rho_{ab}(\underline{r}, t, \underline{v}) = \rho_{ab}^{lin}(\underline{r}, t, \underline{v}) + \rho_{ab}^{NL}(\underline{r}, t, \underline{v}) \quad , \qquad 3.3\text{-}26$$

where the superscripts lin and NL indicate contributions to $\rho_{ab}(\underline{r}, t, \underline{v})$ which are linear and nonlinear, respectively, in the fields. Neglecting antiresonant terms in 3.3-24, we have

$$\rho_{ab}^{lin}(\underline{r}, t, \underline{v}) = \frac{\omega_o NW(\underline{v})}{2i\hbar}\sum_{\omega}\left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\frac{e^{i(\underline{k} \cdot \underline{r} - \omega t + \varphi)}}{\gamma + i(\omega_o - \omega + \underline{k} \cdot \underline{v})} \quad , \qquad 3.3\text{-}27$$

$$\rho_{ab}^{NL}(\underline{r},t,\underline{v}) = \left(\frac{\omega_o}{2i\hbar}\right)^3 \int_0^\infty dt_1 \int_0^\infty dt_2 \int_0^\infty dt_3 \left(e^{-\gamma_a t_2} + e^{-\gamma_b t_2}\right)$$

$$\times\left(\rho_{aa}(\underline{r},t - t_1 - t_2 - t_3,\underline{v}) - \rho_{bb}(\underline{r},t - t_1 - t_2 - t_3,\underline{v})\right)$$

$$\times \sum_{\omega,\omega',\omega''} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega''}}{\omega''}\right) e^{i[1] - [\gamma + i(\omega_o - \omega + \underline{k}\cdot\underline{v})]t_1}$$

$$\times\left[e^{i\Delta + i[\omega' - \omega'' - (\underline{k}' - \underline{k}'')\cdot\underline{v}](t_1 + t_2) - [\gamma - i(\omega_o - \omega'' + \underline{k}''\cdot\underline{v})]t_3} + \text{c.c.}\right] \qquad 3.3\text{-}28$$

where $[1]$ and Δ are given in 3.3-25.

3.3-27 and 28, along with 3.3-21, are the basic results we require for our later use. We will also find it useful to write $\rho_{ab}(\underline{r},t,\underline{v})$ in terms of $\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v})$ starting from 3.3-14. In an identical way, we find

$$\rho_{ab}(\underline{r},t,\underline{v}) = \frac{\omega_o}{2i\hbar}\int_0^\infty dt_1 \, (\rho_{aa}(\underline{r},t - t_1,\underline{v}) - \rho_{bb}(\underline{r},t - t_1,\underline{v}))$$

$$\times \sum_\omega \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right) e^{i[1] - [\gamma + i(\omega_o - \omega + \underline{k}\cdot\underline{v})]t_1} \quad , \qquad 3.3\text{-}29$$

where we have assumed the variation of $\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v})$ to be very much smaller than ω , and have neglected antiresonant terms.

Let us now examine briefly the kinds of approximate solutions obtainable from the above equations. With no field, we have

$$\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v}) = NW(\underline{v}) \qquad 3.3\text{-}30$$

and

$$\rho_{aa}(\underline{r},t) - \rho_{bb}(\underline{r},t) = N \quad ,$$

with

$$\rho_{ab}(\underline{r},t,\underline{v}) = \rho_{ab}(\underline{r},t) = 0 \quad .$$

With fields, the simplest solution is to use the linear solution 3.3-27, which corresponds to putting the zero-field solution for $\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v})$, $NW(\underline{v})$, into 3.3-29. This case is considered in Chapter 4. The lowest order nonlinear terms can be obtained either by putting the zero-field solution for $\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v})$ into the right hand side of 3.3-21 and substituting the resulting second order expression for $\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v})$ into 3.3-29, or by using the zero-field solution for $\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v})$ in the right hand side of 3.3-28. Either method gives a third order expression for $\rho_{ab}(\underline{r},t,\underline{v})$, and the latter method is used in Chapter 5, where the lowest order nonlinear effects are studied in detail. Higher order terms in the perturbation expansions of $\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v})$ and $\rho_{ab}(\underline{r},t,\underline{v})$ in powers of the fields can be obtained by continued iteration of the above approach, which is ideally suited for this purpose. In this expansion, we see that successive iterations for both quantities are two orders higher in the field, ρ_{ab} and $\rho_{aa} - \rho_{bb}$ containing the fields to odd and even powers, respectively. We note that $\rho_{aa} - \rho_{bb}$ will be slowly varying compared to the optical frequencies ω, and that ρ_{ab} will be essentially varying harmonically at these frequencies. We will not explicitly consider these higher order terms except briefly in the discussion of a more approximate method used in Chapter 6 for studying higher order effects. This last method is quite useful when the fields are so large that the perturbation expansion of the solutions becomes invalid. It involves making some assumptions on the character of $\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v})$ such that the integral equation 3.3-21 can be approximately solved.

It will become clear that the two methods of solution (i.e., the perturbation expansion and the approximate solution of 3.3-21) complement each other to a large extent; the former giving an accurate picture of the processes involved as well as accurate expressions for quantities of interest, the latter giving a good picture of high-field effects and expressions which agree to first order with the perturbation expansion. It will also be seen that the perturbation expansion indicates qualitatively the corrections which must be made to the results of the approximate method in cases where the latter does not apply directly.

CHAPTER FOUR

LINEAR AMPLIFICATION

4.1 Introduction

An applied field with a frequency within the transition linewidth will cause transitions between and mixing of the levels $|a)$ and $|b)$ and thus lead to nonlinear effects. For this chapter, we assume that the population inversion density, $\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v})$, retains its zero-field value, $NW(\underline{v})$. In this case $\rho_{ab}(\underline{r},t,\underline{v})$ reduces to $\rho_{ab}^{lin}(\underline{r},t,\underline{v})$, given by 3.3-27. The conditions under which it is valid to neglect nonlinear effects will be derived in Chapter 5, where the lowest order nonlinear effects will be considered.

It is useful to consider these linear effects, since they relate the present formalism to well-known results and their study forms a basis for treating the nonlinear effects, and for comparison. Also, the magnitude, frequency dependence, etc., of the nonlinear effects are of course related to the parameters characterizing linear amplification, so that the latter are important.

In this chapter we consider two specific velocity distributions of excited atoms. First, stationary atoms, for which $W(\underline{v}) = \delta(\underline{v})$, the three-dimensional Dirac delta function, are considered in section 4.2. Then a Maxwellian velocity distribution, i.e.

$$W(\underline{v}) = \left(\frac{1}{\pi u^2}\right)^{\frac{3}{2}} e^{-v^2/u^2} , \qquad 4.1\text{-}1$$

where $u^2 = 2kT/M$ (23), is considered in section 4.3.

4.2 Stationary Atoms

With $W(\underline{v}) = \delta(\underline{v})$, the integration over velocities is trivial and gives from 3.3-27 the result

$$\rho_{ab}(\underline{r},t) = \frac{\omega_o N}{2i\hbar} \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega} \right) \frac{e^{i(\underline{k} \cdot \underline{r} - \omega t + \varphi)}}{\gamma + i(\omega_o - \omega)} \quad . \qquad 4.2\text{-}1$$

Substituting 4.2-1 into 3.2-16, we find the macroscopic polarization

$$\underline{P}(\underline{r},t) = \frac{\underline{P}_o \omega_o N}{2i\hbar} \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega} \right) \left[\frac{e^{i(\underline{k} \cdot \underline{r} - \omega t + \varphi)}}{\gamma + i(\omega_o - \omega)} - \text{c.c.} \right] \quad , \qquad 4.2\text{-}2$$

which can be rewritten as

$$\underline{P}(\underline{r},t) = \frac{\underline{P}_o \omega_o N}{\hbar} \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega} \right) \left[\frac{\gamma \sin(\underline{k} \cdot \underline{r} - \omega t + \varphi) - (\omega_o - \omega)\cos(\underline{k} \cdot \underline{r} - \omega t + \varphi)}{\gamma^2 + (\omega_o - \omega)^2} \right]$$

4.2-3

According to semiclassical radiation theory (3), we must average over all possible directions of the vector

$$\underline{P}_o \equiv (a| - e\underline{r}|b) \qquad 4.2\text{-}4$$

to determine the direction of the induced polarization. For all our work we assume that there is nothing in the unperturbed system (zero-fields) to establish any preferred direction, i.e., that the unperturbed system is isotropic. Then any direction of $\underline{P}_o$ is equally likely, so that the probability of $\underline{P}_o$ being within the incremental solid angle

$d\Omega$ around the direction defined by the angles θ, φ relative to some chosen axis is

$$P(\theta,\varphi)d\Omega = \frac{d\Omega}{4\pi} = \frac{\sin\,\theta d\theta d\varphi}{4\pi} \quad . \qquad 4.2\text{-}4$$

For other cases, where some natural anisotropy exists, such as an external DC magnetic field or anisotropic crystalline sites, the situation will be more complex and will depend on the particular characteristics of the states $|a)$ and $|b)$. Since we are interested in studying any anisotropy induced by nonlinear effects, it is desirable to begin with an isotropic unperturbed medium, which is also of course the simplest case.

The field equations of motion 2.3.2-20 and 21 require the component of $\underline{P}(z,t)$ at frequency ω along the direction of $\underline{E}_\omega$. Taking the latter direction to be the $\theta = 0$ axis, we find the component of $\underline{P}_o(\underline{P}_o \cdot \underline{E}_\omega)$ along $\underline{E}_\omega$ to be

$$\frac{P_o^2 E_\omega}{4\pi} \int_0^\pi d\theta \int_0^{2\pi} d\varphi \, \sin\theta \, \cos^2\theta = \frac{P_o^2 E_\omega}{3} \quad . \qquad 4.2\text{-}5$$

Similarly, the components of $\underline{P}_o(\underline{P}_o \cdot \underline{E}_\omega)$ along the two directions perpendicular to $\underline{E}_\omega$ are

$$\frac{P_o^2 E_\omega}{4\pi} \int_0^\pi \cos\theta \, \sin^2\theta d\theta \int_0^{2\pi} \cos\varphi \, d\varphi = 0$$

$$\frac{P_o^2 E_\omega}{4\pi} \int_0^\pi \cos\theta \ \sin^2\theta d\theta \int_0^{2\pi} \sin\varphi d\varphi = 0$$

Thus the induced polarization has only a component parallel to the field, and we have from 4.2-3 and 5

$$P_{\omega c} = - \frac{(\omega_o - \omega)\, \omega_o N P_o^2 E_\omega}{3\hbar\omega[\gamma^2 + (\omega_o - \omega)^2]} \quad , \qquad 4.2\text{-}6$$

$$P_{\omega s} = \frac{\gamma\omega_o N P_o^2 E_\omega}{3\hbar\omega[\gamma^2 + (\omega_o - \omega)^2]} \quad . \qquad 4.2\text{-}7$$

Taking $\underline{k}_\omega$ to be along the z axis, 2.3.2-20 and 21 become, using 4.2-6 and 7,

$$\left[n(\omega) - 1 + \frac{c}{\omega}\frac{\partial\varphi}{\partial z}\right] E_\omega = \frac{-(\omega_o - \omega)\omega_o N P_o^2 E_\omega}{6\epsilon_o \hbar\omega[\gamma^2 + (\omega_o - \omega)^2]} \quad , \qquad 4.2\text{-}8$$

$$\frac{\partial E_\omega}{\partial z} = \frac{\gamma\omega_o N P_o^2 E_\omega}{6\epsilon_o c\hbar[\gamma^2 + (\omega_o - \omega)^2]} \quad . \qquad 4.2\text{-}9$$

If we define

$$\alpha_o = \frac{P_o^2 N \omega_o}{6\gamma\epsilon_o c\hbar} \qquad 4.2\text{-}10$$

(with dimensions length^{-1}) and neglect $\partial\varphi/\partial z$ in 4.2-8, the latter and 4.2-9 become

$$n(\omega) = 1 - \frac{c}{\omega}\alpha_o \frac{\gamma(\omega_o - \omega)}{\gamma^2 + (\omega_o - \omega)^2} \quad , \qquad 4.2\text{-}11$$

$$\frac{\partial E_\omega}{\partial z} = \frac{\gamma^2}{\gamma^2 + (\omega_o - \omega)^2}\alpha_o E_\omega \quad . \qquad 4.2\text{-}12$$

4.2-12 gives

$$E_\omega(z) = E_\omega(o)\, e^{\alpha_o(\omega)z} \quad , \qquad 4.2\text{-}13$$

where $E_\omega(z) = E_\omega(o)$ at $z = 0$ and we have defined

$$\alpha_o(\omega) = \alpha_o \frac{\gamma^2}{\gamma^2 + (\omega_o - \omega)^2} \quad . \qquad 4.2\text{-}14$$

4.2-14 and 11 are of course the well-known gain per unit length and index of refraction characteristic of an inverted, nautrally broadened line, where 2γ is the total halfwidth of the Lorentzian curve of $\alpha_o(\omega)$ versus ω .

The neglect of $\partial\varphi/\partial z$ in 4.2-8 amounts to grouping it with $n(\omega)$ to give an effective index of refraction, and of course any linear change of φ with distance is equivalent to an index of refraction. Since the field amplitude are slowly varying, we expect other changes in φ to be quite small, and $\partial\varphi/\partial z$ small enough to be neglected. In fact, from 4.28 we see that $\varphi(z)$ could only have a linear dependence on z , and this will be included in the index of refraction. With nonlinear effects, we may have cases where we need to take $\partial\varphi/\partial z \neq 0$.

4.3 Maxwellian Velocity Distribution

For stationary atoms we have seen that the amplitude of each component of the field increases exponentially with the distance it has traveled in the medium. The same behavior will of course be found for the case of moving atoms, only with a different exponent and index of refraction. With a Maxwellian velocity distribution, we will obtain the Doppler gain curve and the corresponding index of refraction.

Putting 4.1-1 into 3.3-27, we find

$$\rho_{ab}(\underline{r},t) = \frac{\omega_o N}{2i\hbar} \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega} \right) e^{i(\underline{k} \cdot \underline{r} - \omega t + \varphi)}$$

$$\times \left(\frac{1}{\pi u^2} \right)^{\frac{3}{2}} \int d\underline{v} \frac{e^{-v^2/u^2}}{\gamma + i(\omega_o - \omega + \underline{k} \cdot \underline{v})} , \qquad 4.3\text{-}1$$

where the integral is over all velocity space. With one of a set of Cartesian axes along $\underline{k}$, the integrations over velocities perpendicular to this direction can be easily carried out leaving for the second line of 4.3-1

$$\frac{1}{\sqrt{\pi} u} \int_{-\infty}^{\infty} dV \frac{e^{-V^2/u^2}}{\gamma + i(\omega_o - \omega + kV)} , \qquad 4.3\text{-}2$$

where V is the velocity component along $\underline{k}$. 4.3-2 is a standard integral which arises in the theory of Doppler broadening and which cannot be analytically evaluated (24). Defining

$$a = \frac{\gamma}{ku} , \; x = \frac{\omega_o - \omega}{ku} , \qquad 4.3\text{-}3$$

4.3-2 can be expressed as

$$\frac{2}{ku}\, e^{(a + ix)^2} \int_{a+ix}^{\infty + ix} dt\; e^{-t^2} \quad , \qquad 4.3\text{-}4$$

as shown in Appendix I. 4.3-4 can be expressed in terms of tabulated functions, viz.,

$$\frac{2}{\sqrt{\pi}}\, e^{(a + ix)^2} \int_{a+ix}^{\infty + ix} dt\; e^{-t^2} = w(-x + ia) = w^*(x + ia) \;, \qquad 4.3\text{-}5$$

where $w(z)$ is the Error Function for Complex Arguments (25), defined by

$$w(z) = e^{-z^2}\, \mathrm{erfc}(-iz) = \frac{2}{\sqrt{\pi}}\, e^{-z^2} \int_z^{\infty} e^{-t^2}\, dt \quad . \qquad 4.3\text{-}6$$

For the interesting case $\gamma \ll ku$ $(a \ll 1)$, we have to first order in a , from Appendix I,

$$e^{(a + ix)^2} \int_{a+ix}^{\infty + ix} dt\; e^{-t^2} \cong \frac{\sqrt{\pi}}{2} \left\{ \left[e^{-x^2} - \frac{2a}{\pi}\,(1 - 2xF(x)) \right] - i\left[\frac{2}{\pi}\, F(x) - 2axe^{-x^2} \right] \right\} . \qquad 4.3\text{-}7$$

From 4.3-7, we see that for $a = 0$

$$w^*(x + io) = e^{-x^2} - \frac{2i}{\sqrt{\pi}}\, F(x) \quad . \qquad 4.3\text{-}8$$

With the above definitions, 4.3-1 becomes

$$\rho_{ab}(\underline{r},t) = \frac{\omega_o N\sqrt{\pi}a}{2i\hbar\gamma} \sum_{\omega} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right) w^*(x + ia)\ e^{i(\underline{k} \cdot \underline{r} - \omega t + \varphi)}, \quad 4.3\text{-}9$$

where x, a are defined by 4.3-3. Putting 4.3-9 into 3.2-16, we find the polarization coefficients

$$P_{\omega c} = \frac{\omega_o N\sqrt{\pi}a\, P_o^2 E_\omega}{3\hbar\gamma\omega}\ \text{Im}\ w^*(x + ia) \quad , \qquad 4.3\text{-}10$$

$$P_{\omega s} = \frac{\omega_o N\sqrt{\pi}a\, P_o^2 E_\omega}{3\hbar\gamma\omega}\ \text{Re}\ w^*(x + ia) \quad , \qquad 4.3\text{-}11$$

where $\text{Re}\ w^*(x + ia)$ and $\text{Im}\ w^*(x + ia)$ are the real and imaginary parts of $w^*(x + ia)$ and the previous results for the integration over directions of $\underline{P}_o$ have been used. Putting 4.3-10 and 11 into the field equations of motion 2.3.2-20 and 21, and taking $\underline{k}_\omega = k_\omega \underline{e}_z$ and $\partial\varphi/\partial z = 0$ as before, we find the results

$$\frac{\partial E_\omega}{\partial z} = \frac{\omega_o N\sqrt{\pi}a\, P_o^2 E_\omega}{6\epsilon_o c\hbar\gamma}\ \text{Re}\ w^*(x + ia) = \alpha\ \text{Re}\ w^*(x + ia)\ E_\omega \quad , \qquad 4.3\text{-}12$$

$$n(\omega) = 1 + \frac{\omega_o N\sqrt{\pi}a\, P_o^2}{6\epsilon_o \omega\hbar\gamma}\ \text{Im}\ w^*(x + ia) = 1 + \frac{c}{\omega}\,\alpha\ \text{Im}\ w^*(x + ia) \ , \quad 4.3\text{-}13$$

where

$$\alpha = \alpha_o \sqrt{\pi}\, a \qquad 4.3\text{-}14$$

and α_o is defined by 4.2-10. For $a \ll 1$, 4.3-14 and 15 become

$$\frac{\partial E_\omega}{\partial z} = \alpha\left[e^{-x^2} - \frac{2a}{\sqrt{\pi}}\,(1 - 2xF(x))\right] E_\omega = \alpha(\omega)\ E_\omega \qquad 4.3\text{-}15$$

$$n(\omega) = 1 - \frac{c}{\omega}\,\alpha\left[\frac{2}{\sqrt{\pi}}\,F(x) - 2axe^{-x^2}\right] \ . \qquad 4.3\text{-}16$$

$\alpha(\omega)$ as defined by 4.3-15 (for $a \ll 1$) is of course the "Gaussian with Lorentzian wings" line shape familiar from the theory of Doppler line broadening (26). $\alpha(\omega)$ and $n(\omega)$ are plotted in Figure 1 for representative values of a, along with $\alpha_o(\omega)$ and $n_o(\omega)$ from the case of stationary atoms.

4.4 Scaling Laws and the Condition for Linearity

In Chapter 5 we will see that the condition for linearity, i.e. that the lowest order nonlinear effects be small compared to the linear effects, requires that the total field amplitude remain small compared to a field E_o, where

$$E_o^2 = \frac{\gamma_a \gamma_b \hbar^2}{P_o^2} \ . \qquad 4.4\text{-}1$$

In other words, the power per unit area must be small compared to

$$P_s = \epsilon_o c E_o^2 \ . \qquad 4.4\text{-}2$$

Since γ_a and γ_b are just the reciprocals of the lifetimes of the states $|a)$ and $|b)$, respectively, we can estimate values for P_s using

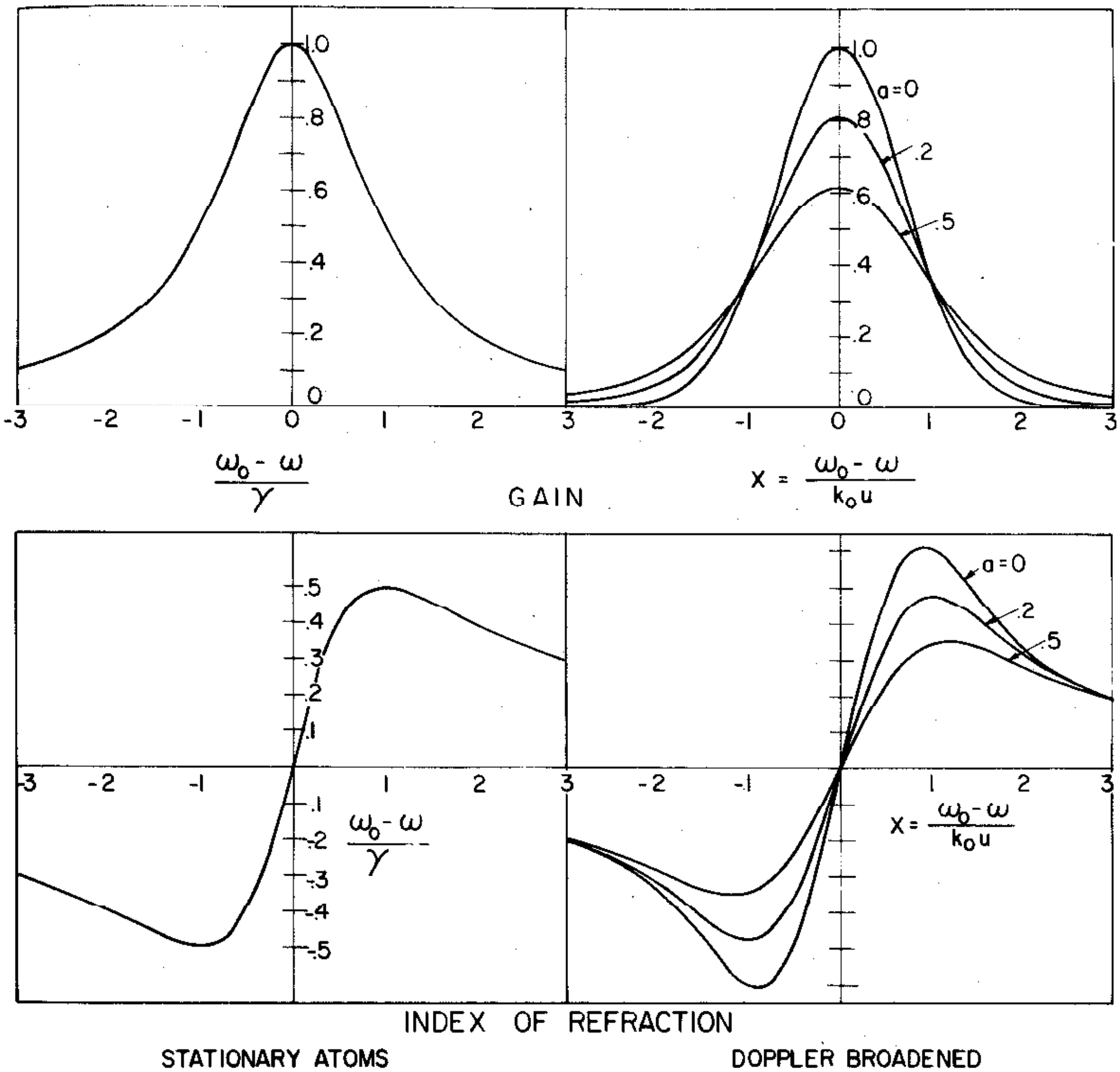


FIGURE I LINEAR GAIN AND INDEX OF REFRACTION FOR STATIONARY AND MOVING ATOMS, AS FUNCTIONS OF FREQUENCY

$$P_o^2 \simeq 2 \times 10^{-58} \quad \text{(MKS)}$$

and some calculated and experimental values of lifetimes. For some lines of interest, we have

.633 micron,	Ne :	$\gamma_a \simeq 10^7$	,	$\gamma_b = 8.3 \times 10^7$ (2)
1.15 "	Ne :	$\gamma_a = 10^7$ (2)	,	$\gamma_b = 8.3 \times 10^7$ (2)
3.39 "	Ne :	$\gamma_a \simeq 10^7$	,	$\gamma_b = 9 \times 10^6$ (27)
3.51 "	Xe :	$\gamma_a = 7.4 \times 10^5$ (27)	,	$\gamma_b = 2.3 \times 10^7$ (27)

We have assumed that γ_a for the upper states of the .633 (3.39) and 1.15 micron lines are about the same, since essentially the same type of state is involved. With these values, we find from 4.4-1 and 2

$$P_s \; .633 \simeq 11 \quad \text{mw/cm}^2$$
$$P_s \; 1.15 \simeq 11 \quad "$$
$$P_s \; 3.39 \simeq 1.2 \quad "$$
$$P_s \; 3.51 \simeq .22 \quad " \quad .$$

Some experimentally observed values are

$$P_s \; 1.15 \text{ (observed)} \simeq 5.9 \quad \text{mw/cm}^2 \quad (28)$$
$$P_s \; 3.39 \quad " \quad \simeq 1 \quad " \quad (16)$$
$$P_s \; 3.51 \quad " \quad \simeq .2 \quad " \quad (29) \; .$$

Thus there is at least order of magnitude agreement.

An alternative assumption is that γ_a arises only from the transition between states $|a)$ and $|b)$. For this case, we can use the result (30)

$$\frac{\gamma_a}{P_o^2} \simeq \frac{\omega_o^3}{3\pi\epsilon_o c^3 \hbar} \quad , \qquad 4.4\text{-}3$$

which with 4.4-1 and 2 gives

$$P_s \cong \frac{\gamma_b}{\lambda^3}\, 8 \times 10^{-8} \text{ mw/cm}^2 \quad , \qquad 4.4\text{-}4$$

where λ is the wavelength expressed in microns. Using 4.4-4 and the above experimental values for P_s, we calculate

$$\begin{aligned} \gamma_b\, 3.39 &\cong 7 \times 10^8 \text{ sec}^{-1} \\ \gamma_b\, .633 = \gamma_b\, 1.15 &\cong 10^8 \text{ sec}^{-1} \\ \gamma_b\, 3.51 &\cong 10^8 \text{ sec}^{-1} \quad . \end{aligned} \qquad 4.4\text{-}5$$

Since $\gamma_b\, 1.15 = \gamma_b\, .633$, we can use 4.4-5 and 4.4-4 to calculate

$$P_s\, .633 \cong 20 \text{ mw/cm}^2 \quad . \qquad 4.4\text{-}6$$

These values may be more accurate, since the computed values given by Horrigan, et al (27) do not take into account broadening due to collisions and radiation trapping (2), and are less favorable for establishing a large population inversion (2). We will return later to the problem of determining γ_a, γ_b and other parameters from various experimental data.

From the above, we see that

$$P_s \propto \gamma_b\, \omega_o^3 \quad , \qquad 4.4\text{-}7$$

so that the saturation power will generally increase rapidly with frequency. Another quantity of interest in the theory is the natural linewidth, defined by

$$(\Delta f)_N = \frac{\gamma_a + \gamma_b}{2\pi} \quad . \qquad 4.4\text{-}8$$

For $\gamma_b \gg \gamma_a$, we see from 4.4-7

$$(\Delta f)_N \propto \frac{P_s}{\omega_o^3} \quad , \qquad 4.4\text{-}9$$

and 4.4-5 gives

$$(\Delta f)_{N3.39} \simeq 110 \quad \text{Mc/sec.}$$

$$(\Delta f)_{N.633} \simeq (\Delta f)_{N1.15} \simeq 16 \quad \text{Mc/sec.} \qquad 4.4\text{-}10$$

$$(\Delta f)_{N3.51} \simeq 16 \quad \text{Mc/sec.}$$

If $(\Delta f)_D$ is the Doppler line total halfwidth (31), we have

$$ku = \frac{2\pi(\Delta f)_D}{2\sqrt{\ln 2}} \quad , \qquad 4.4\text{-}11$$

and therefore for $a = \gamma/ku = .835\ (\Delta f)_N/(\Delta f)_D$ we find

$$a_{3.39} \simeq .26$$

$$a_{1.15} \simeq .028$$

$$a_{.633} \simeq .009 \qquad 4.4\text{-}12$$

$$a_{3.51} \simeq .1$$

using calculated values of $(\Delta f)_D$ (31).

Equation 4.2-10 gives

$$\alpha_o \propto \frac{P_o^2 N \omega_o}{\gamma_a + \gamma_b} \simeq \frac{P_o^2 \lambda_a \omega_o}{\gamma_a \gamma_b}$$

if $\gamma_b \gg \gamma_a$. Using 4.4-1 and 2, we find

$$\alpha_o \propto \frac{\lambda_a \omega_o}{P_s} \quad . \qquad 4.4\text{-}13$$

For $a \ll 1$, we have, using $ku \propto \omega_o \sqrt{T/M}$ and 4.4-4,

$$\alpha = \alpha_o \; a\sqrt{\pi} \propto \frac{\lambda_a}{\omega_o^3} \sqrt{\frac{M}{T}} \quad . \qquad 4.4\text{-}14$$

If we assume constant λ_a and T , 4.4-14 predicts relative values of α for the .633, 1.15, 3.39, 3.51 micron lines

1 : 6 : 160 : 430 ,

while observed gains (32) give the relative values

1 : 6 : 250 : 550 .

The corresponding relative values of α_o are from 4.4-13,

1 : 1.8 : 3.6 : 18 .

The above discussion gives the approximate scaling laws for some quantities of interest, ignoring any "configuration" effects (33), etc. These laws can be of some help in evaluating the potential of a given transition once a few measurements have been made. Most expressions are in terms of the saturation power, P_s , since the latter is probably one of the easiest quantities to measure.

The most interesting result here is that quite small powers can cause strong nonlinear effects for some transitions. We will now continue to study these nonlinear effects, for the case where they are small.

CHAPTER FIVE

LOWEST ORDER NONLINEAR EFFECTS

When the field amplitudes remain small enough that we have only a slight change from linear amplification, it is adequate to use a perturbation expansion of the solutions (12) and keep only the lowest order terms beyond the linear term. As discussed in Chapter 3, this means a correction to the population inversion density which is quadratic in the field amplitudes, and a correction to the polarization which is cubic in the field amplitudes. The approach used in Chapter 3 for solution of the atomic equations of motion is ideal for obtaining these terms, since it only requires putting $\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v}) = NW(\underline{v})$ in 3.3-21 and 28. In this chapter we will be primarily interested in the resulting $\rho_{ab}^{(3)}(\underline{r},t,\underline{v})$ and the corresponding third order polarization and its effect on the behavior of the fields.

In section 5.1 we calculate $\rho_{aa}^{(2)}(\underline{r},t,\underline{v}) - \rho_{bb}^{(2)}(\underline{r},t,\underline{v})$ and $\rho_{ab}^{(3)}(\underline{r},t,\underline{v})$. The latter quantity is used to study lowest order nonlinear effects for stationary atoms in section 5.2 and for a Maxwellian velocity distribution of excited atoms in section 5.3. Section 5.4 gives a discussion of the results.

5.1 The Lowest Order Nonlinear Solutions

From 3.3-21, with $\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v}) = NW(\underline{v})$ in the right hand side, we have

$$\rho_{aa}^{(2)}(\underline{r},t,\underline{v}) - \rho_{bb}^{(2)}(\underline{r},t,\underline{v}) = -\left(\frac{\omega_o}{2\hbar}\right)^2 NW(\underline{v}) \int_0^\infty dt_1 \int_0^\infty dt_2$$

$$\times\left(e^{-\gamma_a t_1} + e^{-\gamma_b t_1}\right) \sum_{\omega,\omega'} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right)$$

$$\times\left[e^{i\Delta + i(\omega - \omega' - (\underline{k} - \underline{k}') \cdot \underline{v})t_1 - [\gamma - i(\omega_o - \omega' + \underline{k}' \cdot \underline{v})]t_2} + c.c.\right]$$

5.1-1

Performing the integrations gives

$$\rho_{aa}^{(2)}(\underline{r},t,\underline{v}) - \rho_{bb}^{(2)}(\underline{r},t,\underline{v}) = -\left(\frac{\omega_o}{2\hbar}\right)^2 NW(\underline{v}) \sum_{\omega,\omega'} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right)$$

$$\times\left\{\frac{e^{i[(\underline{k} - \underline{k}') \cdot \underline{r} - (\omega - \omega')t + \varphi - \varphi']}}{\gamma - i(\omega_o - \omega' + \underline{k}' \cdot \underline{v})} \left[\frac{1}{\gamma_a - i(\omega - \omega' - (\underline{k} - \underline{k}') \cdot \underline{v})}\right.\right.$$

$$\left.\left. + \frac{1}{\gamma_b - i(\omega - \omega' - (\underline{k} - \underline{k}') \cdot \underline{v})}\right] + c.c.\right\} .$$

5.1-2

Putting $\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v})$ in the right hand side of 3.3-27 gives

$$\rho_{ab}^{(3)}(\underline{r},t,\underline{v}) = \left(\frac{\omega_o}{2i\hbar}\right)^3 NW(\underline{v}) \int_0^\infty dt_1 \int_0^\infty dt_2 \int_0^\infty dt_3 \left(e^{-\gamma_a t_2} + e^{-\gamma_b t_2}\right)$$

$$\times \sum_{\omega,\omega',\omega''} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega''}}{\omega''}\right) e^{i[1] - [\gamma + i(\omega_o - \omega + \underline{k} \cdot \underline{v})]t_1}$$

$$\times \left[e^{i\Delta + i[\omega' - \omega'' - (\underline{k}' - \underline{k}'')\cdot \underline{v}](t_1 + t_2) - [\gamma - i(\omega_o - \omega'' + \underline{k}'' \cdot \underline{v})]t_3} + \text{c.c.}\right] , \qquad 5.1\text{-}3$$

where [1] and Δ are given by 3.3-25. Performing the integration gives

$$\rho_{ab}^{(3)}(\underline{r},t,\underline{v}) = \left(\frac{\omega_o}{2i\hbar}\right)^3 NW(\underline{v}) \sum_{\omega,\omega',\omega''} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega''}}{\omega''}\right)$$

$$\times \frac{e^{i[(\underline{k} + \underline{k}' - \underline{k}'') \cdot \underline{r} - (\omega + \omega' - \omega'')t + \varphi + \varphi' - \varphi'']}}{\gamma + i(\omega_o - \omega - \omega' + \omega'' + (\underline{k} + \underline{k}' - \underline{k}'') \cdot \underline{v})}$$

$$\times \left[\frac{1}{\gamma_a - i(\omega' - \omega'' - (\underline{k}' - \underline{k}'') \cdot \underline{v})} + \frac{1}{\gamma_b - i(\omega' - \omega'' - (\underline{k}' - \underline{k}'') \cdot \underline{v})}\right]$$

$$\times \left[\frac{1}{\gamma - i(\omega_o - \omega'' + \underline{k}'' \cdot \underline{v})} + \frac{1}{\gamma + i(\omega_o - \omega' + \underline{k}' \cdot \underline{v})}\right] . \qquad 5.1\text{-}4$$

Equations 5.1-4 is our basic starting point for this chapter. From it we calculate the third order component of the polarization and thereby the lowest order corrections to linear amplification.

5.2 Stationary Atoms

For stationary atoms 5.1-4 gives immediately

$$\rho_{ab}^{(3)}(\underline{r},t) = \left(\frac{\omega_o}{2i\hbar}\right)^3 N \sum_{\omega,\omega',\omega''} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega''}}{\omega''}\right)$$

$$\times \frac{e^{i[(\underline{k} + \underline{k}' - \underline{k}'') \cdot \underline{r} - (\omega + \omega' - \omega'')t + \varphi + \varphi' - \varphi'']}}{\gamma + i(\omega_o - \omega - \omega' + \omega'')}$$

$$\times \left[\frac{1}{\gamma_a - i(\omega' - \omega'')} + \frac{1}{\gamma_b - i(\omega' - \omega'')}\right]\left[\frac{1}{\gamma - i(\omega_o - \omega'')} + \frac{1}{\gamma + i(\omega_o - \omega')}\right] \quad .$$

5.2-1

In this section we treat various cases covered by 5.2-1.

5.2.1 Single Input Frequency

For the case of a single input frequency, the triple summation in 5.2-1 reduces to a single term:

$$\rho_{ab}^{(3)}(\underline{r},t) = \left(\frac{\omega_o}{2i\hbar}\right)^3 N \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)^3 \frac{e^{i(\underline{k} \cdot \underline{r} - \omega t + \varphi)}}{\gamma + i(\omega_o - \omega)}$$

$$\times \frac{2\gamma}{\gamma_a \gamma_b} \, \frac{2\gamma}{\gamma^2 + (\omega_o - \omega)^2} \quad . \qquad 5.2.1\text{-}1$$

In taking the average over all directions of $\underline{P}_o$, using 4.2-4, it is evident that the only component of the polarization will be parallel to $\underline{E}_\omega$. We have for the component of $\underline{P}_o(\underline{P}_o \cdot \underline{E}_\omega)^3$ along $\underline{E}_\omega$:

$$\frac{P_o^4 E_\omega^3}{4\pi} \int_0^{2\pi} d\varphi \int_0^{\pi} d\theta \cos^4\theta \sin\theta = \frac{P_o^4 E_\omega^3}{5} \quad , \qquad 5.2.1\text{-}2$$

with both perpendicular components averaging to zero as in the linear case. Thus we find

$$P_{\omega s}^{(3)} = - \frac{\omega_o^3 N P_o^4 E_\omega^3}{5\omega^3 \hbar^3 \gamma_a \gamma_b} \frac{\gamma^3}{[\gamma^2 + (\omega_o - \omega)^2]^2} \quad , \qquad 5.2.1\text{-}3$$

$$P_{\omega c}^{(3)} = \frac{\omega_o^3 N P_o^4 E_\omega^3}{5\omega^3 \hbar^3 \gamma_a \gamma_b} \frac{\gamma^2(\omega_o - \omega)}{[\gamma^2 + (\omega_o - \omega)^2]} \quad . \qquad 5.2.1\text{-}4$$

If we define E_o by

$$E_o^2 = \hbar^2 \gamma_a \gamma_b / P_o^2 \quad , \qquad 5.2.1\text{-}5$$

5.2.1-3 and 4 become

$$P_{\omega s}^{(3)} = - \frac{\omega_o^3 N P_o^2 E_\omega^3}{5\omega^3 \hbar E_o^2} \frac{\gamma^3}{[\gamma^2 + (\omega_o - \omega)^2]^2} \quad , \qquad 5.2.1\text{-}6$$

$$P_{\omega c}^{(3)} = \frac{\omega_o^3 N P_o^2 E_\omega^3}{5\omega^3 \hbar E_o^2} \frac{\gamma^2(\omega_o - \omega)}{[\gamma^2 + (\omega_o - \omega)^2]^2} \quad . \qquad 5.2.1\text{-}7$$

Putting 5.2.1-6 and 7 into the field equations 2.3.2-20 and 21 and taking $\underline{k}$ to be in the z_ω direction, we find

$$\delta\, n(\omega) = \frac{3}{5}\,\alpha_o\,\frac{c}{\omega}\,\frac{E_\omega^2}{E_o^2}\,\frac{\gamma^3(\omega_o - \omega)}{[\gamma^2 + (\omega_o - \omega)^2]^2}\,, \qquad 5.2.1\text{-}8$$

$$\delta\left(\frac{\partial E_\omega}{\partial z_\omega}\right) = -\frac{3}{5}\,\alpha_o\,\frac{E_\omega^3}{E_o^2}\,\frac{\gamma^4}{[\gamma^2 + (\omega_o - \omega)^2]^2}\,, \qquad 5.2.1\text{-}9$$

where we have taken $\omega_o^2/\omega^2 \simeq 1$ since $\omega_o \simeq \omega$. Comparing 5.2.1-8 and 9 to the linear index of refraction and gain per unit length 4.2-11 and 12, we find to third order:

$$n(\omega) - 1 = \Big[n(\omega) - 1\Big]_{lin}\left(1 - \frac{3}{5}\,\frac{E_\omega^2}{E_o^2}\,\frac{\gamma^2}{\gamma^2 + (\omega_o - \omega)^2}\right), \qquad 5.2.1\text{-}10$$

$$\frac{\partial E_\omega}{\partial z_\omega} = \left(\frac{\partial E_\omega}{\partial z_\omega}\right)_{lin}\left(1 - \frac{3}{5}\,\frac{E_\omega^2}{E_o^2}\,\frac{\gamma^2}{\gamma^2 + (\omega_o - \omega)^2}\right). \qquad 5.2.1\text{-}11$$

Thus for a single input frequency, the effect of the lowest order non-linearity is to introduce intensity-dependent corrections to the index of refraction and gain per unit length.

It is important to note that the amount of the nonlinear correction depends on the size of the field relative to E_o and on how far from resonance the frequency ω is. We see that if $E_\omega^2 \ll E_o^2$ the correction will be small. This condition was used in section 4.4

to define the region of linear amplification; it is also the condition which must be fulfilled if the lowest order correction is to be adequate. As noted in section 4.4, there are many practical cases when this condition is strongly violated.

5.2.2 Two Input Frequencies

For two input frequencies, ω_1, ω_2, we obtain from 5.2-1 the single-frequency corrections at ω_1 and ω_2, and in addition other terms which we will now consider. For convenience we write $\underline{E}_1$ for $\underline{E}_{\omega_1}$, etc.

First, with $\omega = \omega_1$ and $\omega' = \omega'' = \omega_2$ in 5.2-1 we obtain a term in the polarization at ω_1. This gives corrections to $n(\omega_1) - 1$ and $\partial E_1/\partial z_1$ which depend on the field strength at ω_2. The magnitude of this correction will have the same form as the single-frequency correction, except there will be a factor depending on the relative polarization directions of $\underline{E}_1$ and $\underline{E}_2$ which will replace the 1/5 in 5.2.1-10 and 11. This different factor of course comes from the average of the component along $\underline{E}_1$ of

$$\underline{P}_o(\underline{P}_o \cdot \underline{E}_2)^2 \, (\underline{P}_o \cdot \underline{E}_1)$$

over all directions of $\underline{P}_o$. This average replaces the factor 1/5 by

$$\frac{1}{4\pi} \int_0^{2\pi} d\varphi \int_0^{\pi} d\theta \, \sin\theta \, \cos^2\theta \, (\cos^2\theta \, \cos^2 \Theta_{12} + \sin^2\theta \, \cos^2\varphi \, \sin^2 \Theta_{12}$$

$$+ 2 \cos\theta \, \sin\theta \, \cos\varphi \, \cos \Theta_{12} \, \sin \Theta_{12}) \qquad 5.2.2\text{-}1$$

$$= \frac{1 + 2 \cos^2 \Theta_{12}}{15} \; ,$$

where Θ_{12} is the angle between $\underline{E}_1$ and E_2. Thus the correction factor for $n(\omega_1) - 1$ and $\partial E_1/\partial z_1$ becomes

$$\left(1 - \frac{3}{5}\frac{E_1^2}{E_o^2}\frac{\gamma^2}{\gamma^2 + (\omega_o - \omega_1)^2} - \frac{1 + 2\cos^2\Theta_{12}}{5}\frac{E_2^2}{E_o^2}\frac{\gamma^2}{\gamma^2 + (\omega_o - \omega_2)^2}\right) . \qquad 5.2.2\text{-}2$$

Another contribution to the polarization at ω_1 comes from the term in 5.2-1 with $\omega = \omega'' = \omega_2, \omega' = \omega_1$. For this term we find the complicated expressions

$$\delta\left(\frac{\partial E_1}{\partial z_1}\right) = -\alpha_o \frac{1 + 2\cos^2\Theta_{12}}{5}\frac{E_1 E_2^2}{E_o^2}(A_a + A_b) , \qquad 5.2.2\text{-}3$$

$$\delta\, n(\omega_1) = -\alpha_o \frac{c}{\omega_1}\frac{1 + 2\cos^2\Theta_{12}}{5}\frac{E_2^2}{E_o^2}(B_a + B_b) , \qquad 5.2.2\text{-}4$$

where

$$A_a = \frac{\gamma_a\gamma_b\gamma/4}{[\gamma^2 + (\omega_o - \omega_1)^2][\gamma_a^2 + (\omega_1 - \omega_2)^2]}$$

$$\times\left\{\frac{\gamma_a\gamma^2 + \gamma(\omega_o - \omega_1)(\omega_o - \omega_2) - (\omega_1 - \omega_2)^2\gamma}{\gamma^2 + (\omega_o - \omega_2)^2} + \frac{\gamma_a\gamma^2 - \gamma_a(\omega_o - \omega_1)^2 + 2\gamma(\omega_1 - \omega_2)(\omega_o - \omega_1)}{\gamma^2 + (\omega_o - \omega_1)^2}\right\} \qquad 5.2.2\text{-}5$$

$$B_a = \frac{\gamma_a \gamma_b \gamma/4}{[\gamma^2 + (\omega_o - \omega_1)^2][\gamma_a^2 + (\omega_1 - \omega_2)^2]}$$

$$\times \left\{ \frac{(\omega_1 - \omega_2)[\gamma\gamma_a + \gamma^2 + (\omega_o - \omega_1)(\omega_o - \omega_2)]}{\gamma^2 + (\omega_o - \omega_2)^2} + \frac{(\omega_1 - \omega_2)[\gamma^2 - (\omega_o - \omega_1)^2] - 2\gamma\gamma_a(\omega_o - \omega_1)}{\gamma^2 + (\omega_o - \omega_1)^2} \right\}$$

5.2.2-6

and A_b, B_b are obtained by replacing γ_a with γ_b in 5.2.2-5 and 6, respectively. If

$$\omega_1 \to \omega_2 \to \omega_o$$

we find

$$A_a + A_b \to 1$$

$$B_a + B_b \to - \frac{(\omega_o - \omega_1)}{\gamma} + 0 \left(\frac{\omega_1 - \omega_2}{\gamma} \right)$$

as expected. The important characteristic of this term in the polarization is that a factor $1/\gamma_a + 1/\gamma_b$ has been replaced by

$$\frac{1}{\gamma_a - i(\omega_1 - \omega_2)} + \frac{1}{\gamma_b - i(\omega_1 - \omega_2)} ,$$

so that this term decreases more rapidly as $|\omega_1 - \omega_2|$ increases.

The corrections to the gain per unit length and index of refraction at ω_2 are obtained by exchanging indices 1 and 2 in the expressions above for these quantities at ω_1 .

We have thus far dealt with the component of the polarization, at a given frequency, along the field at that frequency, assuming this field to be linearly polarized. In general there will also be components of the polarization along the two perpendicular directions, i.e., along $\underline{E}_1 \times \underline{k}_1$ and $\underline{k}_1$. In Chapter 2 we argued that there should be only a negligible field along $\underline{k}_1$. In section 5.4 we will show that this longitudinal field is of order $\alpha_o\, c/\omega \sim 10^{-6}$ compared to the field in the direction $\underline{E}_1 \times \underline{k}_1$, even though the polarizations are of the same order of magnitude. Since even the field along $\underline{E}_1 \times \underline{k}_1$ will be quite small compared to E_1, the field along $\underline{k}_1$ will be extremely small and will be neglected.

In order to calculate the averaged component of $\underline{P}$ along $\underline{E}_1 \times \underline{k}_1$, we need to define the various directions in the coordinate system used for these calculations. We had chosen $\underline{E}_2$ to be along the z-axis and $\underline{E}_1$ to be in the x-z plane, making an angle Θ_{12} with $\underline{E}_2$. Thus $\underline{k}_2$ is in the x-y plane, and can be defined by the angle Φ_2 made with the x-axis. $\underline{k}_1$ lies in the plane containing the y-axis and making an angle Θ_{12} with the x-axis. If we define a direction in this plane by the direction of the projection of $\underline{E}_2$ into it (or the x-axis when $\Theta_{12} = 0$), the $\underline{k}_1$ direction is defined by the angle Φ_1 made with this direction. If $\underline{e}_x$, $\underline{e}_y$, $\underline{e}_z$ are the unit vectors along the x,y,z axes, respectively, $\underline{e}_1$, $\underline{e}_2$ are the unit vectors along $\underline{E}_1$, $\underline{E}_2$, respectively, and $\underline{e}_1'$, $\underline{e}_2'$ are the unit vectors along the directions $\underline{E}_1 \times \underline{k}_1$, $\underline{E}_2 \times \underline{k}_2$, respectively, we find

$$\underline{e}_2 = \underline{e}_z \quad ,$$

$$\underline{e}_1 = \underline{e}_z \cos \Theta_{12} - \underline{e}_x \sin \Theta_{12} \quad ,$$

$$\underline{k}_1 = k_1 \underline{e}_x \cos \Theta_{12} \cos \Phi_1 + k_1 \underline{e}_y \sin \Phi_1 + k_1 \underline{e}_z \sin \Theta_{12} \cos \Phi_1 \quad ,$$

$$\underline{k}_2 = k_2 \underline{e}_x \cos \Phi_2 + k_2 \underline{e}_y \sin \Phi_2 \quad ,$$

$$\underline{e}_2' = -\underline{e}_x \sin \Phi_2 + \underline{e}_y \cos \Phi_2 \quad ,$$

$$\underline{e}_1' = \underline{e}_x \cos \Theta_{12} \sin \Phi_1 - \underline{e}_y \cos \Phi_1 + \underline{e}_z \sin \Theta_{12} \sin \Phi_1 \quad .$$

5.2.2-7

With these definitions and 4.2-4, we find the average of the component of

$$\underline{P} = \underline{P}_o \, (\underline{P}_o \cdot \underline{E}_2)^2 \, (\underline{P}_o \cdot \underline{E}_1)$$

along $\underline{e}_1'$ to be

$$P_o^4 E_2^2 E_1 \, \frac{\sin 2\Theta_{12} \sin \Phi_1}{15} \quad , \qquad 5.2.2\text{-}8$$

and the averaged component along $\underline{k}_1$ to be

$$P_o^4 E_2^2 E_1 \, \frac{\sin 2\Theta_{12} \cos \Phi_1}{15} \quad . \qquad 5.2.2\text{-}9$$

Thus we find for the component of $\underline{P}$ along $\underline{e}_1'$

$$\frac{\omega_1}{2\epsilon_o c} P_1'(\underline{r},t) = - \frac{\sin 2\,\Theta_{12} \sin \Phi_1}{5} \alpha_o \frac{E_2^2 E_1}{E_o^2}$$

$$\times \left[S_1 \sin(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1) + C_1 \cos(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1) \right] \quad ,$$

5.2.2-10

where

$$S_1 = \frac{\gamma^4}{\left[\gamma^2 + (\omega_o - \omega_1)^2\right]\left[\gamma^2 + (\omega_o - \omega_2)^2\right]} + A_a + A_b \, , \quad 5.2.2\text{-}11$$

$$C_1 = \frac{-\gamma^3 (\omega_o - \omega_1)}{\left[\gamma^2 + (\omega_o - \omega_1)^2\right]\left[\gamma^2 + (\omega_o - \omega_2)^2\right]} + B_a + B_b \, , \quad 5.2.2\text{-}12$$

and A_a , etc., are the same as in 5.2.2-3 and 4.

Since there is initially no field along $\underline{e}_1'$ at frequency ω_1, the polarization $P_1'(\underline{r},t)$ will induce a field E_1' which is out of phase. If we write $P_1'(\underline{r},t)$ in the form

$$\frac{\omega_1}{2\epsilon_o c} P_1'(\underline{r},t) = - \frac{\sin 2\,\Theta_{12} \sin \Phi_1}{5} \alpha_o \frac{E_2^2 E_1}{E_o^2}$$

5.2.2-13

$$\times S_1' \sin(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1 + \varphi_1') \quad ,$$

then

$$S_1' = \sqrt{S_1^2 + C_1^2} \quad ,$$

$$\varphi_1' = \tan^{-1} \frac{C_1}{S_1} \quad ,$$

5.2.2-14

and the field along $\underline{e}_1'$ with frequency ω_1 has the form

$$E_1' \cos(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1 + \varphi_1') \quad , \qquad 5.2.2\text{-}15$$

so that the new field along $\underline{e}_1'$ leads the original field along $\underline{e}_1$ by a phase angle φ_1' .

If we neglect the nonlinear corrections to the gain per unit length and index of refraction at ω_1 , the amplitude E_1' satisfies

$$\frac{\partial E_1'}{\partial z_1} = \alpha_o E_1' \frac{\gamma^2}{\gamma^2 + (\omega_o - \omega_1)^2} - \frac{\sin 2\,\Theta_{12} \sin \Phi_1}{5}\, \alpha_o \frac{E_2^2 E_1}{E_o^2} S_1' \, , \qquad 5.2.2\text{-}16$$

with $E_1' = 0$ at $z_1 = 0$. If $E_2 = E_{20}$ and $E_1 = E_{10}$ at $z_1 = 0$, we can integrate 5.2.2-16 to give

$$E_1'(z_1) = -\sin 2\,\Theta_{12} \sin \Phi_1 \frac{\left[\gamma^2 + (\omega_o - \omega_2)^2\right]}{10\gamma^2 \cos\alpha} S_1'$$

$$\times \frac{E_{20}^2 E_{10}}{E_o^2} \left(e^{2\alpha_o(\omega_2) z_1 \cos\alpha} - 1\right) e^{\alpha_o(\omega_1) z_1} \quad , \qquad 5.2.2\text{-}17$$

where

$$\alpha_o(\omega) = \frac{\alpha_o\, \gamma^2}{\gamma^2 + (\omega_o - \omega)^2} \quad ,$$

and α is the angle between $\underline{k}_1$ and $\underline{k}_2$, i.e.,

$$\cos\alpha = \frac{\underline{k}_1 \cdot \underline{k}_2}{k_1 k_2} = \cos\Theta_{12} \cos\Phi_1 \cos\Phi_2 + \sin\Phi_1 \sin\Phi_2 \quad .$$

The corresponding field at ω_2 is $\underline{E}'_2$ in the direction $\underline{e}'_2$. The magnitude of this field is obtained from 5.2.2-17 by interchanging the subscripts 1 and 2.

With $\omega = \omega' = \omega_2$, $\omega'' = \omega_1$ in 5.2-1, we obtain terms in the polarization at frequency $2\omega_2 - \omega_1 \equiv \omega_{21}$. These will induce a field at the same frequency with components $\underline{E}_{21}$, $\underline{E}'_{21}$ in two directions perpendicular to the direction of propagation, $\underline{k}_{21}$ and the treatment of this effect closely follows that for the introduction of the field $\underline{E}'_1$. The difference arises from the dispersion of the medium, i.e., from the fact that in general

$$\underline{k}_{21} \neq 2\underline{k}_2 - \underline{k}_1 \quad . \qquad 5.2.2\text{-}18$$

Rather than give the complicated general expressions for the unit vectors $\underline{e}_{21}$ and $\underline{e}'_{21}$ perpendicular to $\underline{k}_{21}$, we calculate the average projection of $\underline{P}_o(\underline{P}_o \cdot \underline{E}_2)^2 (\underline{P}_o \cdot E_1)$ along an arbitrary unit vector $\underline{a}$, using 4.2-4. This calculation is carried out exactly as before, and gives the result

$$\frac{P_o^4 E_2^2 E_1}{3} \left(\frac{3a_z \cos \Theta_{12} - a_x \sin \Theta_{12}}{5} \right) = \frac{P_o^4 E_2^2 E_1}{3} \, (GF) \, , \qquad 5.2.2\text{-}19$$

where the quantity in parentheses is a "geometrical factor" which will appear in the following equations.

Putting $\omega = \omega' = \omega_2$ and $\omega'' = \omega_1$ in 5.2-1 gives

$$\rho_{ab}^{(3)}(\underline{r},t) = \left(\frac{\omega_o}{2i\hbar}\right)^3 N \left(\frac{\underline{P}_o \cdot \underline{E}_2}{\omega_2}\right)^2 \left(\frac{\underline{P}_o \cdot \underline{E}_1}{\omega_1}\right)\left[\frac{1}{\gamma_a - i(\omega_2 - \omega_1)} + \frac{1}{\gamma_b - i(\omega_2 - \omega_1)}\right]$$

$$\frac{e^{i\left[(2\underline{k}_2 - \underline{k}_1)\cdot \underline{r} - (2\omega_2 - \omega_1)t + 2\varphi_2 - \varphi_1\right]}}{\gamma + i(\omega_o - 2\omega_2 + \omega_1)}\left[\frac{1}{\gamma - i(\omega_o - \omega_1)} + \frac{1}{\gamma + i(\omega_o - \omega_2)}\right].$$

5.2.2-20

Using 5.2.2-19, we then find the corresponding polarization to be

$$\frac{\omega_{21}}{2\epsilon_o c} P_{21}(\underline{r},t) = -(GF)\,\alpha_o \frac{E_2^2 E_1}{E_o^2}\left\{S'_{21} \sin\left[(2\underline{k}_2 - \underline{k}_1)\cdot \underline{r} - \omega_{21}t + 2\varphi_2 - \varphi_1\right]\right.$$

$$\left. + C'_{21} \cos\left[(2\underline{k}_2 - \underline{k}_1)\cdot \underline{r} - \omega_{21}t + 2\varphi_2 - \varphi_1\right]\right\} .$$

5.2.2-21

where

$$S'_{21} = S'_{21a} + S'_{21b} \quad ,$$

$$C'_{21} = C'_{21a} + C'_{21b} \quad ,$$

$$S'_{21a} = \frac{\gamma_a \gamma_b \gamma / 4}{\left[\gamma^2 + (\omega_o - 2\omega_2 + \omega_1)^2\right]\left[\gamma_a^2 + (\omega_2 - \omega_1)^2\right]}$$

$$\times \left\{\frac{\gamma_a\left[\gamma^2 + (\omega_o - 2\omega_2 + \omega_1)(\omega_o - \omega_1)\right] - 2\gamma(\omega_2 - \omega_1)^2}{\gamma^2 + (\omega_o - \omega_1)^2}\right.$$

$$\left. + \frac{\gamma_a\left[\gamma^2 - (\omega_o - 2\omega_2 + \omega_1)(\omega_o - \omega_2)\right] + \gamma(\omega_2 - \omega_1)(2\omega_o - 3\omega_2 + \omega_1)}{\gamma^2 + (\omega_o - \omega_2)^2}\right\} ,$$

5.2.2-22

$$C'_{21a} = \frac{\gamma_a \gamma_b \gamma/4}{\gamma^2 + (\omega_o - 2\omega_2 + \omega_1)^2 \quad \gamma_a^2 + (\omega_2 - \omega_1)^2}$$

$$\times \left\{ \frac{(\omega_2 - \omega_1)\ 2\gamma\gamma_a + \gamma^2 + (\omega_o - 2\omega_2 + \omega_1)(\omega_o - \omega_1)}{\gamma^2 + (\omega_o - \omega_1)^2} \right.$$

$$\left. + \frac{(\omega_2 - \omega_1)\ \gamma^2 - (\omega_o - 2\omega_2 + \omega_1)(\omega_o - \omega_2)] - \gamma\gamma_a(2\omega_o - 3\omega_2 + \omega_1)}{\gamma^2 + (\omega_o - \omega_2)^2]} \right\}$$

5.2.2-23

and S'_{21b} and C'_{21b} are 5.2.2-22 and 23 with γ_a and γ_b interchanged.

The field induced by the polarization 5.2.2-21 will have the form

$$E_{21}(\underline{r},t) = E_{21}\ \cos[\underline{k}_{21} \cdot \underline{r} - \omega_{21}t + \varphi_{21}(\underline{r})] \quad . \qquad 5.2.2\text{-}24$$

As discussed before, the initial phase $\varphi_{21}(0)$ must be such that the polarization is initially out of phase with the field. Because of 5.2.2-18 the polarization will not remain out of phase with the field, but will have a component in phase with the field. This in-phase component will induce phase fluctuations in the field, and these are of course strongly correlated with the amplitude fluctuations due to the changing out-of-phase component of the polarization.* We now proceed

* According to 2.3.2-20, the effect of an in-phase component of the polarization can be interpreted as contributing to either $[n(\omega) - 1]$ or $c/\omega\ \partial\varphi/\partial z$. Since the phase change due to the polarization now being considered will in general not increase smoothly with z, it is preferable to study the actual phase rather than some equivalent index of refraction.

to calculate these effects. In the following calculations we will drop the subscript 21 so that $\underline{k}$ means $\underline{k}_{21}$, z means z_{21} , etc. We will also use the notation $\cos(k,k_1)$ to mean the cosine of the angle between $\underline{k}$ and $\underline{k}_1$, etc., i.e.,

$$\cos(k,k_1) = \frac{\underline{k} \cdot \underline{k}_1}{kk_1} \quad . \qquad 5.2.2\text{-}24$$

We define

$$\Delta\underline{k} = \underline{k} - (2\underline{k}_2 - \underline{k}_1) \quad , \qquad 5.2.2\text{-}25$$

so that

$$(\Delta k)_z = k - |2\underline{k}_2 - \underline{k}_1| \cos(k, 2k_2 - k_1) \quad . \qquad 5.2.2\text{-}26$$

In order to find the initial phase of the field, we rewrite 5.2.2-21 as a sine wave, i.e.,

$$\frac{\omega}{2\epsilon_o c} P(\underline{r},t) = -(GF)\,\alpha_o \frac{E_2^2 E_1}{E_o^2} S_{21} \sin\left[(2\underline{k}_2 - \underline{k}_1)\cdot \underline{r} - \omega t + 2\varphi_2 - \varphi_1 + \varphi'\right]$$

5.2.2-27

where

$$S_{21} = \sqrt{S_{21}'^2 + C_{21}'^2} \quad ,$$

$$\varphi' = \tan^{-1} \frac{C_{21}'}{S_{21}'} \quad . \qquad 5.2.2\text{-}27$$

Figure 2 shows a plot of S_{21}', C_{21}', and S_{21} as functions of frequency.

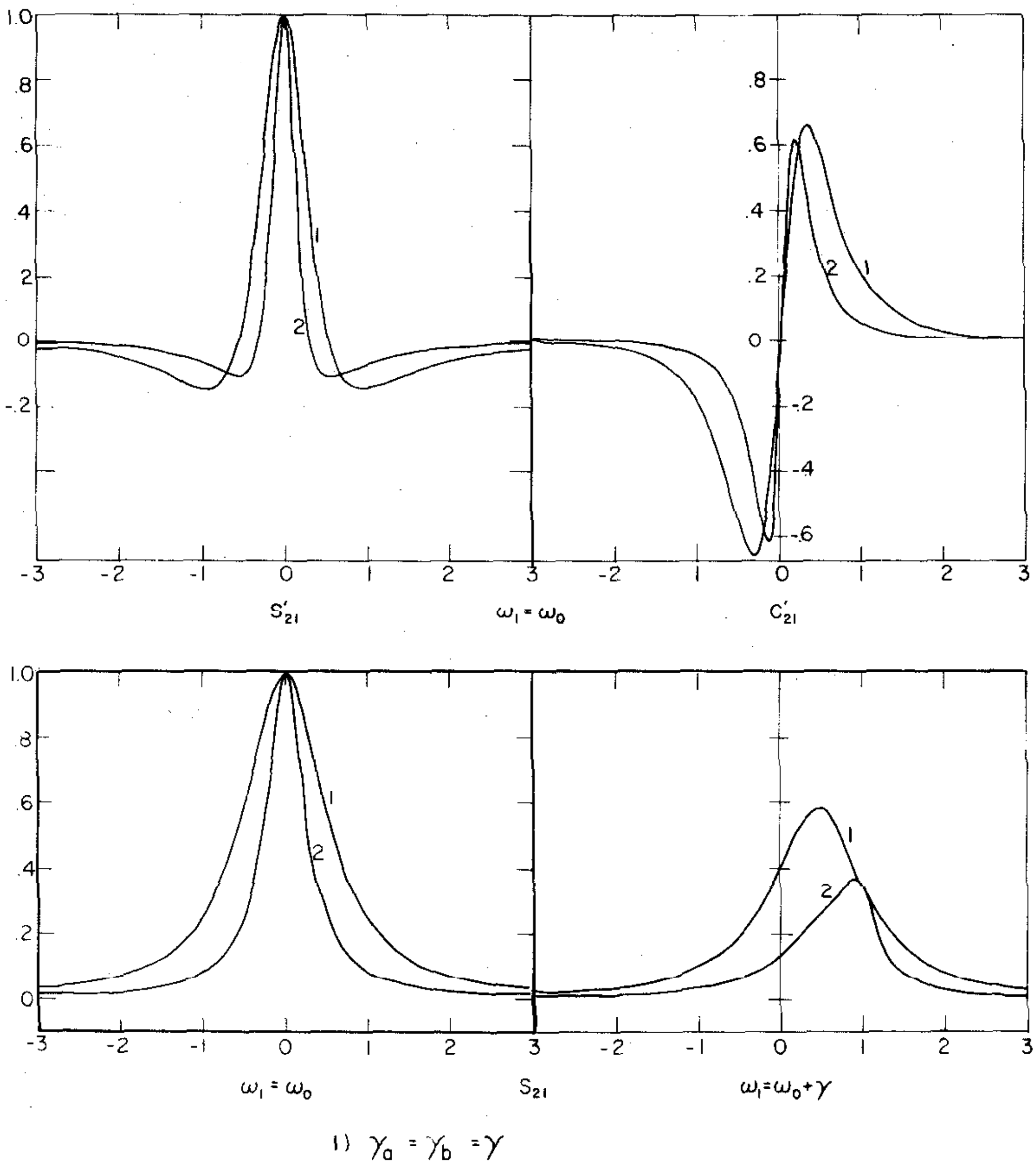


FIGURE 2 LOWEST ORDER INDUCED POLARIZATION AND GAIN AT $2\omega_2-\omega_1$ DUE TO WAVES AT ω_1 AND ω_2

The argument of the sine can be written as

$$\underline{k} \cdot \underline{r} - \Delta\underline{k} \cdot \underline{r} - \omega t + 2\varphi_2 - \varphi_1 + \varphi'$$

$$= kz - \Delta k_z z - \omega t + 2\varphi_2 - \varphi_1 + \varphi' \quad ,$$

taking $x = y = 0$, which only affects the relative phase. Thus at $z = 0$ we must have

$$\varphi(0) = 2\varphi_2 - \varphi_1 + \varphi' \quad . \qquad 5.2.2\text{-}27$$

We can now write the polarization in the form

$$\frac{\omega}{2\epsilon_o c} P(z,t) = -(GF)\,\alpha_o \frac{E_2^2 E_1}{E_o^2} S_{21} \sin\left[kz - \omega t + \varphi - (\Delta k_z z + \varphi - \varphi(o))\right]$$

$$= -(GF)\,\alpha_o \frac{E_2^2 E_1}{E_o^2} S_{21} \left\{ \sin(kz - \omega t + \varphi)\cos(\Delta k_z z + \varphi - \varphi(o)) - \cos(kz - \omega t + \varphi)\sin(\Delta k_z z + \varphi - \varphi(o)) \right\} \quad ,$$

$$5.2.2\text{-}28$$

giving

$$\frac{\omega P_{\omega s}}{2\epsilon_o c} = -(GF)\,\alpha_o \frac{E_2^2 E_1}{E_o^2} S_{21} \cos(\Delta k_z z + \varphi - \varphi(o)) \quad , \qquad 5.2.2\text{-}28$$

$$\frac{\omega P_{\omega c}}{2\epsilon_o c} = (GF)\,\alpha_o \frac{E_2^2 E_1}{E_o^2} S_{21} \sin(\Delta k_z z + \varphi - \varphi(o)) \quad . \qquad 5.2.2\text{-}29$$

If we neglect the nonlinear corrections to the linear index of refraction and gain per unit length, we can write the equations of motion for E and φ , using 2.3.2-20 and 21, as

$$\frac{\partial E}{\partial z} = \alpha_o(\omega)E - (GF)\, \alpha_o \frac{E_2^2 E_1}{E_o^2} S_{21} \cos(\Delta k_z z + \varphi(z) - \varphi(o)) \quad , \qquad 5.2.2\text{-}30$$

$$E \frac{\partial \varphi}{\partial z} = + (GF)\, \alpha_o \frac{E_2^2 E_1}{E_o^2} S_{21} \sin(\Delta k_z z + \varphi(z) - \varphi(o)) \quad , \qquad 5.2.2\text{-}31$$

where

$$E_2(z) - E_{20}\, e^{\alpha_o(\omega_2)\cos(k,k_2)z} \quad ,$$

$$E_1(z) = E_{10}\, e^{\alpha_o(\omega_1)\cos(k,k_1)z} \quad . \qquad 5.2.2\text{-}32$$

Equations 5.2.2-30 and 31 are a set of two coupled differential equations. Their solution is found relatively easily by a transformation to the complex function

$$\mathcal{E}(z) = E(z)\, e^{i\varphi(z)} \quad . \qquad 5.2.2\text{-}33$$

We find directly

$$e^{-i\varphi} \frac{\partial \mathcal{E}}{\partial z} = \frac{\partial E}{\partial z} + iE \frac{\partial \varphi}{\partial z} = \alpha_o(\omega)E - (GF)\, \alpha_o \frac{E_2^2 E_1}{E_o^2} S_{21}\, e^{-i(\Delta k_z z + \varphi(z) - \varphi(o))} ,$$

5.2.2-34

or

$$\frac{\partial \mathcal{E}}{\partial z} = \alpha_o(\omega)\, \mathcal{E} - (GF)\, \alpha_o \frac{E_2^2 E_1}{E_o^2} S_{21}\, e^{-i(\Delta k_z z - \varphi(o))} \quad . \qquad 5.2.2\text{-}35$$

From 5.2.2-33 we see that

$$E^2 = |\mathcal{E}|^2$$
$$\varphi(z) = \arg \mathcal{E} \quad . \qquad 5.2.2\text{-}36$$

Using 5.2.2-32, the solution of 5.2.2-35 with $E(0) = 0$, is found in a straightforward way to be

$$\mathcal{E}(z) = - e^{\alpha_o(\omega)z} (GF)\, \alpha_o \frac{E_{20}^2 E_{10}}{E_o^2} S_{21}\, e^{i\varphi(o)} \frac{e^{(K - i\Delta k_z)z} - 1}{K - i\Delta k_z} \qquad 5.2.2\text{-}37$$

where

$$K = 2\alpha_o(\omega_2) \cos(k,k_2) + \alpha_o(\omega_1) \cos(k,k_1) - \alpha_o(\omega) \quad . \qquad 5.2.2\text{-}38$$

Using 5.2.2-37 and 5.2.2-36, we find

$$E^2 = e^{2\alpha_o(\omega)z} \left[(GF)\, \alpha_o \frac{E_{20}^2 E_{10}}{E_o^2} S_{21}\right]^2 \frac{e^{2Kz} + 1 - 2 \cos \Delta k_z z\, e^{Kz}}{K^2 + (\Delta k_z)^2} \quad , \qquad 5.2.2\text{-}39$$

and

$$\varphi(z) = \varphi(o) + \tan^{-1} \left\{ \frac{\Delta k_z \left[e^{Kz} \cos \Delta k_z z - 1\right] - K \sin \Delta k_z z\, e^{Kz}}{K\left[e^{Kz} \cos \Delta k_z z - 1\right] + \Delta k_z \sin \Delta k_z z\, e^{Kz}} \right\} \quad . \qquad 5.2.2\text{-}40$$

With $\Delta k_z = 0$, $\omega = \omega_1$ and the appropriate geometrical factor, we obtain the previous result 5.2.2-17. If we have $\Delta k_z z \ll 1$, we find

$$\varphi(z) \simeq \varphi(o) + \tan^{-1} \frac{\Delta k_z\left[e^{Kz}(1 - Kz) - 1\right]}{K\left[e^{Kz} - 1\right]} \quad . \qquad 5.2.2\text{-}41$$

If in addition we have $Kz \ll 1$, 5.2.2-41 gives

$$\varphi(z) \simeq \varphi(o) - \frac{1}{2}\Delta k_z z \quad , \qquad 5.2.2\text{-}42$$

while if $Kz \gg 1$, we have

$$\varphi(z) \simeq \varphi(o) - \Delta k_z z \quad . \qquad 5.2.2\text{-}43$$

If Δk_z is large enough for $\cos \Delta k_z$ to go through several cycles in the region of interest and applicability, we see that the quantity in curley brackets in 5.2.2-40 fluctuates between positive and negative infinity. Setting the denominator equal to zero gives

$$\cos \Delta k_z z - e^{-Kz} + \frac{\Delta k_z}{K} \sin \Delta k_z z = 0 \quad . \qquad 5.2.2\text{-}$$

For $Kz \gg 1$, this becomes

$$\tan \Delta k_z z = - \frac{K}{\Delta k_z} \quad ,$$

which occurs twice during each cycle of $\cos \Delta k_z z$. Thus we expect that $\varphi(z) - \varphi(o)$ will increase uniformly with z , at least for $Kz \gg 1$. This behavior can be seen in Figure 3, where the relative E^2 and $\varphi(z) - \varphi(o)$ are plotted for some (small) values of Δk_z . For larger values of Δk_z , we expect more fluctuations in $\varphi(z) - \varphi(o)$

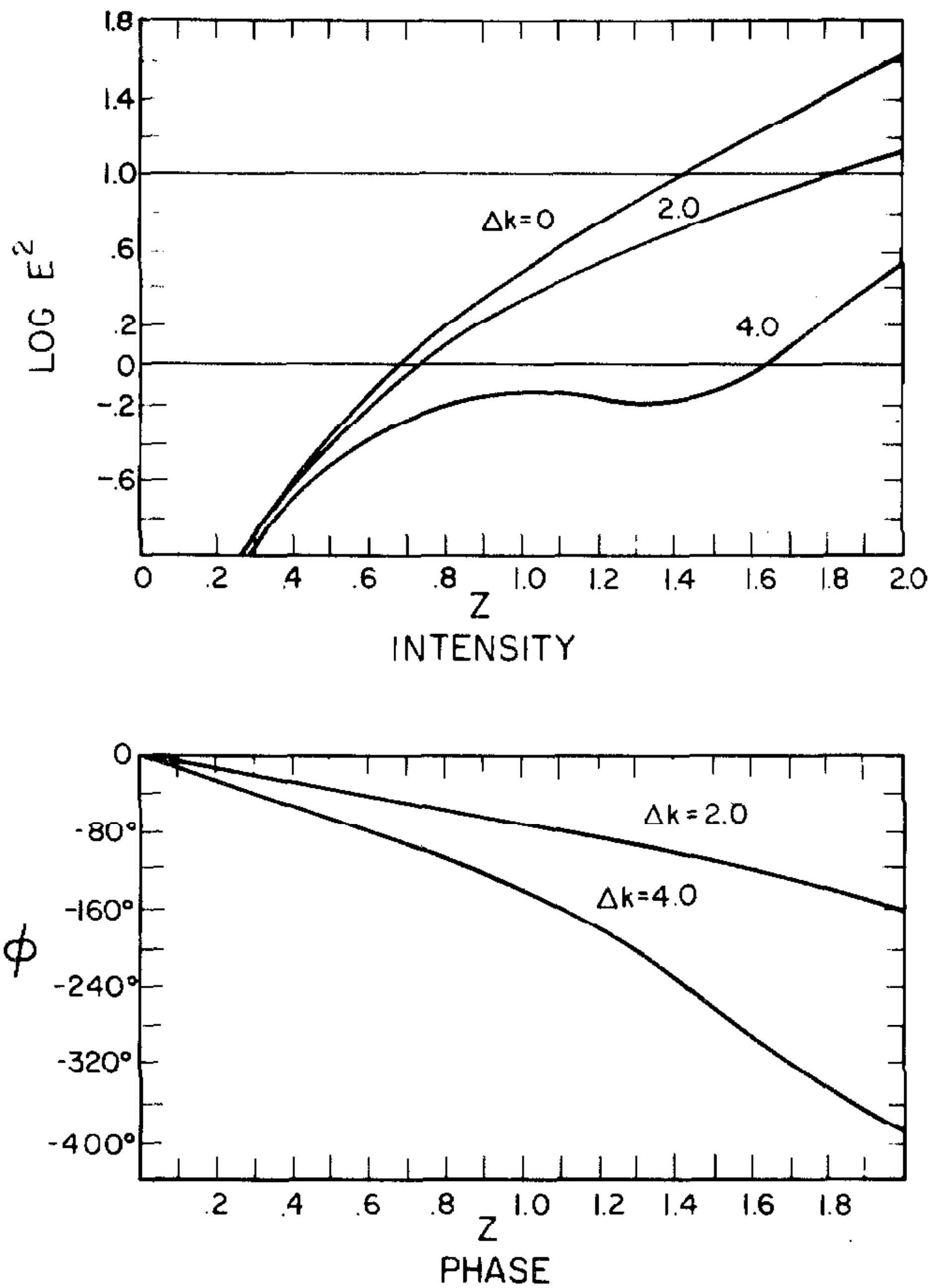


FIGURE 3 INTENSITY AND PHASE AS A FUNCTION OF Z FOR K=1 AND VARIOUS AMOUNTS OF MISMATCH

near $z = 0$.

From 5.2.2-39 we see that for $Kz \gg 1$, the intensity E^2 depends on Δk_z as

$$\frac{1}{K^2 + (\Delta k_z)^2} ,$$

so that the intensity will be a Lorentzian function of angle, with the angular width depending on the gain characteristics of the medium and the directions of propagation of the incident waves.

We will delay further discussion of these results until after we have considered the case of more than two input frequencies and studied similar processes for moving atoms.

5.2.3 Three or More Input Frequencies

With three or more input frequencies, 5.2-1 gives single-frequency terms for each frequency and two-frequency terms for each pair of frequencies. In addition there are terms with $\omega, \omega', \omega''$ all different, e.g., if $\omega = \omega_1$, $\omega' = \omega_2$, $\omega'' = \omega_3$, we have

$$\rho_{ab}^{(3)}(\underline{r},t) = \left(\frac{\omega_o}{2i\hbar}\right)^3 N \left(\frac{\underline{P}_o \cdot \underline{E}_1}{\omega_1}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_2}{\omega_2}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_3}{\omega_3}\right)$$

$$\times \frac{e^{i[(\underline{k}_1 + \underline{k}_2 - \underline{k}_3)\cdot \underline{r} - (\omega_1 + \omega_2 - \omega_3)t + \varphi_1 + \varphi_2 - \varphi_3]}}{\gamma + i(\omega_o - \omega_1 - \omega_2 + \omega_3)}$$

$$\times \left[\frac{1}{\gamma_a - i(\omega_2 - \omega_3)} + \frac{1}{\gamma_b - i(\omega_2 - \omega_3)}\right]\left[\frac{1}{\gamma - i(\omega_o - \omega_3)} + \frac{1}{\gamma + i(\omega_o - \omega_2)}\right]$$

5.2.3-1

giving a polarization and therefore a field at the frequency $\omega_1 + \omega_2 - \omega_3$. The calculation of the field amplitude and phase at this frequency follows in all respects the similar calculation for the field at the frequency $2\omega_2 - \omega_1$ given in the last section. Since there are no new physical results, we will not consider these terms further.

5.3 Maxwellian Velocity Distribution

With a Maxwellian velocity distribution of excited atoms, we must integrate over the velocity spectrum to obtain $\rho_{ab}^{(3)}(\underline{r},t)$. From 4.1-1 and 5.1-4, we have

$$\rho_{ab}^{(3)}(\underline{r},t) = \left(\frac{\omega_o}{2i\hbar}\right)^3 N \sum_{\omega,\omega',\omega''} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega''}}{\omega''}\right)$$

$$\times e^{i[(\underline{k} + \underline{k}' - \underline{k}'') \cdot \underline{r} - (\omega + \omega' - \omega'')t + \varphi + \varphi' - \varphi'']}$$

$$\times \left(\frac{1}{\pi u^2}\right)^{\frac{3}{2}} \int \frac{d\underline{v}\ e^{-v^2/u^2}}{\gamma + i(\omega_o - \omega - \omega' + \omega'' + (\underline{k} + \underline{k}' - \underline{k}'') \cdot \underline{v})}$$

$$\times \left[\frac{1}{\gamma_a - i(\omega' - \omega'' - (\underline{k}' - \underline{k}'') \cdot \underline{v})} + \frac{1}{\gamma_b - i(\omega' - \omega'' - (\underline{k}' - \underline{k}'') \cdot \underline{v})}\right]$$

$$\times \left[\frac{1}{\gamma - i(\omega_o - \omega'' + \underline{k}'' \cdot \underline{v})} + \frac{1}{\gamma + i(\omega_o - \omega' + \underline{k}' \cdot \underline{v})}\right] .$$

5.3-1

By inspection of 5.3-1, we see that the same frequencies will be present in the polarization, and therefore in the fields, as were present for the case of stationary atoms. The difference is that the frequency denominators, which determine the magnitude of the response, contain the effective "interaction" frequencies, e.g., $\omega - \underline{k} \cdot \underline{v}$, rather than the field frequencies, e.g., ω . The result of this is that for a large spread of velocities, so that $ku > \gamma$, the response will extend over a wider range of frequencies. Physically, the interaction frequencies are the (Doppler shifted) frequencies which the atom "sees", and the Doppler shift can move a frequency effectively within the atomic interaction linewidth (the natural linewidth, 2γ) . Thus the results will be qualitatively the same as for stationary atoms, and the response will be weaker and "smeared out" over a wider frequency band, the larger ku is.

There is one qualitative difference between the result 5.3-1 and the usual approach (34) to the treatment of atomic motion. In the usual approach one calculates the response for a stationary atom and then integrates this response over a distribution of center frequencies

$$\omega_o + \underline{k} \cdot \underline{v} \quad . \qquad 5.3\text{-}2$$

Since $ku \ll \omega \approx \omega_o$ in practice, this difference will usually not have a large effect. However, when there are waves traveling in opposite directions, there will be a significant difference in the result. For example, for waves traveling in the same direction we have

$$\underline{k} + \underline{k}' - \underline{k}'' \sim \underline{k} \quad ,$$

while for waves traveling in opposite directions we may have

$$\underline{k} + \underline{k}' - \underline{k}'' \sim 3\underline{k} \quad ,$$

i.e., a Doppler shift which is three times as large as otherwise predicted.

Without rather drastic approximations it is not possible to represent the velocity integrals in 5.3-1 for general $\underline{k}$'s in a useable form. However, for the case where all $\underline{k}$'s are along one direction, we can express these integrals in terms of the single Doppler broadening integral studied in Appendix I. This case includes the possibility of waves traveling in opposite directions, and, for example, for the case of a polarization wave with $\omega = 2\omega_2 - \omega_1$ and $\underline{k} = 2\underline{k}_2 - \underline{k}_1$, we may still take $\underline{k}_{21}$ to be not parallel to $2\underline{k}_2 - \underline{k}_1$. Thus this restriction on the $\underline{k}$'s allows us to consider quantitatively most cases of interest, and we expect that other cases will not differ qualitatively.

With the above restriction on $\underline{k}$'s, the integrals over the two perpendicular directions can be done immediately, and we are left with the requirement of evaluating integrals like

$$I_{a\mp} = \frac{1}{\sqrt{\pi}u} \int_{-\infty}^{\infty} \frac{dV\, e^{-V^2/u^2}}{[\gamma + i(\omega_o - \nu + \underline{K} \cdot \underline{V})][\gamma_a - i(\omega' - \omega'' - (\underline{k}' - \underline{k}'') \cdot \underline{V})]}$$

$$\times \frac{1}{\gamma \begin{Bmatrix} -i(\omega_o - \omega'' + \underline{k}'' \cdot \underline{V}) \\ +i(\omega_o - \omega' + \underline{k}' \cdot \underline{V}) \end{Bmatrix}} \quad , \qquad 5.3\text{-}3$$

where

$$\nu = \omega + \omega' - \omega'' \ ,$$

$$\underline{K} = \underline{k} + \underline{k}' - k'' \ , \qquad 5.3\text{-}4$$

and the notation $\underline{K} \cdot \underline{V}$, etc., is used to preserve the fact that the various $\underline{k}$'s may lie in opposite directions, i.e.,

$$\underline{k}' \cdot \underline{V} = \pm k'V \ .$$

$I_{b\mp}$ is the same as 5.3-3, with γ_a replaced by γ_b . From 5.3-1 we see that we require the integrals

$$I(\omega\omega'\omega'') = I_{a-} + I_{b-} + I_{a+} + I_{b+} \ . \qquad 5.3\text{-}5$$

Expressions for these various integrals are given in Appendix II in terms of the tabulated Doppler broadening integral of Appendix I. The results are often cumbersome in their general form, and in the following we will often use limiting cases and graphical presentation to emphasize their interesting characteristics. The "geometrical factors" are of course identical to those encountered for stationary atoms, and therefore they will be carried over without comment.

5.3.1 Single Input Wave

For a single input wave, the sum in 5.3.1 reduces to a single term, with $\omega = \omega' = \omega''$, and we find from I.1

$$\frac{\omega}{2\epsilon_o c} P_{\omega s} = - \alpha \frac{3}{5} \frac{E_\omega^3}{2E_o^2} \mathrm{Re} \{ \quad \} \ , \qquad 5.3.1\text{-}1$$

$$\frac{\omega}{2\varepsilon_o c} P_{\omega c} = - \alpha \frac{3}{5} \frac{E_\omega^3}{2E_o^2} \text{Im}\{ \quad \} \qquad 5.3.1\text{-}2$$

where

$$\{ \quad \} = \{a[2/\sqrt{\pi} - 2(a+ix)w^*(x+ia)] + \text{Re } w(x+ia)\} . \qquad 5.3.1\text{-}3$$

The real and imaginary parts of 5.3.1-3 are plotted* in Figure 4 for $a = .01, .2, .5$, along with the corresponding quantities for stationary atoms, from 5.2.1-8 and 9 for comparison. The most interesting feature of the Doppler broadened curves is that even though the gain saturation is essentially independent of a, and in fact is larger for small a, the corresponding index correction is smaller for smaller a, and negligible for $a = .01$. This is physically explained by the observation that for $a \ll 1$ the line is effectively inhomogeneously broadened, and the atoms responsible for the gain,

---

* The plots in Figure 4 and other figures of this chapter are of $\omega P_{s,c}/2\varepsilon_o c$, relative to

$$\alpha(GF)\, E_\omega^3/2E_o^2 \quad .$$

The horizontal (relative frequency) coordinate is uniform in all plots, and the vertical coordinate is indicated in each plot. Where the sign is significant, the plots give the corrections to be subtracted from the linear gain and index of refraction. In all cases $a = \gamma/k_o u$, $A = \gamma_a/2k_o u$, $B = \gamma_b/2k_o u$, so that $a = A + B$. The curves were calculated by digital computer and plotted automatically. The figures were traced from these curves and unfortunately sometimes contain small "wiggles" due to the plotter.

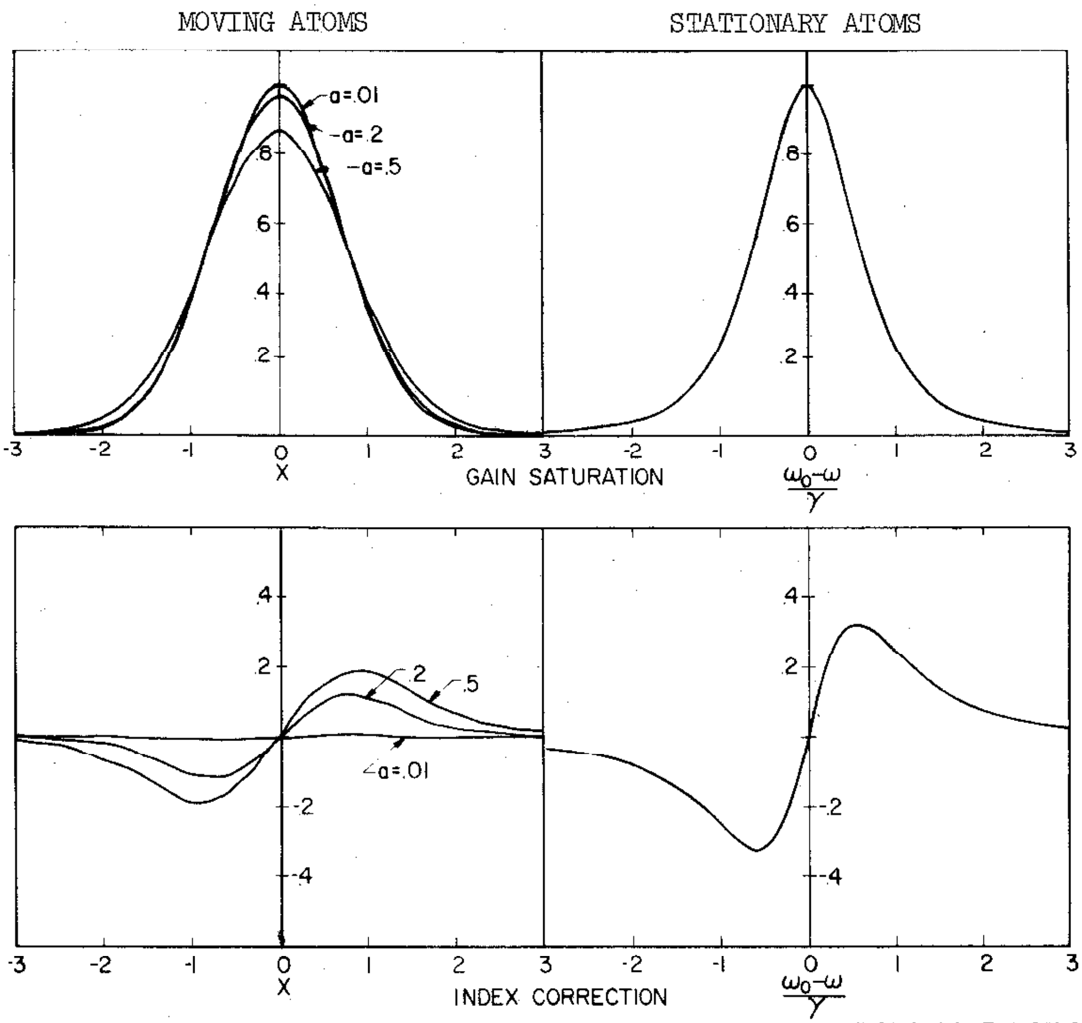


FIGURE 4 LOWEST ORDER GAIN SATURATION AND INDEX CORRECTIONS, AS FUNCTIONS OF FREQUENCY; COMPARED FOR MOVING AND STATIONARY ATOMS

and therefore susceptible to saturation, do not appreciably contribute to the index of refraction (15). If we use I-18 and 19 to expand 5.3.1-1 and 2 to first order in a, we find

$$\frac{\omega}{2\epsilon_o c} P_{\omega s} = -\alpha \frac{3}{5} \frac{E_\omega^3}{2E_o^2} e^{-x^2} , \qquad 5.3.1\text{-}4$$

$$\frac{\omega}{2\epsilon_o c} P_{\omega c} = \alpha \frac{3}{5} \frac{E_\omega^3}{2E_o^2} 2axe^{-x^2} , \qquad 5.3.1\text{-}5$$

for $a \ll 1$, which clearly demonstrate the features discussed above.

5.3.2 Two Input Waves

For two input waves at frequencies ω_1 and ω_2, as in the case of stationary atoms, 5.3-1 gives the single-wave corrections of 5.3.1, and in addition terms depending on both fields. We shall divide the discussion into two parts, first considering two waves traveling in the same direction, and then two waves traveling in opposite directions.

A. Two Waves in the Same Direction

In this case the results of II.1 are sufficient for all terms, since we always have

$$\underline{k} + \underline{k}' - \underline{k}'' \sim \underline{k} .$$

Also, the effects of Doppler broadening are equivalent to a distribution of resonance frequencies, ω_o.

As in the case of stationary atoms, there are three kinds of terms, viz., saturation at ω_2 due to the wave at $\omega_1 (\omega = \omega_2, \omega' = \omega'' = \omega_1)$, modulation" at $\omega_1(\omega = \omega'' = \omega_2,\ \omega' = \omega_1)$, and modulation at $2\omega_2 - \omega_1(\omega = \omega' = \omega_2,\ \omega'' = \omega_1)$. Of course similar terms occur with the indices 1 and 2 interchanged.

For the first case, with $\omega = \omega_2,\ \omega' = \omega'' = \omega_1$ in 5.3-1, we find using the geometrical factor 5.2.2-1,

$$\frac{\omega_2 P_{2s}}{2\epsilon_o c} = -\alpha\,\frac{1 + 2\cos^2\Theta_{12}}{5}\,\frac{E_1^2 E_2}{2E_o^2}\,\mathrm{Re}\,\{\quad\}\,, \qquad 5.3.2\text{-}1$$

$$\frac{\omega_2 P_{2c}}{2\epsilon_o c} = -\alpha\,\frac{1 + 2\cos^2\Theta_{12}}{5}\,\frac{E_1^2 E_2}{2E_o^2}\,\mathrm{Im}\,\{\quad\}\,, \qquad 5.3.2\text{-}2$$

with

$$\{\quad\} = \left\{a\left[\frac{w^*(x_2 + ia) + w(x_1 + ia)}{2a - i(x_1 - x_2)} - \frac{w^*(x_1 + ia) - w^*(x_2 + ia)}{i(x_1 - x_2)}\right]\right\}\,, \qquad 5.3.2\text{-}3$$

5.3.2-3 is plotted in Figure 5 as a function of x_2 for a = .01, .2, .5, and $x_1 = 0,\ 1$. The behavior of the curves is determined primarily by the first term which has a denominator $2a + i(x_1 - x_2)$. Thus the curves are essentially Lorentzian (more so for small a) and have a total half-width 4a. The effects are strongest in the region $x_1 \simeq x_2$, and for $x_1 = 1$ the weighting of the approximately Gaussian "envelope" function is evident. The physical interpretation of these results is as follows: The wave at ω_1 depletes the population inversion of atoms with velocities which allow them to

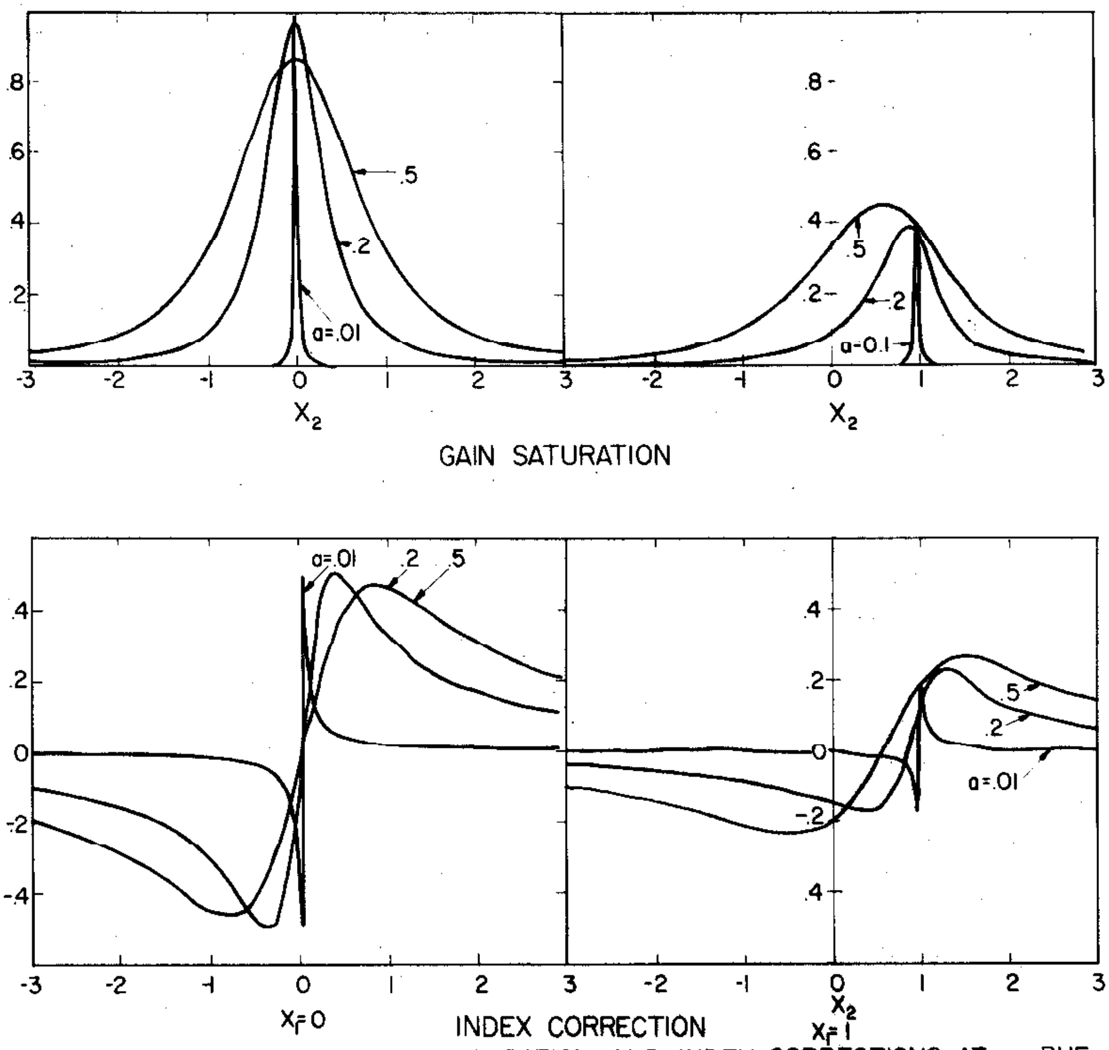


FIGURE 5 LOWEST ORDER GAIN SATURATION AND INDEX CORRECTIONS AT ω_2 DUE TO WAVE AT ω_1 FOR a=.01, .2, .5

$X_1 = \frac{\omega_0 - \omega_1}{k_0 u}$; $X_2 = \frac{\omega_0 - \omega_2}{k_0 u}$

interact strongly, i.e., over a frequency width 2γ. Since the wave at ω_2 also interacts with atoms over a frequency range 2γ, the total width of the gain reduction at ω_2 is 4γ, or $4a$ in Figure 5. This of course agrees with Bennett's revised "hole burning" arguments (2). The sign of the index correction shown in Figure 5 is correct, since for $\omega_2 > \omega_1$ the atoms removed by the field at ω_1 would have made a positive contribution to the index at ω_2.

It is interesting to note that, from the form of 5.3.2-3, Figure 5 also applies for the corrections at ω_1 if the sign of the index correction is reversed, and E_1 and E_2 are interchanged.

For the second case, with $\omega = \omega'' = \omega_2$ and $\omega' = \omega_1$ in 5.3-1, we have

$$\frac{\omega_1 P_{1s}}{2\epsilon_o c} = -\alpha \frac{1 + 2\cos^2\Theta_{12}}{5} \frac{E_1 E_2^2}{2E_o^2} \operatorname{Re}\{\ \ \} , \qquad 5.3.2\text{-}4$$

$$\frac{\omega_1 P_{1c}}{2\epsilon_o c} = -\alpha \frac{1 + 2\cos^2\Theta_{12}}{5} \frac{E_1 E_2^2}{2E_o^2} \operatorname{Im}\{\ \ \} , \qquad 5.3.2\text{-}5$$

where

$$\{\ \ \} = \left\{ AB\left(\frac{1}{A + i\frac{x_1 - x_2}{2}} + \frac{1}{B + i\frac{x_1 - x_2}{2}}\right)\right.$$

$$\left. \times \left[2/\sqrt{\pi} - 2(a + ix_1)\, w^*(x_1 + ia) + \frac{w^*(x_1 + ia) + w(x_2 + ia)}{2a + i(x_1 - x_2)}\right]\right\} . \qquad 5.3.2\text{-}6$$

For $x_1 = x_2$, 5.3.2-6 and 3 are equal. However for $x_1 \neq x_2$, 5.3.2-6 drops off considerably more rapidly than 5.3.2-3, due to the Lorentzian denominators with widths 2A and 2B. This behavior can be seen graphically by comparing Figure 5 to Figure 6, where 5.3.2-6 is plotted. The shape of these curves is again determined primarily by the Lorentzian denominators.

We can interpret this process as follows: For $\omega' \neq \omega''$ in 5.3-1, we have $\omega \neq \omega'$ in 5.1-2, and therefore we see that this correction to the zero field population inversion density is a traveling <u>wave</u> with frequency $\omega - \omega'$ and propagation vector $\underline{k} - \underline{k}'$. This effect might be called a "coherent modulation" of the population inversion. This modulation gives rise to a modulation of the gain at all frequencies, and will generate "sidebands" for all waves present in the medium. These sidebands will be separated by $\pm \Delta\omega$ from the frequency of each wave, where $\Delta\omega = \omega' - \omega''$. For the present case, $\Delta\omega = \omega_1 - \omega_2$, and, for example, the sidebands of the wave at ω_2 will be at ω_1 and $2\omega_2 - \omega_1$. The sideband at ω_1 appears as corrections to the gain and index at that frequency, given by 5.3.2-4 through 6. The sideband at $2\omega_2 - \omega_1$ gives rise to a field introduced at that frequency, and this is the remaining term which we have yet to discuss. Of course this same interpretation applies for the corresponding processes in the case of stationary atoms, treated in 5.2.

The term in the polarization at $2\omega_2 - \omega_1$ is given by $\omega = \omega' = \omega_2$ and $\omega'' = \omega_1$ in 5.3-1:

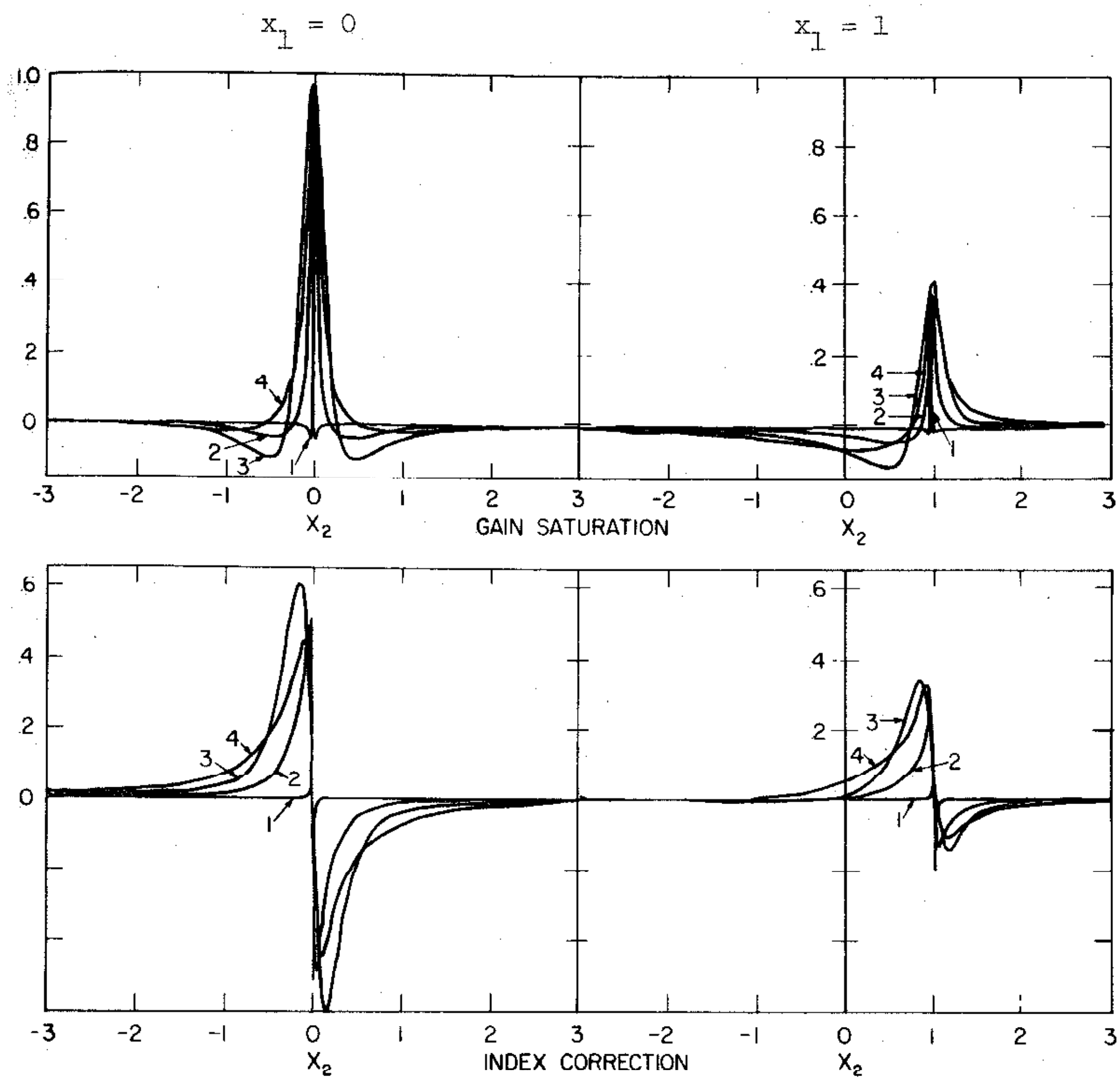


FIGURE 6 LOWEST ORDER GAIN SATURATION AND INDEX CORRECTIONS AT ω_1 DUE TO MODULATION PRODUCED BY E_1 AND E_2 AT ω_1 AND ω_2.
(1) a=.01, A=.001, B=.009
(2) a=.2, A=.02, B=.18
(3) a=.2, A=.1, B=.1
(4) a=.5, A=.05, B=.45

$$\frac{\omega_{21} P_{21s}}{2\epsilon_o c} = - \alpha \ (GF) \ \frac{E_1 E_2^2}{2E_o^2} \ \mathrm{Re} \ \{ \quad \} \quad , \qquad 5.3.2\text{-}7$$

$$\frac{\omega_{21} P_{21c}}{2\epsilon_o c} = - \alpha \ (GF) \ \frac{E_1 E_2^2}{2E_o^2} \ \mathrm{Im} \ \{ \quad \} \quad . \qquad 5.3.2\text{-}8$$

where

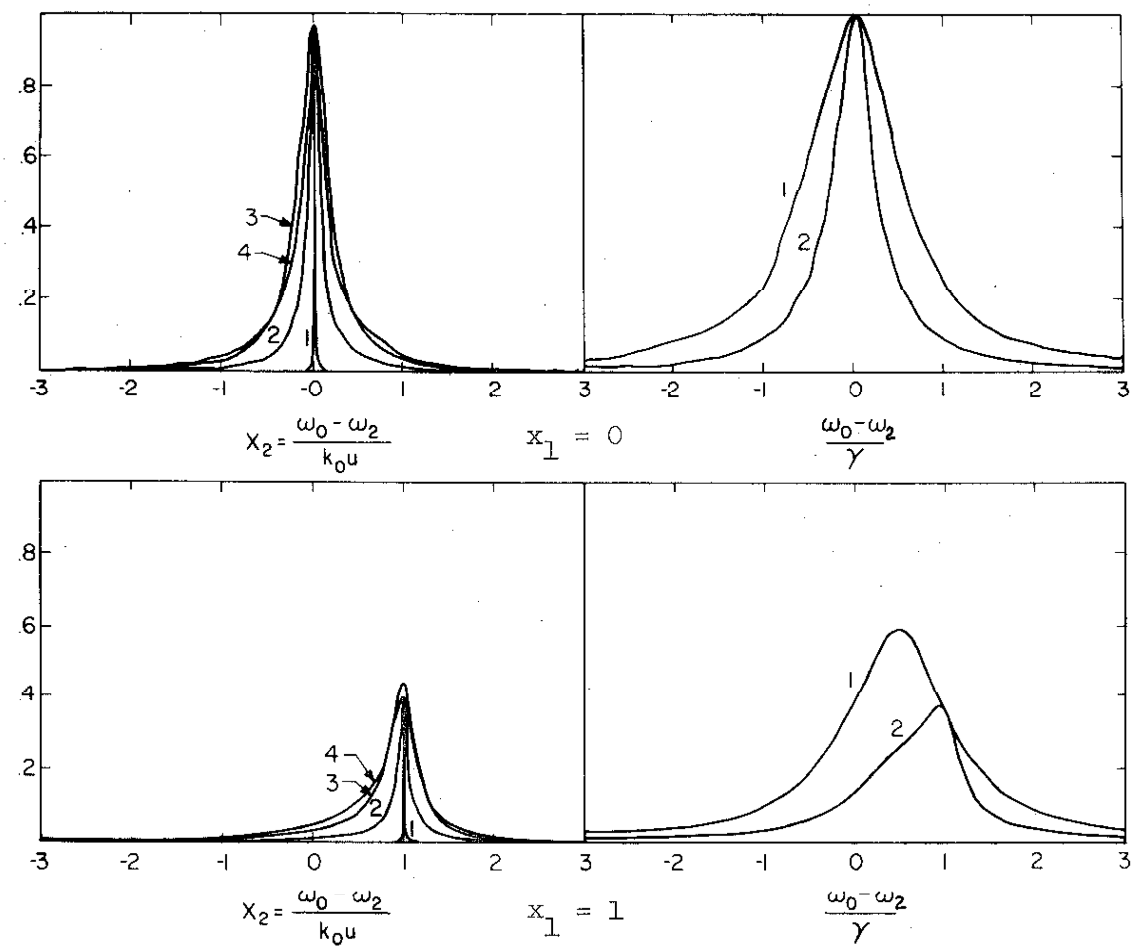
$$\{ \quad \} \left\{ AB \left(\frac{1}{A + i \dfrac{x_1 - x_2}{2}} + \frac{1}{B + i \dfrac{x_1 - x_2}{2}} \right) \right.$$

$$\left. \times \left[\frac{w^*(2x_2 - x_1 + ia) - w(x_1 + ia)}{2a + 2i(x_2 - x_1)} + \frac{w^*(2x_2 - x_1 + ia) - w^*(x_2 + ia)}{i(x_1 - x_2)} \right] \right\} \quad , \qquad 5.3.2\text{-}9$$

and (GF) is the geometrical factor 5.2.2-19. As in the case of stationary atoms, for the introduction of the new field we are interested only in the magnitude of the polarization,

$$\frac{\omega_{21} P_{21s}}{2\epsilon_o c} = - \alpha \ (GF) \ \frac{E_1 E_2^2}{2E_o^2} \ |\{ \quad \}| \quad , \qquad 5.3.2\text{-}10$$

and the discussion given for that case regarding Δk , the phase fluctuations, etc., holds completely. The magnitude of 5.3.2-9 is plotted in Figure 7 as a function of x_2 , for several cases, along with S_{21} of 5.2.2-27, the comparable quantity for stationary atoms.



MOVING ATOMS

1. a=.01, A=.001, B=.009
2. a=.2, A=.02, B=.18
3. a=2, A=.1, B=.1
4. a=.5, A=.05, B=.45

STATIONARY ATOMS

1. $\gamma_a = \gamma_b = \gamma$
2. $\gamma_a = \frac{1}{5}\gamma$, $\gamma_b = \frac{9}{5}\gamma$

FIGURE 7. LOWEST ORDER INDUCED GAIN AT $2\omega_2 - \omega_1$ DUE TO FIELDS AT ω_1, ω_2

B. Two Waves in Opposite Directions

For this case, we sometimes have $\underline{k}' - \underline{k}'' \sim 2\underline{k}'$ so that the results of II.2 must be used. The processes involved are of course the same as in A, and it will be interesting to note the differences in the response caused by reversing the direction of one of the waves.

We consider first the saturation effect of one wave on another. For $\omega = \omega_2$, $\omega' = \omega'' = \omega_1$, we find for the saturation effect at ω_2 due to a wave at ω_1 traveling oppositely,

$$\frac{\omega_2 P_{2s}}{2\epsilon_o c} = -\alpha \frac{1 + 2\cos^2 \Theta_{12}}{5} \frac{E_1^2 E_2}{2E_o^2} \operatorname{Re}\{\ \} , \qquad 5.3.2\text{-}11$$

$$\frac{\omega_2 P_{2c}}{2\epsilon_o c} = -\alpha \frac{1 + 2\cos^2 \Theta_{12}}{5} \frac{E_1^2 E_2}{2E_o^2} \operatorname{Im}\{\ \} , \qquad 5.3.2\text{-}12$$

where

$$\{\ \} = \left\{ a \left[\frac{w^*(x_1 + ia) + w^*(x_2 + ia)}{2a + i(x_1 + x_2)} + \frac{w(x_1 + ia) - w^*(x_2 + ia)}{i(x_1 + x_2)} \right] \right\} . \qquad 5.3.2\text{-}13$$

It is evident that 5.3.2-13 has its widest excursions near $x_2 = -x_1$, compared to $x_2 = x_1$ for both waves in the same direction (5.3.2-3). If in 5.3.2-13 either $x_1 \to -x_1$, or $x_2 \to -x_2$ with complex conjugation, 5.3.2-13 and 3 are the same. Thus the Figure 5 curves also apply for 5.3.2-13 if we reflect the curves through the vertical axis and change the sign of the P_c curves, for the same values of x_1. This is the behavior we expect, since waves at $+x$ and $-x$ interact with the same atoms.

An interesting limiting case for the saturation effect of two oppositely traveling waves is when they have the same frequency. This corresponds to a single cavity mode excited, and we find

$$\frac{\omega P_{s,c}}{2\epsilon_o c} = -\alpha \frac{3}{5} \frac{E^3}{2E_o^2} \text{Re, Im} \{ \quad \} , \qquad 5.3.2\text{-}14$$

with

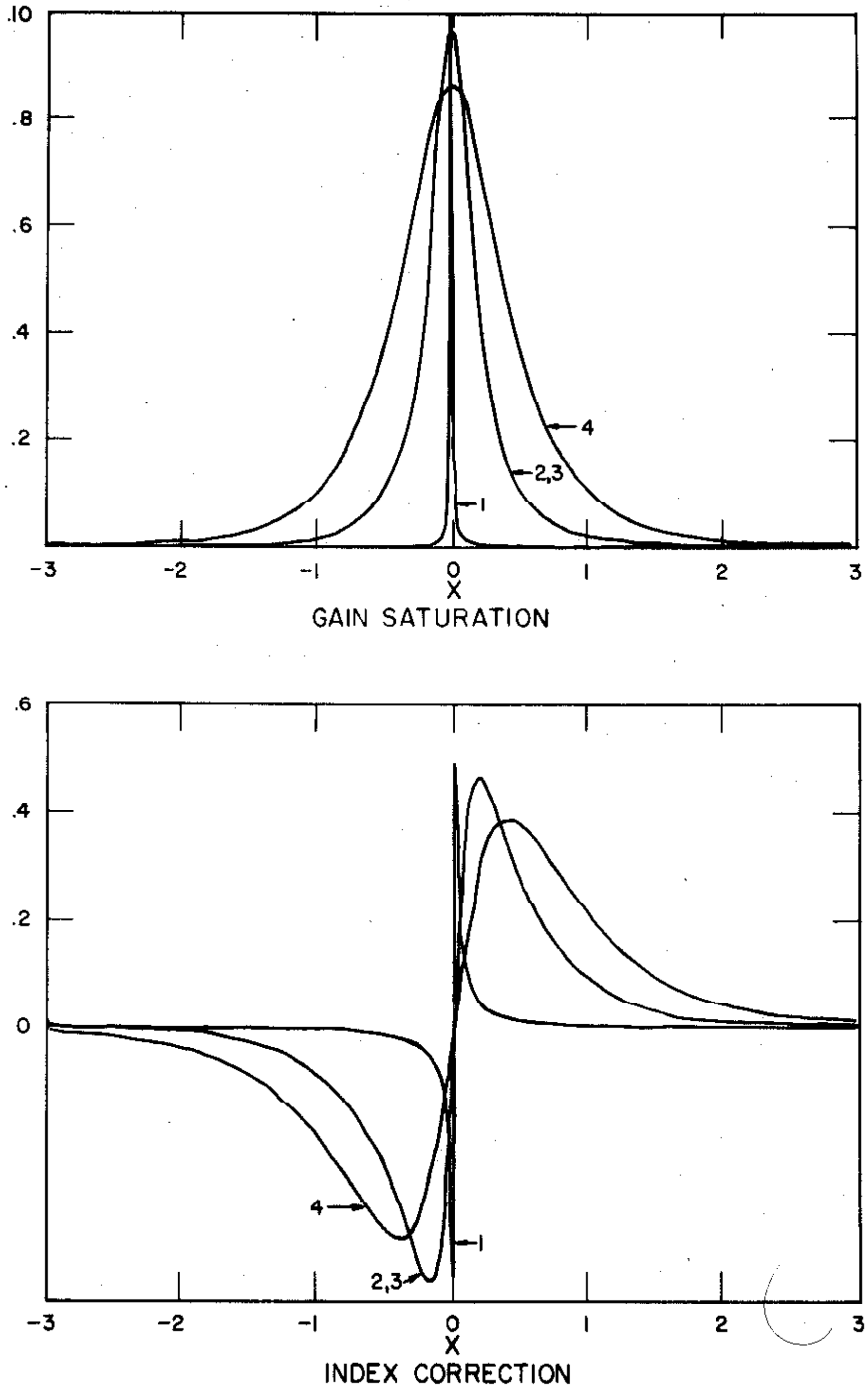
$$\{ \quad \} = \left\{ a \left[\frac{w^*(x + ia)}{a + ix} - \frac{\text{Im } w(x + ia)}{x} \right] \right\} . \qquad 5.3.2\text{-}15$$

where we have assumed waves of equal amplitude and polarized in the same direction. 5.3.2-15 is plotted in Figure 8 for several values of a . Note that this process has a much narrower frequency width (especially for the smaller values of a) than the self-saturation plotted in Figure 4. It is this feature, along with the fact that the two self-saturation processes affect the same atoms, which gives rise to the well-known "Lamb dip" observed in laser oscillators (12,35).

With $\omega = \omega'' = \omega_2$ and $\omega' = \omega_1$ in 5.3-1 we have the modulation effect at ω_1, for which

$$\frac{\omega_1 P_{1s,c}}{2\epsilon_o c} = -\alpha \frac{1 + 2\cos^2 \Theta_{12}}{5} \frac{E_1 E_2^2}{2E_o^2} \text{Re, Im} \{ \quad \} , \qquad 5.3.2\text{-}16$$

where, from II.2,



(1) a = .01 , A = .001 , B = .009
(2) a = .2 , A = .02 , B = .18
(3) a = .2 , A = .1 , B = .1
(4) a = .5 , A = .05 , B = .45

$$x = \frac{\omega_0 - \omega}{k_0 u}$$

FIGURE 8 LOWEST ORDER GAIN SATURATION AND INDEX CORRECTION FOR ONE HALF OF A CAVITY MODE DUE TO THE OTHER HALF

$$\{\ \} = \Bigg\{ AB \Bigg[- \frac{2/\sqrt{\pi} - 2(a + ix_1)\, w^*(x_1 + ia)}{A + i(x_1 + x_2)/2}$$

$$+ \frac{w((x_2 - x_1)/2 + iA) - w^*(x_1 + ia)}{(B + i(x_1 + x_2)/2)^2}$$

$$+ \Big(- (B - i\,\frac{x_1 + x_2}{2})\, w^*(x_1 + ia) + (B + i\,\frac{x_1 + x_2}{2})\, w(x_2 + ia)$$

$$- i(x_1 + x_2) w(\frac{x_2 - x_1}{2} + iA)\Big) \Big/ \Big((a + A - i\frac{3x_2 - x_1}{2})(B - i\frac{x_1 + x_2}{2})(a + ix_1)$$

$$- (a + A + i\,\frac{3x_1 - x_2}{2})(B + i\,\frac{x_1 + x_2}{2})(a - ix_2) + i(x_1 + x_2)(A + i\,\frac{x_1 - x_2}{2})$$

$$\times (2a + i(x_1 - x_2)\Big) + \text{same with A, B interchanged} \Bigg] \Bigg\} \ .$$

5.3.2-17

5.3.2-17 is plotted in Figure 9 for several cases with $x_1 = 0$, and compared with the corresponding curves for two waves traveling in the same direction. We see that the effect for waves in opposite directions is more than an order of magnitude less than for waves in the same direction, but extends over a wider frequency range. Thus the effects of Doppler broadening are considerably different for these two cases. In Figure 10 we have plotted several cases of 5.3.2-17 for $x_1 = 1$, and one case for a range of x_1 values to demonstrate the interesting qualitative change in the curves with x_1 . This behavior is also different from that found for waves traveling in the same direction.

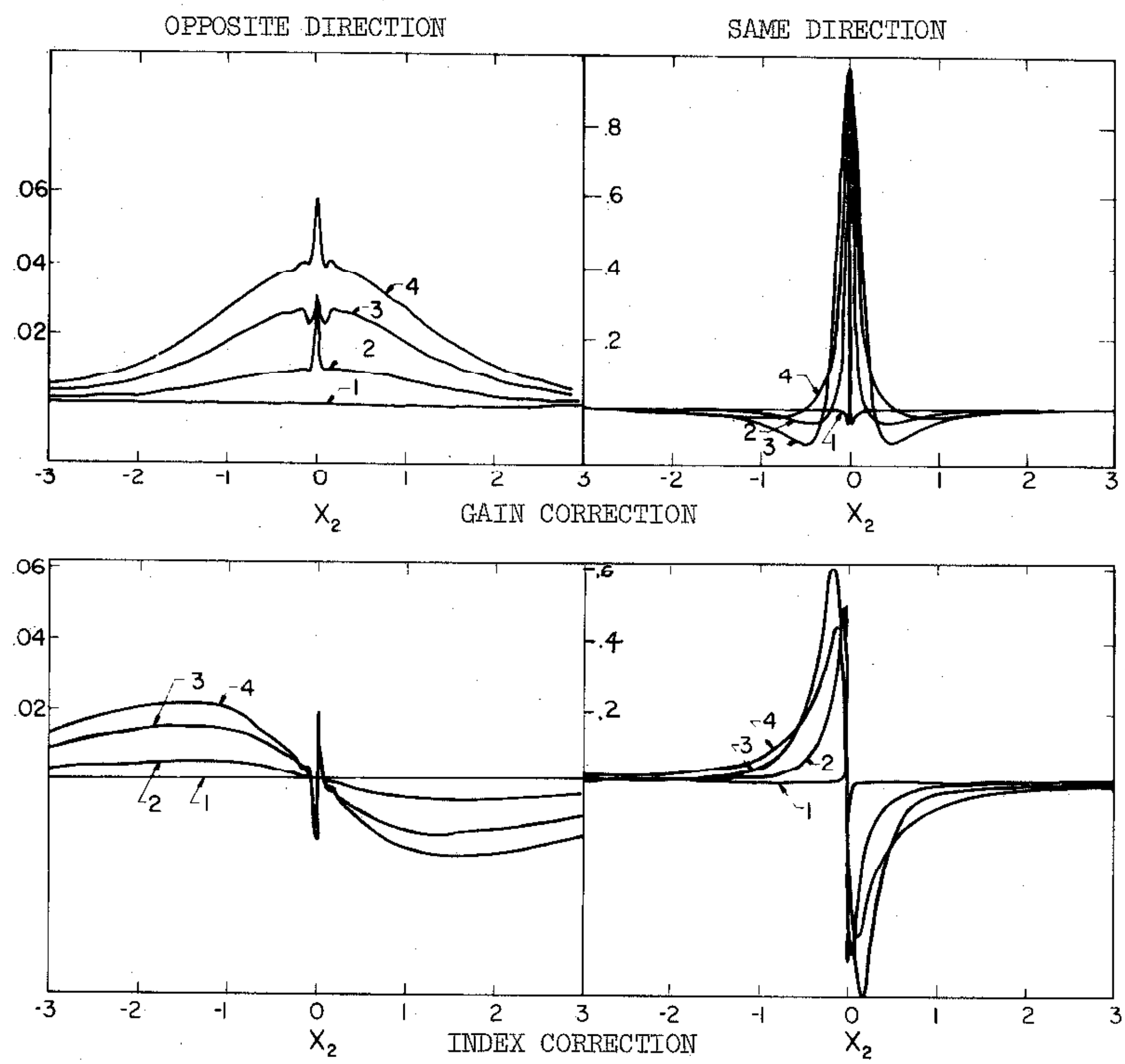


FIGURE 9 LOWEST ORDER GAIN AND INDEX CORRECTIONS AT ω_1 DUE TO WAVES AT ω_1 AND ω_2 FOR $x_1=0$

$x_1 = \frac{\omega_0 - \omega_1}{k_0 u}$, $x_2 = \frac{\omega_0 - \omega_2}{k_0 u}$

1. a=.01, A=.001, B=.009
2. a=.2, A=.02, B=.18
3. a=.2, A=.1, B=.1
4. a=.5, A=.05, B=.45

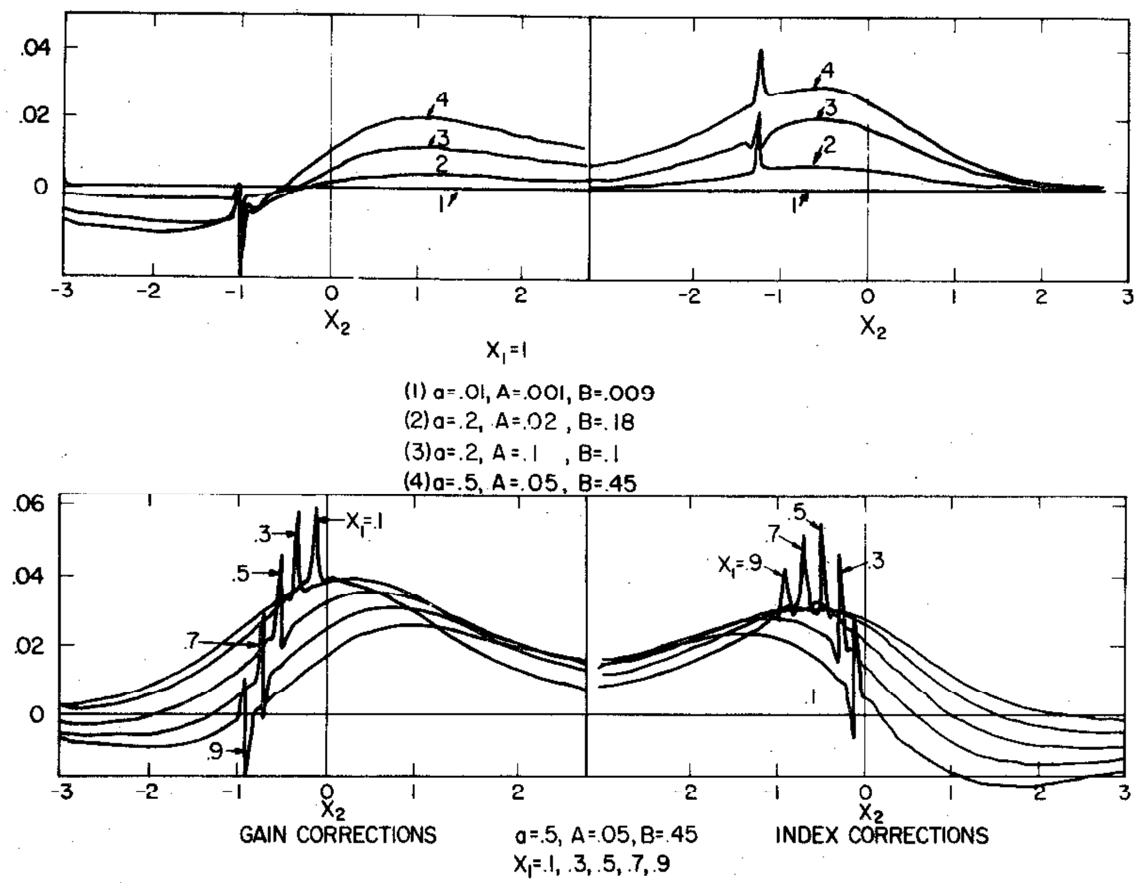


FIGURE 10 LOWEST ORDER GAIN AND INDEX CORRECTIONS AT ω_1 DUE TO MODULATION BY OPPOSITELY-DIRECTED WAVES AT ω_1 AND ω_2.

$$X_1 = \frac{\omega_0 - \omega_1}{k_0 u} \quad , \quad X_2 \frac{\omega_0 - \omega_2}{k_0 u}$$

For the modulation effect at $2\omega_2 - \omega_1$, 5.3.2-9 is replaced by the complicated expression

$$\left\{\frac{AB}{3}\left[-\left(B - i\,\frac{x_1 - x_2}{2}\right)w^*\left(\frac{2x_2 - x_1}{3} + i\,\frac{a}{3}\right) + \left(\frac{a}{3} - A + i\,\frac{x_2 + x_1}{6}\right)w(x_1 + ia)\right.\right.$$

$$\left. + \left(\frac{2}{3}a - 2i\,\frac{x_1 + x_2}{3}\right) w\left(\frac{x_1 - x_2}{2} + iA\right)\right] \Big/ \left[\left(\frac{a}{3} + A + i\,\frac{7x_2 - 5x_1}{6}\right)\right.$$

$$\times\left(A - \frac{a}{3} - i\,\frac{x_1 + x_2}{2}\right)(a - ix_1) + \left(a + A - i\,\frac{3x_1 - x_2}{2}\right)\left(B - i\,\frac{x_1 + x_2}{2}\right)$$

$$\times\left(\frac{a}{3} + i\,\frac{2x_2 - x_1}{3}\right) + \left(\frac{4}{3}a + 2i\,\frac{x_2 - 2x_1}{3}\right)\left(-\frac{2}{3}a + 2i\,\frac{x_1 + x_2}{3}\right)$$

$$\left.\times\left(A - i\,\frac{x_1 - x_2}{2}\right)\right] + \frac{AB}{3}\left[-\left(B + i\,\frac{x_1 + x_2}{2}\right) w^*\left(\frac{2x_2 - x_1}{3} + i\,\frac{a}{3}\right)\right.$$

$$\left. + \left(\frac{a}{3} - A + i\,\frac{x_1 + x_2}{6}\right) w^*(x_2 + ia) + \left(\frac{2}{3}a + i\,\frac{x_1 + x_2}{3}\right) w\left(\frac{x_1 - x_2}{2} + iA\right)\right] \Big/$$

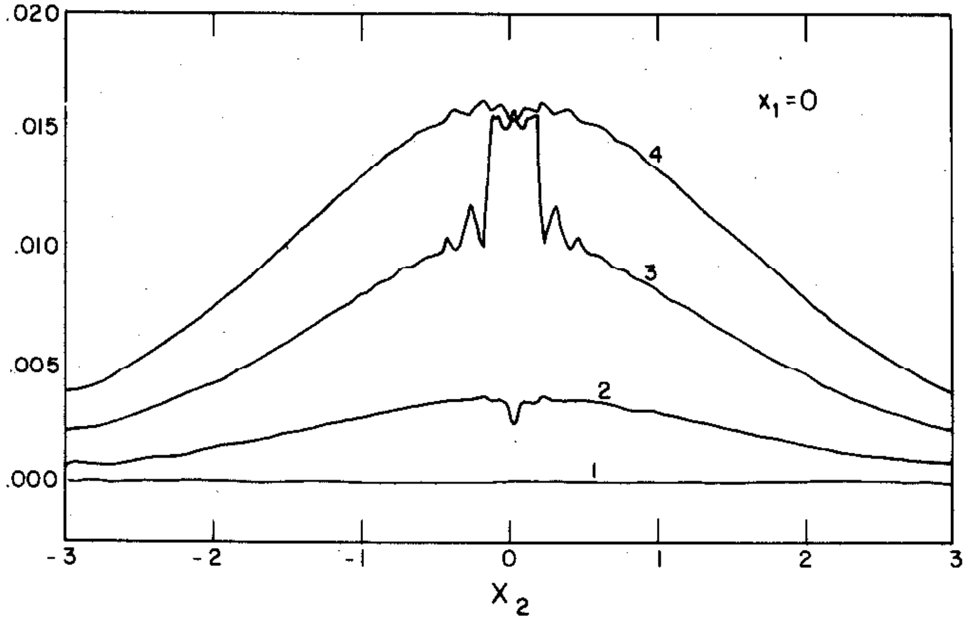
$$\left[\left(\frac{a}{3} + A + i\,\frac{7x_2 - 5x_1}{6}\right)\left(A - \frac{a}{3} - i\,\frac{x_1 + x_2}{6}\right)(a + ix_2) + \left(a + A - i\,\frac{x_1 - 3x_2}{2}\right)\right.$$

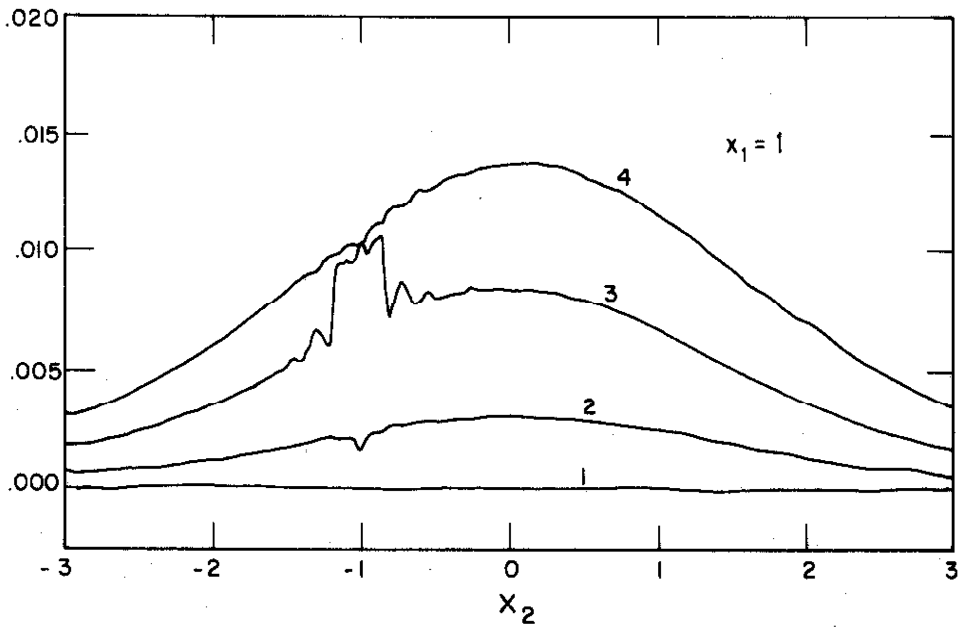
$$\times\left(B + i\,\frac{x_1 + x_2}{2}\right)\left(\frac{a}{3} + i\,\frac{2x_2 - x_1}{3}\right) + \left(\frac{4}{3}a + i\,\frac{5x_2 - x_1}{3}\right)$$

$$\left.\left.\times\left(-\frac{2}{3}a - i\,\frac{x_1 + x_2}{3}\right)\left(A - i\,\frac{x_1 - x_2}{2}\right)\right] \quad + \text{same with A, B, interchanged}\right\}.$$

5.3.2-18

The magnitude of 5.3.2-18 is plotted in Figure 11 for several cases. These curves should be compared with Figure 7 for the case of two waves in the same direction. We see that the effect for oppositely





$X_1 = \frac{\omega_0 - \omega_1}{k_0 u}$, $X_2 = \frac{\omega_0 - \omega_2}{k_0 u}$

1) a = .01 , A = .001 , B = .009
2) a = .2 , A = .02 , B = .18
3) a = .2 , A = .1 , B = .1
4) a = .5 , A = .05 , B = .45

FIGURE 11 INDUCED GAIN AT $2\omega_2 - \omega_1$ DUE TO OPPOSITELY DIRECTED WAVES AT ω_1, ω_2

directed waves is more than 50 times smaller and again has a considerably larger frequency width.

It is interesting to again consider the case of a cavity mode. In this case both 5.3.2-17 and 18 give modulation effects at the mode frequency. The dependence on frequency is obtained by putting $x_1 = x_2 = x$ in 5.3.2-17 and 18, and the resulting curves are shown in Figure 12. Again we see the relatively stronger interaction near the origin.

We have not in the above discussion considered the perpendicular components of the polarization, or the resulting components of the field. These effects are treated exactly as for stationary atoms, using the proper geometrical factor and the magnitude of the response, and need not be considered here.

5.3.3 More Than Two Input Waves

As in the case of stationary atoms, we will not consider explicitly cases where more than two waves are incident on the medium, since no new physical results are to be expected. The results of Appendix II can be immediately applied to study any particular term of interest. We should note, however, that we will have the same qualitative dependence on the directions of propagation as was found in 5.3.2 .

5.4 Discussion

In this chapter we have studied in detail the lowest order nonlinear processes for both stationary atoms and atoms with a Maxwellian velocity distribution at the time of excitation. The various

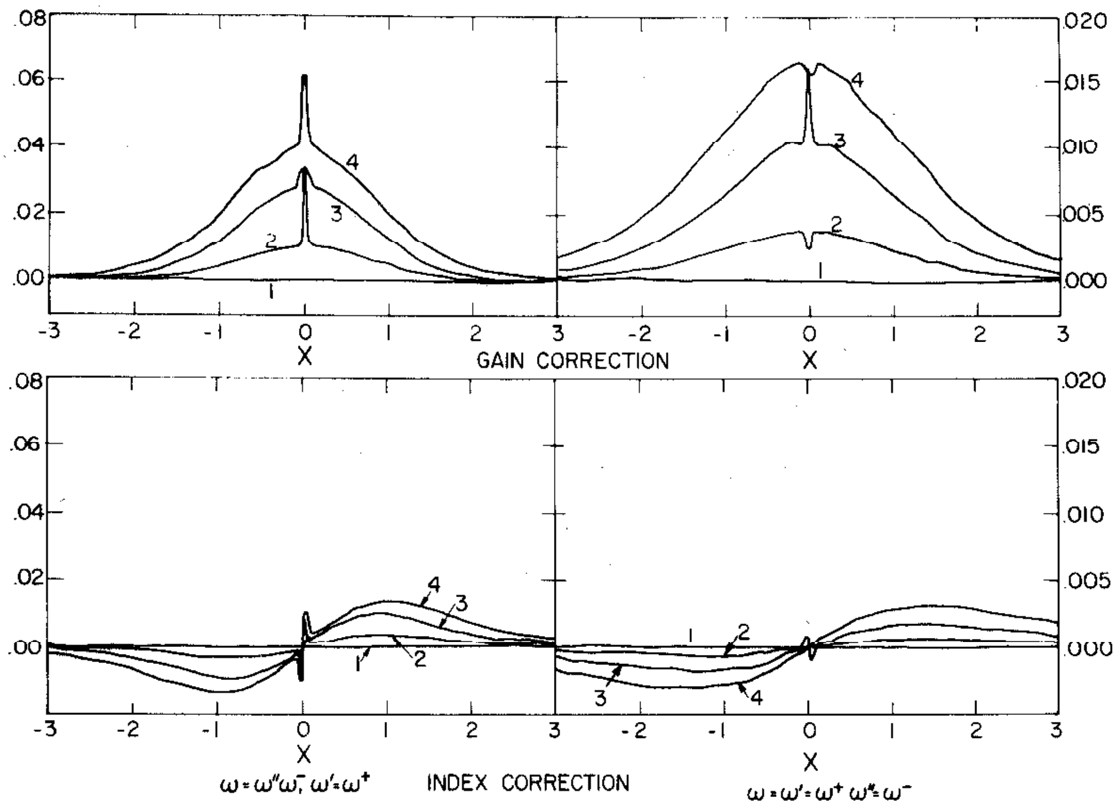


FIGURE 12 LOWEST ORDER GAIN AND INDEX CORRECTIONS FOR ONE HALF OF A CAVITY MODE DUE TO MODULATION

$X = \dfrac{\omega_0 - \omega}{k_0 u}$

1) a = .01 , A = .001 , B = .009
2) a = .2 , A = .02 , B = .18
3) a = .2 , A = .1 , B = .1
4) a = .5 , A = .05 , B = .45

nonlinear processes encountered have been briefly interpreted in terms of saturation and coherent modulation of the population inversion density.

In summary, the presence of two or more strong fields induces corrections to the gain and index of refraction for each field. In addition there are in general new fields introduced, at the old frequencies with perpendicular polarizations, and at new frequencies. The fields at new frequencies are limited to propagation directions close to that of the inducing polarization, i.e., small Δk . This "phase matching" condition and the existence of the perpendicularly polarized fields constitute an effective anisotropy of the medium, induced by the nonlinear processes. For gaseous laser media the difference between $\underline{k}_{21}$ and $2\underline{k}_2 - \underline{k}_1$ will be small, except in the cases where, for example, $2\underline{k}_2 - \underline{k}_1 \sim 3\underline{k}_2$. The latter cases were also characterized by a particularly weak nonlinear response.

Several of the nonlinear processes which have been discussed should be observable in practical situations. For example, the perpendicularly polarized wave could be observed with an appropriately oriented polarizer and beating techniques. As another example, the new field at $2\omega_2 - \omega_1$ should be observable even under conditions where the lowest order nonlinear corrections are sufficient, viz. $(E_1^2 + E_2^2)/E_0^2 \ll 1$. If $|\omega_1 - \omega_2| \ll \gamma_a, \gamma_b$, 5.3.2-9 becomes essentially unity, and neglecting the nonlinear corrections to the phase-independent gain and the geometrical factor, we have at line center

$$\frac{\partial E_{21}}{\partial z} = \alpha\, E_{21} - \alpha\, \frac{E_1 E_2^2}{2E_o^2} \quad . \qquad 5.4\text{-}1$$

5.4-1 gives

$$E_{21}(z) = -\frac{E_{10}E_{20}^2}{4E_o^2}\,(e^{2\alpha z} - 1)\, e^{\alpha z} \quad , \qquad 5.4\text{-}2$$

where E_{10} and E_{20} are the incident field strengths. Using the values of E_o^2 of Chapter 4, and requiring $E_{1,2}^2/2E_o^2 \leq .1$ we find: for $\alpha L = 1$ for the 3.4 micron transition in neon, we obtain .05 $\mu w/cm^2$ output at $2\omega_1 - \omega_2$ and $2\omega_2 - \omega_1$ for 4 $\mu w/cm^2$ input at ω_1 and ω_2 ; for αL - .05 for the .633 neon transition, inputs of about 5 mw/cm^2 give outputs of about .02 mw/cm^2 . Of course larger inputs are easily achievable, and we shall return later to consider the possible observation of these effects. We shall continue now with some more discussion of the physical interpretation of the nonlinear effects in 5.4.1, and of the longitudinal field in 5.4.2.

5.4.1 Physical Interpretation

The nonlinear response for moving atoms is very dependent on the directions of propagation of the interacting waves, in contrast to the response for stationary atoms. As previously discussed these effects cannot be adequately treated using a distribution of atomic resonance frequencies. This makes the inhomogeneous Doppler broadening somewhat unique from other forms of inhomogeneous broadening, characterized by such distributions. The inhomogeneous Doppler broadening is characterized by nonlinear interaction frequency widths much smaller than the width characterizing linear amplification.

We have previously interpreted some of the nonlinear processes in terms of a gain modulation associated with waves in the population inversion density, $\rho_{aa} - \rho_{bb}$. For two incident waves closely spaced in frequency, the atoms see a field with beats at the difference frequency $\omega_1 - \omega_2$. In fact, the beats move through the medium with the propagation constant $\Delta\underline{k} = \underline{k}_1 - \underline{k}_2$. For $\Delta\omega \ll \gamma_a, \gamma_b$, the time between successive field maxima for one atom is

$$\tau = \frac{\text{beat wavelength}}{\text{group velocity}} \quad .$$

Since the velocity is essentially that of light in free space,

$$\tau \approx \frac{2\pi}{c\Delta k} \approx \frac{1}{\Delta f} \gg \frac{1}{\gamma_a} , \frac{1}{\gamma_b}$$

where $\Delta f = \Delta\omega/2\pi$. Thus the population inversion can easily follow the beats in the field, for $\Delta\omega \ll \gamma_a, \gamma_b$, resulting in waves in the population inversion, traveling with the beats in the field.

The effect of these waves was previously discussed in terms of a gain modulation which generated sidebands for all the waves present in the medium. Another interesting viewpoint is that they represent a moving diffraction grating, which scatters the incident light into the sideband frequencies. This interpretation also gives the proper propagation directions for the sideband waves.

Some further insight into these nonlinear processes can be obtained by thinking of them as traveling wave parametric processes (36). Thus the waves in the population inversion density constitute a time- and space-varying propagation medium. The parametric effects of such a medium are well known (36), and result here in the "sideband" waves, with their characteristic propagation directions.

5.4.2 Neglect of Longitudinal Field

In 5.2.2 we found a component of the polarization along the direction of propagation at one frequency due to the field at another frequency. According to the equations of motion 2.3.2-20 and 21 for the fields, this component of the polarization should induce a longitudinal field. Actually, this longitudinal field will be much smaller than predicted by 2.3.2-20 and 21, and this is because the latter equations were derived by neglecting the $\nabla(\nabla \cdot \underline{E})$ term in 2.3.2-8, with the simultaneous assumption that the longitudinal field would be of higher order. We shall now show that this argument was indeed valid and that the longitudinal field is of order $\alpha_o c/\omega \lesssim 10^{-6}$ compared to the other induced fields.

If we take the vector $\underline{k}$ to be in the z direction, the term $\nabla(\nabla \cdot P)/\epsilon_o$ in 2.3.2-9 becomes

$$- \left(2k \frac{\partial P_{cz}}{\partial z} + k^2 P_{sz}\right) \sin(kz - \omega t + \varphi) +$$

$$+ \left(2k \frac{\partial P_{sz}}{\partial z} - k^2 P_{cz}\right) \cos(kz - \omega t + \varphi) \quad , \qquad 5.4.2\text{-}1$$

and including this term in 2.3.2-15 gives the field equations for the z component of the field:

$$\left(k^2 - \frac{\omega^2}{c^2}\right) E_z = \frac{\omega^2}{\epsilon_o c^2} P_{cz} + \frac{2k}{\epsilon_o} \frac{\partial P_{sz}}{\partial z} - \frac{k^2}{\epsilon_o} P_{cz} \quad , \qquad 5.4.2\text{-}2$$

$$2k \frac{\partial E_z}{\partial z} = \frac{\omega^2}{\epsilon_o c^2} P_{sz} - \frac{2k}{\epsilon_o} \frac{\partial P_{cz}}{\partial z} - \frac{k^2}{\epsilon_o} P_{sz} \quad . \qquad 5.4.2\text{-}3$$

The equations for E_x and E_y are the same as 2.3.2-20 and 21. With the same approximations used in 2.3.2 we find for 5.4.2-3

$$\frac{\partial E_z}{\partial z} = - \frac{(n - 1)\omega}{\epsilon_o c} P_{sz} - \frac{1}{\epsilon_o} \frac{\partial P_{cz}}{\partial z} \quad , \qquad 5.4.2\text{-}4$$

where n is the index of refraction. Using the linear n, 5.4.2-4 becomes

$$\frac{\partial E_z}{\partial z} = \frac{\alpha_o}{\epsilon_o} \frac{\gamma(\omega_o - \omega)}{\gamma^2 + (\omega_o - \omega)^2} P_{sz} - \frac{1}{\epsilon_o} \frac{\partial P_{cz}}{\partial z} \quad . \qquad 5.4.2\text{-}5$$

From 5.3 we have

$$\frac{1}{\epsilon_o} P_{cz} \precsim \frac{3}{5} \frac{c\alpha_o}{\omega} \frac{E_2^2 E_1}{E_o^2} \quad , \qquad 5.4.2\text{-}6$$

$$\frac{1}{\epsilon_o} P_{sz} \precsim \frac{3}{5} \frac{c\alpha_o}{\omega} \frac{E_2^2 E_1}{E_o^2} \quad , \qquad 5.4.2\text{-}7$$

where frequency-dependent factors of order unity or less have been neglected. Substituting 5.4.2-6 and 7 into 5.4.2-5 and assuming linear amplification of the fields E_2 and E_1, we find

$$\frac{\partial E_z}{\partial z} \lesssim \frac{3}{5} \frac{c\alpha_o^2}{\omega} \frac{E_2^2 E_1}{E_o^2} \left[\frac{\gamma(\omega_o - \omega)}{\gamma^2 + (\omega_o - \omega)^2} - 3 \right] \quad . \qquad 5.4.2\text{-}8$$

Since the factor in square brackets is again of order unity, we have

$$\frac{\partial E_z}{\partial z} \lesssim \frac{c\alpha_o^2}{\omega} \frac{E_2^2 E_1}{E_o^2} \quad . \qquad 5.4.2\text{-}9$$

The corresponding nonlinear contribution to the transverse field is, from 5.2.2-16,

$$\frac{\partial E_x}{\partial z} \lesssim \alpha_o \frac{E_2^2 E_1}{E_o^2} \quad . \qquad 5.4.2\text{-}10$$

Comparing 5.4.2-9 and 10, we see that we will have

$$\frac{E_z}{E_x} \sim \frac{\alpha_o c}{\omega} \lesssim 10^{-6} \quad , \qquad 5.4.2\text{-}11$$

where E_x is the transverse component induced by the nonlinearity. Since the latter is small, the longitudinal field E_z will be negligible.

CHAPTER SIX

STRONG NONLINEAR EFFECTS

In Chapter 5 we have studied in detail the lowest order nonlinear effects by making a perturbation expansion of the response and keeping only the lowest order terms which were nonlinear in the field strengths. These terms were of order E^2/E_o^2 or less relative to the linear response of Chapter 4. Thus the lowest order solutions are valid if $E^2 \ll E_o^2$, and if the latter condition is not fulfilled, these solutions will not be mathematically valid (the perturbation solution will converge slowly or not at all). We have already seen that this condition is strongly violated in several practical cases.

In this chapter we shall first examine two different approaches to the study of strong nonlinear effects (section 6.1). Then, using an approximate solution, we study several strong nonlinear effects in sections 6.2 through 6.4. The results are discussed in section 6.5.

6.1 Two Approaches to the Study of Strong Nonlinear Effects

In this section we first examine the perturbation expansion as a means of studying strong nonlinear effects, and then introduce an approximate solution which will be used in later sections to study some strong nonlinear effects.

6.1.1 The Perturbation Expansion

The integral equation formulation of the equations of motion for the medium, given in Chapter 3, is convenient for obtaining the perturbation expansion solution for the response of the medium to incident fields. As previously indicated, equation 3.3-21 for the population inversion density can be iterated, and the resulting series of terms can be used in 3.3-29 to give an expression for the response in the form of an expansion in powers of the field strengths. The lowest order nonlinear terms in this expansion were used in Chapter 5. Even though this expansion will not be mathematically valid for strong nonlinear effects where $E^2 > E_o^2$, we can reasonably expect that it will give an indication of the higher order physical processes involved. Thus the next higher order terms beyond those considered in Chapter 5, containing the fifth power of the field strengths, will include terms in the response at frequencies $3\omega_1 - 2\omega_2$, $2\omega_1 + \omega_2 - 2\omega_3$, and so on, as well as terms at ω_1, ω_2, $2\omega_2 - \omega_1$, etc. Just as some third order terms at ω_1 represented saturation of the response at ω_1, some of the fifth order terms at ω_1, $2\omega_2 - \omega_1$, etc., represent saturation of the response at these frequencies. Similarly, other fifth order terms at $3\omega_1 - 2\omega_2$, etc., correspond to new, higher order effects. In general, for two input fields at ω_1 and ω_2 and neglecting the fields induced at other frequencies, the terms containing the $2n + 1$ th powers of the field strengths contribute to: the higher order (modulation-induced) response at $\omega_1 + n(\omega_1 - \omega_2)$ and $\omega_2 - n(\omega_1 - \omega_2)$, the first order saturation of the response at $\omega_1 + (n - 1)(\omega_1 - \omega_2)$ and $\omega_2 - (n - 1)(\omega_1 - \omega_2)$, the second order saturation of the response at $\omega_1 + (n - 2)(\omega_1 - \omega_2)$ and

$\omega_3 - (n - 2)(\omega_1 - \omega_2)$, . . . , and the nth order saturation of the response at ω_1 and ω_2 .

Thus the higher order terms of the expansion are of two rather distinct types, viz., those which introduce higher order effects and those which contribute to the saturation of a lower order effect. Also, it is evident that the latter type includes essentially all of the terms. These characteristics suggest that if we could somehow sum over all the saturation terms giving the response at some given frequency, then the remaining terms would each represent the response at a different frequency and would be convergent. The results obtained in this chapter verify this conclusion, although they are not obtained by explicitly summing terms of the perturbation expansion.

We note that in general the summation would be very difficult to actually carry out, especially for the case of moving atoms, due to the large number and complexity of the terms involved. In some simple, limiting cases it might be feasible to perform the summation, but the process would in any case be rather tedious. The approximate solution which will now be introduced surmounts some of these difficulties and gives results valid in certain limiting cases.

6.1.2 An Approximate Solution

Our previous results have indicated that the source of the nonlinear effects we have been studying is saturation and modulation of the population inversion density. Thus we are led to consider again the integral equation 3.3-21 for this quantity:

$$\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v}) = NW(\underline{v}) - \left(\frac{\omega_o}{2\hbar}\right)^2 \int_0^\infty dt_1 \int_0^\infty dt_2$$

$$\times \left(e^{-\gamma_a t_1} + e^{-\gamma_b t_1}\right)\left(\rho_{aa}(\underline{r}, t - t_1 - t_2, \underline{v}) - \rho_{bb}(\underline{r}, t - t_1 - t_2, \underline{v})\right)$$

$$\times \sum_{\omega,\omega'} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right) \left[e^{i\Delta + i(\omega - \omega' - (\underline{k} - \underline{k}') \cdot \underline{v})t_1}\right.$$

$$\left. \times\ e^{-[\gamma - i(\omega_o - \omega' + \underline{k}' \cdot \underline{v})]t_2} + c.c.\right] \quad , \qquad 6.1.2\text{-}1$$

where

$$\Delta = (\underline{k} - \underline{k}') \cdot \underline{r} - (\omega - \omega')t + \varphi - \varphi' \quad . \qquad 6.1.2\text{-}2$$

Examination of 6.1.2-1 shows that the solution is characterized by the rate of variation of $\rho_{aa} - \rho_{bb}$ compared to γ_a and γ_b : rates slow compared to γ_a, γ_b will appear strongly, while rates very rapid compared to γ_a, γ_b will have very small amplitude. From the perturbation solution we know that $\rho_{aa} - \rho_{bb}$ contains terms at frequencies like $n(\omega - \omega')$, so that if we restrict ourselves to frequencies such that $|\omega - \omega'| \ll \gamma_a, \gamma_b$ or $|\omega - \omega'| \gg \gamma_a, \gamma_b$, then we can predict fairly well what the results will be. In particular, if $|\omega - \omega'| \gg \gamma_a, \gamma_b$ there will be negligible variation in $\rho_{aa} - \rho_{bb}$ at $\omega - \omega'$; while if $|\omega - \omega'| \ll \gamma_a, \gamma_b$, the variations in $\rho_{aa} - \rho_{bb}$ will be so slow as to have no effect on the integral. In the latter case, we can remove $\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v})$ from the integral and obtain

$$\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v}) = NW(\underline{v}) -$$

$$-\left(\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v})\right) \left(\frac{\omega_o}{2\hbar}\right)^2 \int_0^\infty dt_1 \int_0^\infty dt_2 \left(e^{-\gamma_a t_1} + e^{-\gamma_b t_1}\right)$$

$$\times \sum_{\omega,\omega'} \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right) \left[e^{i\Delta + i(\omega - \omega' - (\underline{k} - \underline{k}') \cdot \underline{v})t_1}\right.$$

$$\left.\times\ e^{-[\gamma - i(\omega_o - \omega' + \underline{k}' \cdot \underline{v})]t_2} + c.c.\right] \quad . \qquad 6.1.2\text{-}3$$

Performing the integration and collecting terms gives

$$\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v}) = NW(\underline{v})$$

$$\times \left\{1 + \sum_{\omega,\omega'} \left(\frac{\omega_o}{2\hbar}\right)^2 \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right)\left(\frac{\underline{P}_o \cdot \underline{E}_{\omega'}}{\omega'}\right)\left[\frac{e^{i[(\underline{k} - \underline{k}') \cdot \underline{r} - (\omega - \omega')t + \varphi - \varphi']}}{\gamma - i(\omega_o - \omega' + \underline{k}' \cdot \underline{v})}\right.\right.$$

$$\left.\left.\times \left(\frac{1}{\gamma_a - i(\omega - \omega' - (\underline{k} - \underline{k}') \cdot \underline{v})} + \frac{1}{\gamma_a - i(\omega - \omega' - (\underline{k} - \underline{k}') \cdot \underline{v})}\right) + c.c.\right]\right\}^{-1} \qquad 6.1.2\text{-}4$$

According to the above discussion, we should neglect terms in the summation for which $|\omega - \omega'| \gg \gamma_a, \gamma_b$, and keep the terms for which $|\omega - \omega'| \ll \gamma_a, \gamma_b$. For intermediate frequency separations, 6.1.2-4 will be only approximately valid.

From 6.1.2-4 we see that the population inversion density will contain all harmonics of each frequency difference, $\omega - \omega'$, for which the harmonic frequency is small compared to γ_a, γ_b , and will contain higher harmonics to a lesser degree than is indicated by 6.1.2-4. We will find below that the amplitude of succeeding harmonics decreases rather rapidly even according to 6.1.2-4.

In order to calculate the polarization, we use 3.3-39:

$$\rho_{ab}(\underline{r},t,\underline{v}) = \frac{\omega_o}{2i\hbar} \int_0^\infty dt_1 \; \rho_{aa}(\underline{r},t - t_1,\underline{v}) - \rho_{bb}(\underline{r},t - t_1,\underline{v})$$

$$\times \sum_\omega \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right) e^{i(\underline{k} \cdot \underline{r} - \omega t + \varphi) - [\gamma + i(\omega_o - \omega + \underline{k} \cdot \underline{v})]t_1} \qquad 6.1.2\text{-}5$$

Since $\rho_{aa} - \rho_{bb}$ will be varying slowly compared to γ , we may take $\rho_{aa} - \rho_{bb}$ out of the integral, giving

$$\rho_{ab}(\underline{r},t,\underline{v}) = \frac{\omega_o}{2i\hbar} \left|\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v})\right|$$

$$\times \sum_\omega \left(\frac{\underline{P}_o \cdot \underline{E}_\omega}{\omega}\right) \frac{e^{i(\underline{k} \cdot \underline{r} - \omega t + \varphi)}}{\gamma + i(\omega_o - \omega + \underline{k} \cdot \underline{v})} \; . \qquad 6.1.2\text{-}6$$

Using 6.1.2-4 in 6.1.2-6 gives us an approximate solution, subject to the above restrictions. Although this solution is good for arbitrary $\underline{k}$ vectors, it is simplest for waves traveling in the same direction, so that $\underline{k} - \underline{k}' \approx 0$, and we will for simplicity consider only this case. For other cases we can use the qualitative result of Chapter 5, along with the saturation characteristics to be derived below, to obtain a good picture of the expected results. Furthermore, since the interpretation over all directions of $\underline{P}_o$ would be very complicated and would mask the basic saturation characteristics we want to study, we will not attempt to study nonlinear polarization effects in this chapter, but rather will rely on the results of Chapter 5. Here we replace $\underline{P}_o(\underline{P}_o \cdot \underline{E}_\omega)$ in 6.1.2-6 by $\underline{E}_\omega P_o^2/3$ in order to get the correct linear result, and replace $(\underline{P}_o \cdot \underline{E}_\omega)(\underline{P}_o \cdot \underline{E}_{\omega'})$ in 6.1.2-4 by $P_o^2 E_\omega E_{\omega'}$ for simplicity. With these simplifications, 6.1.2-4 becomes

$$\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v}) = NW(\underline{v})$$

$$\times\left\{1 + \sum_{\omega',\omega''} \frac{P_o^2 E_{\omega'} E_{\omega''}}{4\hbar^2} \frac{2\gamma}{\gamma_a\gamma_b}\left[\frac{e^{i[(\underline{k}' - \underline{k}'') \cdot \underline{r} - (\omega' - \omega'')t + \varphi' - \varphi'']}}{\gamma - i(\omega_o - \omega'' + \underline{k}'' \cdot \underline{v})} + c.c.\right]\right\}^{-1}$$

$$= NW(\underline{v}) \left\{1 + \sum_{\omega',\omega''} \frac{\gamma E_{\omega'} E_{\omega''}}{2E_o^2}\left[\frac{e^{i\Delta}}{\gamma - i(\omega_o - \omega'' + k'' \cdot v)} + c.c.\right]\right\}^{-1} , \quad 6.1.2\text{-}7$$

where

$$\Delta = (\underline{k}' - \underline{k}'') \cdot \underline{r} - (\omega' - \omega'')t + \varphi' - \varphi'' . \quad 6.1.2\text{-}8$$

Similarly, from 6.1.2-6 the polarization becomes

$$\underline{P}(\underline{r},t,\underline{v}) = \left\langle \underline{P}_o(\rho_{ab}(\underline{r},t,\underline{v}) + \rho^*_{ab}(\underline{r},t,\underline{v})) \right\rangle$$

$$= \frac{\omega_o P_o^2}{6i\hbar} (\rho_{aa}(\underline{r},t,v) - \rho_{bb}(\underline{r},t,\underline{v}) \sum_{\omega} \frac{E_\omega}{\omega} \left(\frac{e^{i(\underline{k} \cdot \underline{r} - \omega t + \varphi)}}{\gamma + i(\omega_o - \omega + \underline{k} \cdot \underline{v})} - c.c. \right) \quad .$$

6.1.2-9

6.1.2-7 and 9 form the approximate solution which will now be used to study several strong nonlinear effects.

6.2 Saturation with a Single Traveling Wave

With a single traveling wave 6.1.2-7 gives

$$\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v}) = \frac{NW(\underline{v})}{1 + \frac{E^2}{E_o^2} \frac{\gamma^2}{\gamma^2 + (\omega_o - \omega + \underline{k} \cdot \underline{v})^2}} \quad . \qquad 6.2\text{-}1$$

Putting 6.2-1 into 6.1.2-9 gives the polarization components

$$\frac{\omega P_s}{2\epsilon_o c} = \frac{\alpha_o \, E \, W(\underline{v}) \, \gamma^2}{\gamma^2(1 + E^2/E_o^2) + (\omega_o - \omega + \underline{k} \cdot \underline{v})^2} \quad , \qquad 6.2\text{-}2$$

$$\frac{P_c}{2\epsilon_o} = - \frac{c}{\omega} \frac{\alpha_o \, E \, W(\underline{v}) \, \gamma \, (\omega_o - \omega + \underline{k} \cdot \underline{v})}{\gamma^2(1 + E^2/E_o^2) + (\omega_o - \omega + \underline{k} \cdot \underline{v})^2} \quad . \qquad 6.2\text{-}3$$

6.2.1 Stationary Atoms

For stationary atoms, 6.2-2 and 3 along with 2.3.2-20 and 21 immediately give

$$\frac{\partial E}{\partial z} = \alpha_o E \frac{\gamma^2}{\gamma^2(1 + E^2/E_o^2) + (\omega_o - \omega)^2} \quad , \qquad 6.2.1\text{-}1$$

and

$$n(\omega) = 1 - \frac{c}{\omega}\alpha_o \frac{\gamma(\omega_o - \omega)}{\gamma^2(1 + E^2/E_o^2) + (\omega_o - \omega)^2} \quad . \qquad 6.2.1\text{-}2$$

If we put $E^2 = 0$ in the denominator of 6.2.1-1 and 2, we find the linear results 4.2-11 and 12. If we expand the denominator to first order in E^2/E_o^2, we obtain the lowest order nonlinear corrections, 5.2.1-8 and 9, except for the geometrical factor 3/5 which arose from the average over directions of $\underline{P}_o$, neglected in the present calculation.

Writing the intensity as

$$I = \epsilon_o c\, E^2 \qquad 6.2.1\text{-}3$$

and defining a relative intensity

$$\mathcal{I} = I/I_o = E^2/E_o^2 \quad , \qquad 6.2.1\text{-}4$$

we can write 6.2.1-1 as

$$\frac{1}{\mathcal{I}}\frac{\partial \mathcal{I}}{\partial z} = \frac{2\alpha_o\gamma^2}{\gamma^2(1 + \mathcal{I}) + (\omega_o - \omega)^2} \quad . \qquad 6.2.1\text{-}5$$

If $\omega_o = \omega$, 6.2.1-5 becomes

$$\frac{1}{I}\frac{\partial I}{\partial z} = \frac{2\alpha_o}{1 + I} \quad , \qquad 6.2.1\text{-}6$$

which is equation 15 of Gordon, White and Rigden (16), with $2\alpha_o = k_o/\epsilon\sqrt{\pi}$. The latter authors obtained this result using a rate equation type approach.

6.2.1-6 can be integrated (28) over an amplifying length L to provide a relationship between the input and output relative intensities, I_{in} and I_{out} , viz.,

$$2\alpha_o L = \ell n(I_{out}/I_{in}) + I_{out} - I_{in} \qquad 6.2.1\text{-}7$$

Using 6.2.1-7, we can plot curves of the gain, defined by

$$G = I_{out}/I_{in} \quad , \qquad 6.2.1\text{-}8$$

as a function of I_{in} . The small signal or linear gain is $G_o = e^{2\alpha_o L}$. Examples of such curves for several values of G_o are shown in Figure 13.

6.2.2 Maxwellian Velocity Distribution

For a Maxwellian distribution of excited atoms we use 4.1-1 and integrate over the velocity components perpendicular to $\underline{k}$ to obtain from 6.2-2 and 3

$$\frac{\omega}{2\epsilon_o c} P_s = \alpha_o E\gamma^2 \frac{1}{\sqrt{\pi}u} \int_{-\infty}^{\infty} \frac{e^{-V^2/u^2}\, dV}{\gamma^2(1 + E^2/E_o^2) + (\omega_o - \omega + kV)^2} \quad , \qquad 6.2.2\text{-}1$$

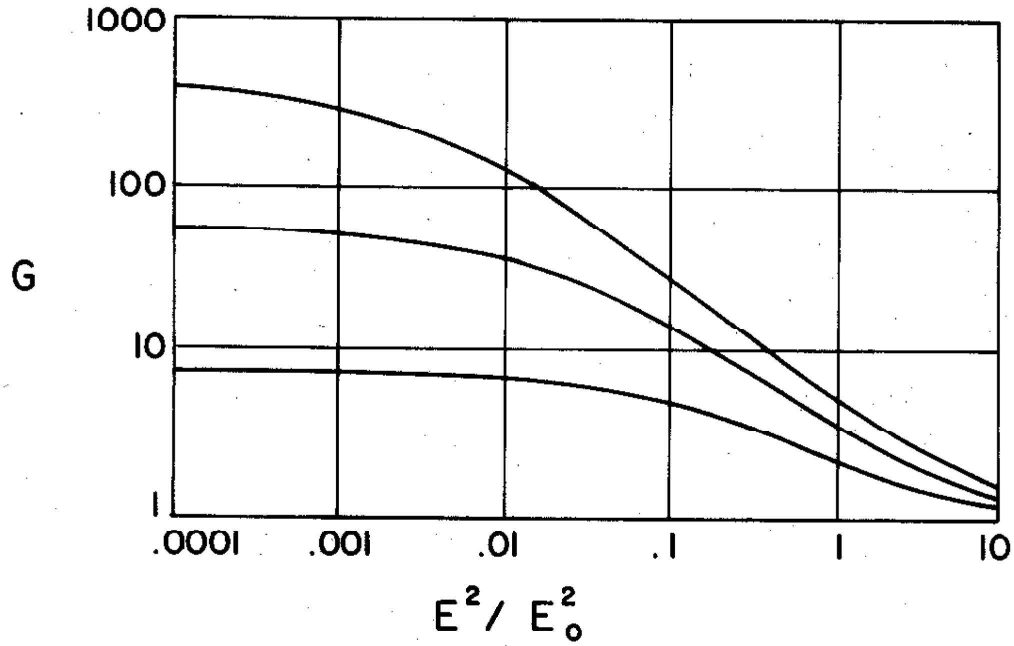


FIGURE 13 SINGLE FREQUENCY GAIN SATURATION FOR STATIONARY ATOMS

$$\frac{1}{2\epsilon_o} P_c = -\frac{c}{\omega}\alpha_o E\gamma \frac{1}{\sqrt{\pi}u}\int_{-\infty}^{\infty} \frac{(\omega_o - \omega + kV)\, e^{-V^2/u^2}\, dV}{\gamma^2(1 + E^2/E_o^2) + (\omega_o - \omega + kV)^2} \quad . \qquad 6.2.2\text{-}2$$

Defining

$$b^2 = \gamma^2(1 + E^2/E_o^2)/(k_o u)^2 \quad , \qquad 6.2.2\text{-}3$$

6.2.2-1 and 2 become

$$\frac{\omega}{2\epsilon_o c} P_s = \frac{\alpha_o E\, a^2\sqrt{\pi}}{b} \operatorname{Re} w(x + ib) \qquad 6.2.2\text{-}4$$

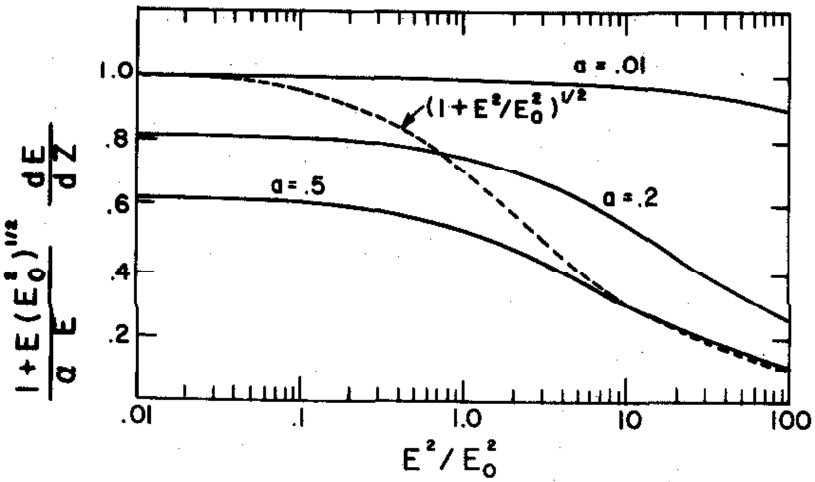
$$\frac{P_c}{2\epsilon_o} = -\frac{c}{\omega}\alpha_o E\, a\sqrt{\pi} \operatorname{Im} w(x + ib) \quad , \qquad 6.2.2\text{-}5$$

where $x = (\omega_o - \omega)/k_o u$, $a = \gamma/k_o u$, w is the complex error function as before, and the results of Appendix I have been used. Using $\alpha = \alpha_o a\sqrt{\pi}$, 6.2.2-4 and 5 in 2.3.2-20 and 21 give

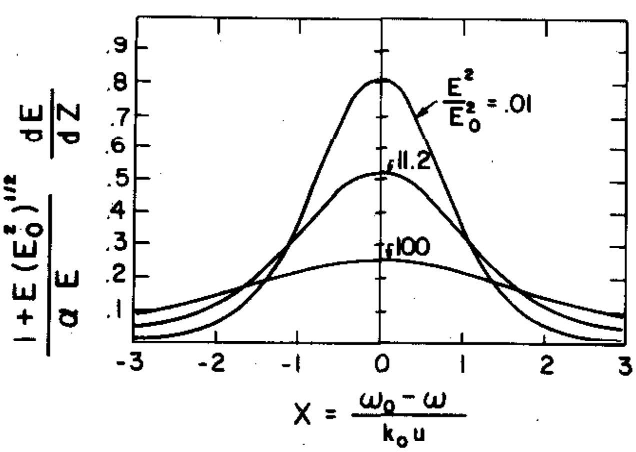
$$\frac{\partial E}{\partial z} = \frac{\alpha E}{(1 + E^2/E_o^2)^{\frac{1}{2}}} \operatorname{Re} w(x + ib) \quad , \qquad 6.2.2\text{-}6$$

$$n(\omega) = 1 - \frac{c}{\omega}\alpha \operatorname{Im} w(x + ib) \quad . \qquad 6.2.2\text{-}7$$

In Figure 14, $(1 + E^2/E_o^2)^{\frac{1}{2}}/(\alpha E)$ times 6.2.2-6 is plotted for several values of a and compared to homogeneous saturation. For $E \ll E_o$, 6.2.2-6 and 7 reduce to the linear results 4.3-14 and 15. Using II.3-4 to expand 6.2.2-6 and 7 to lowest order in E^2/E_o^2 gives the lowest order nonlinear corrections of 5.3, again without the 3/5 geometrical



HIGH FIELD SATURATION AT LINE CENTER IN EXCESS OF INHOMOGENEOUS SATURATION, COMPARED TO HOMOGENEOUS SATURATION; FOR VARIOUS VALUES OF $a = \gamma/k_0u$



HIGH FIELD SATURATION AND BROADENING FOR a = .2, AS A FUNCTION OF FREQUENCY

FIGURE 14 HIGH FIELD SATURATION AND BROADENING FOR MOVING ATOMS factor.

For $b = a(1 + E^2/E_o^2)^{\frac{1}{2}} \ll 1$, we can use I-18 and 19 to expand 6.2.2-6 and 7 to first order in b, obtaining

$$\frac{\partial E}{\partial z} = \alpha E \left[\frac{e^{-x^2}}{(1 + E^2/E_o^2)^{\frac{1}{2}}} - \frac{2a}{\sqrt{\pi}} (1 - 2\, x\, F(x)) \right] , \qquad 6.2.2\text{-}8$$

$$n(\omega) = 1 - \frac{c}{\omega} \alpha \left[\frac{2}{\sqrt{\pi}} F(x) - 2a(1 + E^2/E_o^2)^{\frac{1}{2}}\, x\, e^{-x^2} \right] . \qquad 6.2.2\text{-}9$$

These should be compared with 4.3-17 and 18 for the linear case. 6.2.2-8 and 9 are consistent with the "hole-burning" description of an inhomogeneously broadened line (2). Thus the gain comes primarily from those atoms whose velocities allow them to interact strongly with the field, while the phase shift (index of refraction) arises primarily due to atoms outside this region. Correspondingly, the primary gain term is saturated, while the primary index term is not. Also, the phase shift due to atoms within the hole increases with the width of the hole, while the contribution of atoms outside the hole to the gain is to first order independent of the width of the hole.

Using 6.2.1-3 and 4, 6.2.2-8 becomes

$$\frac{1}{I} \frac{\partial I}{\partial z} = 2\alpha \left[\frac{e^{-x^2}}{(1 + I)^{\frac{1}{2}}} - \frac{2a}{\sqrt{\pi}} (1 - 2x F(x)) \right] . \qquad 6.2.2\text{-}10$$

For $x = 0$, 6.2.2-10 becomes

$$\frac{1}{I}\frac{\partial I}{\partial z} = \frac{2\alpha}{(1 + I)^{\frac{1}{2}}}\left[1 - \frac{2a}{\sqrt{\pi}}(1 + I)^{\frac{1}{2}}\right] , \qquad 6.2.2\text{-}11$$

which is equation 16 of Gordon, White and Rigden (16), with $a = \epsilon$ and $2\alpha = k_o$. Equation 6.2.2-11 can also be integraded (16,28), giving

$$2\alpha L = 2(1 + I_{out})^{\frac{1}{2}} - 2(1 + I_{in})^{\frac{1}{2}}$$

$$+ \ln\left[\frac{(1 + I_{in})^{\frac{1}{2}} + 1}{(1 + I_{in})^{\frac{1}{2}} - 1}\right]\left[\frac{(1 + I_{out})^{\frac{1}{2}} - 1}{(1 + I_{out})^{\frac{1}{2}} + 1}\right] \qquad 6.2.2\text{-}12$$

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

Again we can plot gain versus I_{in} . Some of these curves are plotted in Figure 15 for several values of the small signal gain $G_o = e^{2\alpha L}$ for $a = 0$. We have also plotted some experimental points, due to Hotz (37), for the 3.39 micron line in neon. There is excellent agreement.

6.3 Strong Nonlinear Effects with Two Traveling Waves

For the case of two waves traveling in the same direction, 6.1.2-7 gives, according to section 6.1,

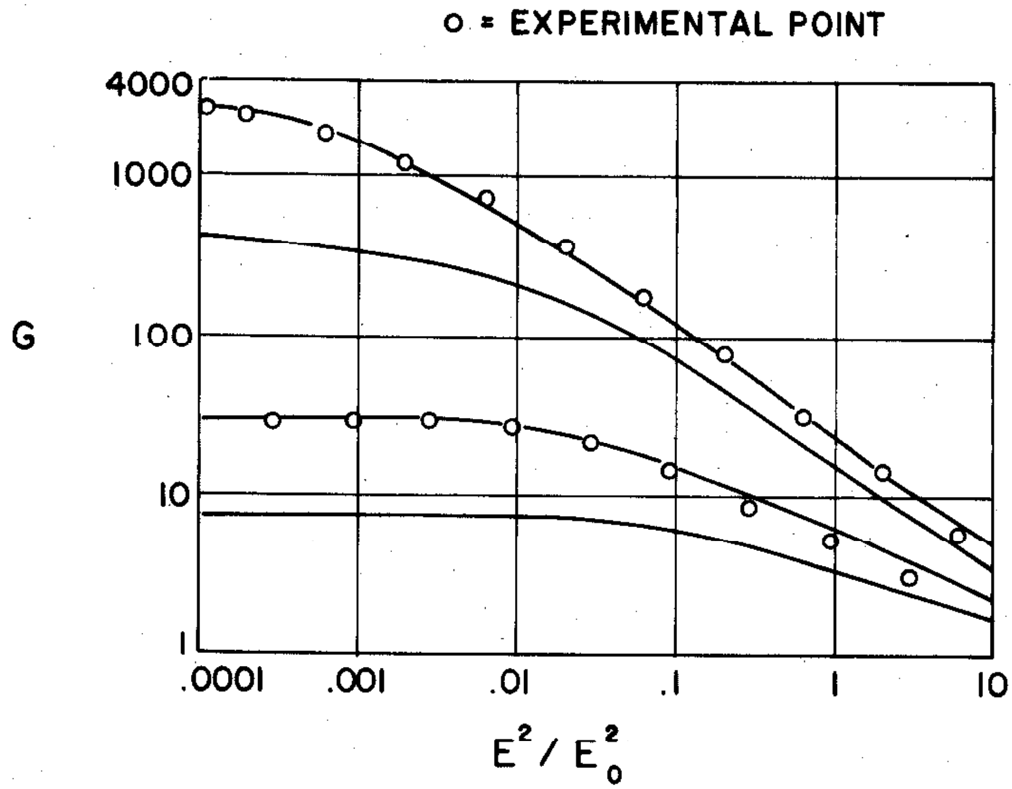


FIGURE 15 DOPPLER BROADENED SINGLE FREQUENCY GAIN SATURATION

$$\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v}) = NW(\underline{v})$$

$$\times \left[1 + \frac{E_1^2}{E_o^2}\frac{\gamma^2}{\gamma^2 + (\omega_o - \omega_1 + \underline{k}_1 \cdot \underline{v})^2} + \frac{E_2^2}{E_o^2}\frac{\gamma^2}{\gamma^2 + (\omega_o - \omega_2 + \underline{k}_2 \cdot \underline{v})^2}\right]^{-1}$$

6.3-1

if $|\omega_1 - \omega_2| \gg \gamma_a, \gamma_b$; and

$$\rho_{aa}(\underline{r},t,\underline{v}) - \rho_{bb}(\underline{r},t,\underline{v}) = NW(\underline{v})$$

$$\times \left[1 + \frac{E_1^2 + E_2^2 + 2E_1E_2 \cos\Delta_{12}}{E_o^2}\frac{\gamma^2}{\gamma^2 + (\omega_o - \omega + \underline{k} \cdot \underline{v})}\right]^{-1} ,$$

6.3-2

if $|\omega_1 - \omega_2| \ll \gamma_a, \gamma_b$, where $\omega_1 - \omega_2$ has been neglected compared to γ in 6.3-2 and

$$\Delta_{12} = (\underline{k}_1 - \underline{k}_2) \cdot \underline{r} - (\omega_1 - \omega_2)t + \varphi_1 - \varphi_2 \quad . \qquad 6.3\text{-}3$$

6.3.1 $|\omega_1 - \omega_2| \gg \gamma_a,\gamma_b$

Substituting 6.3-1 into 6.1.2-9 gives

$$\frac{\omega_1 P_1}{2\epsilon_o c} = \alpha_o \, W(\underline{v}) \, \gamma\left[E_1 \gamma \sin(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1) - (\omega_o - \omega_1 + \underline{k}_1 \cdot \underline{v})\cos(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1)\right]$$

$$\times \left\{ \gamma^2 \left[1 + \frac{E_1^2}{E_o^2} + \frac{E_2^2}{E_o^2} \frac{\gamma^2 + (\omega_o - \omega_1 + \underline{k}_1 \cdot \underline{v})^2}{\gamma^2 + (\omega_o - \omega_2 + \underline{k}_2 \cdot \underline{v})^2} \right] + (\omega_o - \omega_1 + \underline{k}_1 \cdot \underline{v})^2 \right\}^{-1} .$$

6.3.1-1

for the polarization at ω_1 . The polarization at ω_2 is obtained from 6.3.1-1 by exchanging subscripts 1 and 2.

For stationary atoms, we find immediately

$$\frac{\partial E_1}{\partial z} = \frac{\alpha_o \gamma^2 E_1}{\gamma^2 \left[1 + \frac{E_1^2}{E_o^2} + \frac{E_2^2}{E_o^2} \frac{\gamma^2 + (\omega_o - \omega_1)^2}{\gamma^2 + (\omega_o - \omega_2)^2} \right] + (\omega_o - \omega_1)^2} , \qquad 6.3.1\text{-}2$$

$$n(\omega_1) = 1 - \frac{c}{\omega_1} \frac{\alpha_o \gamma (\omega_o - \omega_1)}{\gamma^2 \left[1 + \frac{E_1^2}{E_o^2} + \frac{E_2^2}{E_o^2} \frac{\gamma^2 + (\omega_o - \omega_1)^2}{\gamma^2 + (\omega_o - \omega_2)^2} \right] + (\omega_o - \omega_1)^2}$$

6.3.1-3

with corresponding expressions at ω_2 . Since for this case one or both fields must be strongly off resonance, these results are not of much practical interest. However they do show the effect at one frequency of saturation due to a field at another frequency, and indicate that both fields interact with the same atoms. With $E_2 = 0$, we obtain the single-wave results 6.2.1-1 and 2. Expanded to first order in E_1^2/E_o^2 and E_2^2/E_o^2, 6.3.1-2 and 3 give the lowest order corrections 5.2.2-2, but of course do not include a contribution due to the modulation term 5.2.2-3. Again the geometrical factors are absent.

For a Maxwellian velocity distribution, 6.3.1-1 gives

$$\frac{\partial E_1}{\partial z} = \frac{\alpha a E_1}{\pi} \int_{-\infty}^{\infty} \frac{e^{-y^2}\, dy}{a^2\left(1 + \frac{E_1^2}{E_o^2} + \frac{E_2^2}{E_o^2}\frac{a^2 + (x_1 + y)^2}{a^2 + (x_2 + y)^2}\right) + (x_1 + y)^2}, \qquad 6.3.1\text{-}4$$

$$n(\omega_1) = 1 - \frac{c\alpha}{\omega_1 \pi} \int_{-\infty}^{\infty} \frac{e^{-y^2}\,(x_1 + y)\, dy}{a^2\left(1 + \frac{E_1^2}{E_o^2} + \frac{E_2^2}{E_o^2}\frac{a^2 + (x_1 + y)^2}{a^2 + (x_2 + y)^2}\right) + (x_1 + y)^2}.$$

6.3.1-5

Although 6.3.1-4 and 5 contain quite complicated integrals and will not be evaluated here, we can make some qualitative statements about the effect of the field E_2 for the case $a(1 + E_1^2/E_o^2 + E_2^2/E_o^2)^{\frac{1}{2}} \ll 1$. For this case, the integral in 6.3.1-4 is determined essentially by the contribution near $y \approx -x_1$. Defining a new variable $z = x_1 + y$, 6.3.1-4 becomes

$$\frac{\partial E_1}{\partial z} = \frac{\alpha a E_1}{\pi} \int_{-\infty}^{\infty} \frac{e^{-(z - x_1)^2} dz}{a^2 \left(1 + \frac{E_1^2}{E_o^2} + \frac{E_2^2}{E_o^2} \frac{a^2 + z^2}{a^2 + (x_2 - x_1 + z)^2} \right) + z^2} \quad . \qquad 6.3.1\text{-}6$$

Thus the integral is determined primarily by the contribution near $z = 0$. Limiting the integration to this range gives

$$\frac{\partial E_1}{\partial z} \simeq \frac{\alpha a E_1}{\pi} \int_{-\delta}^{\delta} \frac{e^{-x_1^2} dz}{a^2 \left(1 + \frac{E_1^2}{E_o} + \frac{E_2^2}{E_o^2} \frac{a^2}{a^2 + (x_2 - x_1)^2} \right) + z^2} \quad , \qquad 6.3.1\text{-}7$$

where we have put $z = 0$ except in the rapidly-varying denominator. We can now extend the limits of integration in 6.3.1-7 to $\pm \infty$ without appreciably changing the value of the integral, and obtain

$$\frac{\partial E_1}{\partial z} \simeq \frac{\alpha E_1 e^{-x_1^2}}{\left(1 + \frac{E_1^2}{E_o^2} + \frac{E_2^2}{E_o^2} \frac{a^2}{a^2 + (x_2 - x_1)^2} \right)^{\frac{1}{2}}} \quad , \qquad 6.3.1\text{-}8$$

which reduces to the dominant term of the single-wave result, 6.2.2-8, for $E_2 = 0$. Thus the saturation due to the field E_2 is $a^2/(a^2 + (x_2 - x_1)^2) = \gamma^2/(\gamma^2 + (\omega_2 - \omega_1)^2)$ as strong as that due to E_1 . Since $|\omega_2 - \omega_1| \gg \gamma$, this is small. This is in agreement with the inhomogeneous character of the strongly Doppler broadened line, and is markedly different from the results for the homogeneously broadened line, 6.3.1-2.

The integral in 6.3.1-5 for the index of refraction is more difficult, due to the factor $(x_1 + y)$ in the numerator which makes the contribution outside the range $y \sim - x_1$ more important. By comparing 6.3.1-5 with 6.2.2-2, we see that the factor $(a^2 + (x_1 + y)^2)/(a^2 + (x_2 + y)^2)$ in the denominator of 6.3.1-5 becomes very large near $y \sim - x_2$ and effectively removes the contribution to the index of refraction which would have come from that range of integration. The width of this negation, relative to the width of the integral is approximately aE_2/E_o. The saturation effect of E_2 on the index at ω_1 will therefore be small, since we have assumed $a(1 + E_1^2/E_o^2 + E_2^2/E_o^2)^{\frac{1}{2}} \ll 1$; and the index will be given essentially by the single-wave value, 6.2.2-9, which is very nearly equal to the linear index, 4.3-18. However, the effect of E_2 in 6.3.1-5 is in general more pronounced than that of E_1, since the latter comes from the range of integration $y \sim - x_1$ where the integrand is nearly zero.

The above discussion has a simple physical interpretation in terms of the "hole-burning" ideas introduced by Bennett (2,15). As pointed out in the discussion following 6.2.2-9, the effect of E_1 on the index at ω_1 is small because the latter arises primarily due to atoms outside the range of saturation of E_1. The effect of E_2 on the index at ω_1 is small because for $aE_2/E_o \ll 1$ a relatively small number of atoms is affected by the E_2 saturation. If aE_2/E_o becomes larger, the effect of E_2 correspondingly increases because more atoms are affected.

Another interesting case which we shall mention only briefly is the following: if we repeat the derivation of 6.3.1-4 for the case of two waves traveling in opposite directions, we find

$$\frac{\partial E_1}{\partial z} = \frac{\alpha a E_1}{\pi} \int_{-\infty}^{\infty} \frac{e^{-y^2}\, dy}{a^2 \left(1 + \frac{E_1^2}{E_o^2} + \frac{E_2^2}{E_o^2} \frac{a^2 + (x_1 + y)^2}{a^2 + (x_2 - y)^2} \right) + (x_1 + y)^2} \quad . \qquad 6.3.1\text{-}9$$

The interesting case arises when $x_2 = - x_1$, in which case 6.3.1-9 becomes

$$\frac{\partial E_1}{\partial z} = \frac{\alpha a E_1}{\pi} \int_{-\infty}^{\infty} \frac{e^{-y^2}\, dy}{a^2 \left(1 + \frac{E_1^2}{E_o^2} + \frac{E_2^2}{E_o^2} \right) + (x_1 + y)^2} \quad . \qquad 6.3.1\text{-}10$$

We see that this is the same as the single-wave results, 6.2.2-4 or 6.2.2-8, except the saturation is determined by the sum of the intensities of the fields. This is of course physically due to the fact that for this case the gain for the two waves is determined by the same atoms, even though $|\omega_1 - \omega_2| \gg \gamma$.

6.3.2 $|\omega_1 - \omega_2| \ll \gamma_a, \gamma_b$; Stationary Atoms

Subsituting 6.3-2 into 6.1.2-9, we obtain for this case

$$\frac{\omega P(\underline{r},t)}{2\epsilon_o c} = \frac{\alpha_o \gamma}{\gamma^2 \left(1 + \frac{E_1^2 + E_2^2 + 2E_1E_2 \cos\Delta_{12}}{E_o^2}\right) + (\omega_o - \omega)^2}$$

$$\times \Big[E_1 \gamma \sin(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1) - E_1(\omega_o - \omega_1) \cos(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1)$$

$$+ E_2 \gamma \sin(\underline{k}_2 \cdot \underline{r} - \omega_2 t + \varphi_2) - E_2(\omega_o - \omega_2) \cos(\underline{k}_2 \cdot \underline{r} - \omega_2 t + \varphi_2)\Big] ,$$

6.3.2-1

where in the denominator we have neglected $\omega_2 - \omega_1$ compared to γ. Because of the $\cos\Delta_{12}$ in the denominator, 6.3.2-1 contains the frequencies $\omega_1 \pm n(\omega_o - \omega_2)$ and $\omega_2 \pm n(\omega_1 - \omega_2)$ for integral n. We can obtain the terms at each frequency by expanding 6.3.2-1 in a Fourier series. This is done in Appendix III and we obtain

$$\frac{\omega P(\underline{r},t)}{2\epsilon_o c} = \frac{\alpha_o}{\gamma} \Big[E_1 \gamma \sin(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1) - E_1(\omega_o - \omega_1)\cos(\underline{k} \cdot \underline{r} - \omega_1 t + \varphi_1)$$

$$+ E_2 \gamma \sin(\underline{k}_2 \cdot \underline{r} - \omega_2 t + \varphi_2) - E_2(\omega_o - \omega_2) \cos(\underline{k}_2 \cdot \underline{r} - \omega_2 t + \varphi_2)\Big]$$

$$\times \sum_{m=o}^{\infty} C_m \cos m\Big[(\underline{k}_1 - \underline{k}_2) \cdot \underline{r} - (\omega_1 - \omega_2)t + \varphi_1 - \varphi_2\Big] ,$$

6.3.2-2

where

$$C_m = \frac{2 - \delta_{om}}{1 + y^2 + (E_1^2 + E_2^2)/E_o^2} \frac{\left[\sqrt{1 - a_o^2} - 1\right]^m}{a_o^m \sqrt{1 - a_o^2}} , \qquad 6.3.2\text{-}3$$

$$a_o = \frac{2E_1E_2/E_o^2}{1 + y^2 + (E_1^2 + E_2^2)/E_o^2} \quad , \qquad 6.3.2\text{-}4$$

and

$$y = (\omega_o - \omega)/\gamma \quad . \qquad 6.3.2\text{-}5$$

By expanding 6.3.2-2, we find

$$\frac{\omega P(\underline{r},t)}{2\epsilon_o c} = \frac{\alpha_o}{2\gamma} \sum_{m=o}^{\infty} C_m \Big\{ \gamma E_1 \sin\Big[(\underline{k}_1 + m(\underline{k}_1 - \underline{k}_2)) \cdot \underline{r} - (\omega_1 + m(\omega_1 - \omega_2))t$$

$$+ (\varphi_1 + m(\varphi_1 - \varphi_2))\Big] + \gamma E_1 \sin\Big[(\underline{k}_1 - m(\underline{k}_1 - \underline{k}_2)) \cdot \underline{r} - (\omega_1 - m(\omega_1 - \omega_2))t + \varphi_1 - m(\varphi_1 - \varphi_2)\Big]$$

$$- (\omega_o - \omega_1)E_1 \cos\Big[(\underline{k}_1 + m(\underline{k}_1 - \underline{k}_2)) \cdot \underline{r} - (\omega_1 + m(\omega_1 - \omega_2))t + (\varphi_1 + m(\varphi_1 - \varphi_2))\Big]$$

$$- (\omega_o - \omega_1)E_1 \cos\Big[(\underline{k}_1 - m(\underline{k}_1 - \underline{k}_2)) \cdot \underline{r} - (\omega_1 - m(\omega_1 - \omega_2))t + (\varphi_1 - m(\varphi_1 - \varphi_2))\Big]$$

$$+ \text{(same with subscripts 1 and 2 interchanged)} \Big\} \quad . \qquad 6.3.2\text{-}6$$

Collecting terms in 6.3.2-6, we finally have, for the polarization induced at various frequencies by the fields E_1 and E_2 ,

$$\frac{\omega P_s(\omega_1 \pm m(\omega_1 - \omega_2))}{2\epsilon_o c} = \frac{\alpha_o}{2} \Big[E_1 \, C_m (1 + \delta_{om}) + E_2 \, C_{m\pm 1}(1 + \delta_{o,m\pm 1}) \Big] \quad , \qquad 6.3.2\text{-}7$$

$$\frac{\omega P_c(\omega_1 \pm m(\omega_1 - \omega_2))}{2\epsilon_o c} = - \frac{\alpha_o(\omega_o - \omega)}{2\gamma} \Big[E_1 \, C_m (1 + \delta_{om}) + E_2 \, C_{m\pm 1}(1 + \delta_{o,m\pm 1}) \Big] \quad . \qquad 6.3.2\text{-}8$$

6.3.2-7 and 8 hold only as long as none of the fields induced at frequencies other than ω_1,ω_2 become appreciable compared to E_o, and only for m's such that $\omega_1 \pm m(\omega_1 - \omega_2) \ll \gamma_a,\gamma_b$. For other m's, the polarization will be smaller than indicated by 6.3.2-7 and 8, and will have a more complex frequency dependence. Thus 6.3.2-7 and 8 set an upper limit on the magnitude of the polarization at a given frequency due to E_1, E_2.

As previously discussed, for frequencies other than ω_1,ω_2 we are interested only in the magnitude of the polarization due to E_1 and E_2. From 6.3.2-7 and 8 this is, for $\omega_1 \pm m(\omega_1 - \omega_2) \neq \omega_1,\omega_2$,

$$\frac{\omega P'_s(\omega_1 \pm m(\omega_1 - \omega_2)}{2\epsilon_o c} = \frac{\alpha_o}{2}\sqrt{1 + y^2}\left[E_1 C_m + E_2 C_{m\pm1}\right] . \quad 6.3.2\text{-}9$$

In addition, the induced fields at frequencies other than ω_1 or ω_2 will themselves induce a saturated polarization given by

$$\frac{\omega P_s}{2\epsilon_o c} = \alpha_o C_o E , \qquad 6.3.2\text{-}10$$

$$\frac{\omega P_c}{2\epsilon_o c} = -\frac{\alpha_o(\omega_o - \omega)}{\gamma} C_o E , \qquad 6.3.2\text{-}11$$

where E is the field amplitude at the particular frequency.

Using the above results in 2.3.2-20 and 21, we find

$$\frac{\partial E_1}{\partial z} = \alpha_o \left(E_1 C_o + \frac{E_2 C_1}{2}\right) , \qquad 6.3.2\text{-}12$$

$$\frac{\partial E_2}{\partial z} = \alpha_o \left(E_2 \, C_o + \frac{E_1 C_1}{2} \right) \quad , \qquad 6.3.2\text{-}13$$

$$n(\omega_1) = 1 - \frac{c\alpha_o}{\omega_1} \frac{(\omega_o - \omega_1)}{\gamma} \left[C_o + \frac{E_2 C_1}{2E_1} \right] \quad , \qquad 6.3.2\text{-}14$$

$$n(\omega_2) = 1 - \frac{c\alpha_o}{\omega_2} \frac{(\omega_o - \omega_2)}{\gamma} \left[C_o + \frac{E_1 C_1}{2E_2} \right] \quad , \qquad 6.3.2\text{-}15$$

and for $\omega \pm m = \omega_1 \pm m(\omega_1 - \omega_2) \neq \omega_1$ or ω_2 ,

$$\frac{\partial E_{\pm m}}{\partial z} = \alpha_o \quad E_{\pm m} \, C_o + \frac{\sqrt{1 + y^2}}{2} \left[(E_1 C_m + E_2 C_{m \pm 1}) \right] \quad , \qquad 6.3.2\text{-}16$$

$$n(\omega_{\pm m}) = 1 - \frac{c\alpha_o}{\omega_{\pm m}} \frac{(\omega_o - \omega_{\pm m})}{\gamma} C_o \quad , \qquad 6.3.2\text{-}17$$

where for 6.3.2-16 and 17 we have assumed $\Delta k = 0$ because $|\omega_1 - \omega_2| \ll \gamma_a, \gamma_b$.

Some of the coefficients C_m are plotted in Figure 16 for $E_1 = E_2$, and $y = 0$. For $E_1 = E_2$, the polarization coefficients 6.3.2-7, 8 and 9 are generally determined by the sum of two successive C_m , and we note that this sum is given by the vertical distance between the corresponding curves, taking into account the log scale. Thus we can see directly from Figure 16 how the polarization at various frequencies changes with increasing field strength.

If E_1/E_o and E_2/E_o are small compared to one, we can find the lowest order nonlinear corrections. From 6.3.2-3 and 4 we find

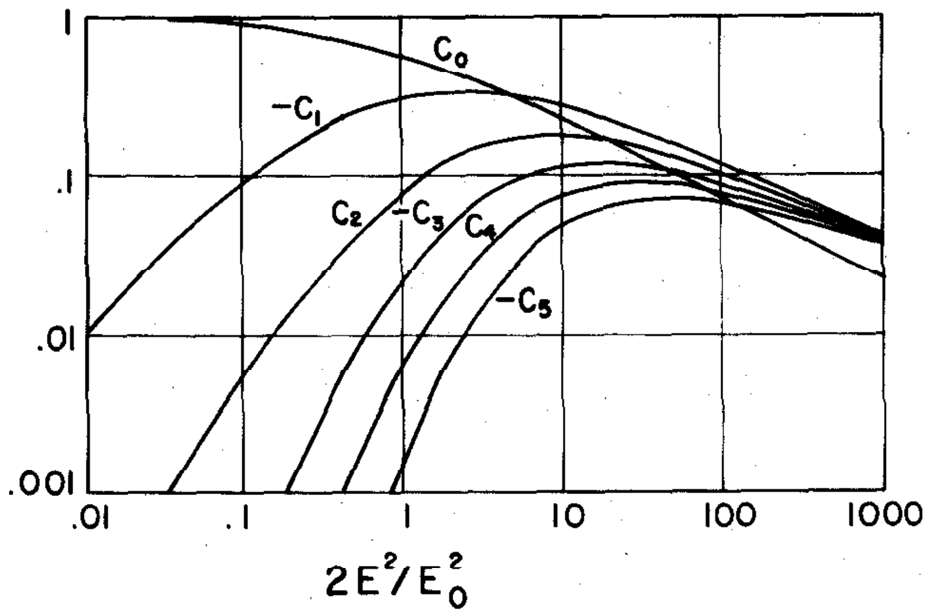


FIGURE 16 THE FOURIER COEFFICIENTS FOR HOMOGENEOUS BROADENING

$$C_o \simeq \frac{1}{1 + y^2 + (E_1^2 + E_2^2)/E_o} \simeq \frac{1}{1 + y^2}\left[1 - \frac{E_1^2 + E_2^2}{E_o^2}\,\frac{1}{1 + y^2}\right] , \quad 6.3.2\text{-}18$$

and

$$C_1 \simeq \frac{2E_1E_2/E_o^2}{[1 + y^2]^2} ,$$

where again $y = (\omega_o - \omega)/\gamma$. Then, for example, 6.3.2-12 becomes

$$\frac{\partial E_1}{\partial z} = \frac{\alpha_o E_1}{1 + y^2}\left[1 - \frac{E_1^2 + 2E_2^2}{E_o^2}\,\frac{1}{1 + y^2}\right] , \quad 6.3.2\text{-}19$$

which gives the same lowest order nonlinear corrections, including the modulation contribution, as were obtained in 5.2.2, for $|\omega_1 - \omega_2| \ll \gamma_a, \gamma_b$.

Another interesting limiting case is $E_1 = E_2 = E$ and $E^2/E_o^2 \gg 1$. From 6.3.2-4 we have

$$a_o \simeq 1 - E_o^2(1 + y^2)/2E^2 , \quad 6.3.2\text{-}20$$

and from 6.3.2-3 we find

$$C_m \simeq (2 - \delta_{om})\,(-1)^m E_o \frac{(1 - mE_o\sqrt{1 + y^2}/E)}{2E\sqrt{1 + y^2}} . \quad 6.3.2\text{-}21$$

Therefore,

$$2C_o + C_1 \simeq E_o^2/E^2 , \quad 6.3.2\text{-}22$$

and for $m, m - 1 \neq 0$,

$$C_m + C_{m\pm 1} \simeq \pm(-1)^m E_o^2/E^2 \quad . \qquad 6.3.2\text{-}23$$

The polarization due to E_1 and E_2 at each frequency is therefore equal in magnitude at all frequencies, and is of order $E_o^2/E^2 \ll 1$, compared to the linear polarization. This of course does not take into account the frequency-dependent decrease in magnitude for m's such that $m|\omega_1 - \omega_2| > \gamma_a, \gamma_b$. Physically, this is a situation where the fields are so strong that they drive the upper and lower levels to equal population, so that $\rho_{aa} - \rho_{bb} \approx 0$.

6.3.3 $|\omega_1 - \omega_2| \ll \gamma_a, \gamma_b$; Maxwellian Velocity Distribution

Substituting 6.3-2 into 6.1.2-9 and using 4.1-1, we obtain for a Maxwellian velocity distribution of excited atoms

$$\frac{\omega P(\underline{r},t)}{2\epsilon_o c} = \frac{\alpha}{\pi}\int_{-\infty}^{\infty} \frac{e^{-y^2}\,dy}{a^2\left(1 + \dfrac{E_1^2 + E_2^2 + 2E_1E_2\cos\Delta_{12}}{E_o^2}\right) + (x+y)^2}$$

$$\times\Big[E_1 a \sin(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1) - E_1(x_1 + y)\cos(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1)$$

$$+ E_2 a \sin(\underline{k}_2 \cdot \underline{r} - \omega_2 t + \varphi_2) - E_2(x_2 + y)\cos(\underline{k}_2 \cdot \underline{r} - \omega_2 t + \varphi_2)\Big] \,,$$

6.3.3-1

where the two velocity integrations over velocities perpendicular to $\underline{k}$ have been carried out, and the various quantities have their previous meanings, i.e., $a = \gamma/k_o u$, $\alpha = \alpha_o a\sqrt{\pi}$, $x = (\omega_o - \omega)/k_o u$, $y = V/u$. As was the case with stationary atoms, the polarization 6.3.3-1 contains the frequencies $\omega_1 \pm m(\omega_1 - \omega_2)$ for integral m. We can write 6.3.3-1 in terms of the complex error function discussed in Appendix I, but in general we could not separate the various frequency contributions. We shall instead work only with the interesting limiting case

$$a\left(1 + \frac{E_1^2 + E_2^2}{E_o^2}\right)^{\frac{1}{2}} \ll 1 \quad , \qquad 6.3.3\text{-}2$$

corresponding to a strongly inhomogeneously broadened line, for which we can expand 6.3.3-1 to first order in a. Using the results of Appendix I, we find for this case

$$\frac{\omega P(\underline{r},t)}{2\epsilon_o c} = \alpha \left\{ \left[e^{-x^2}\left(1 + \frac{E_1^2 + E_2^2 + 2E_1E_2\cos\Delta_{12}}{E_o^2}\right)^{-\frac{1}{2}} - \frac{2a}{\sqrt{\pi}}\left(1 - 2xF(x)\right) \right] \left[E_1 \sin(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1) + E_2 \sin(\underline{k}_2 \cdot \underline{r} - \omega_2 t + \varphi_2) \right] - \left[\frac{2F(x)}{\sqrt{\pi}} - 2a\left(1 + \frac{E_1^2 + E_2^2 + 2E_1E_2\cos\Delta_{12}}{E_o^2}\right)^{\frac{1}{2}} x\, e^{-x^2} \right] \times \left[E_1 \cos(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1) + E_2 \cos(\underline{k}_2 \cdot \underline{r} - \omega_2 t + \varphi_2) \right] \right\} . \qquad 6.3.3\text{-}3$$

Since for the case 6.3.3-2 the cosine terms of 6.3.3-3 are not appreciably changed by saturation, we shall deal only with the sine terms and shall assume that the index of refraction is given by its linear value,

$$n(\omega) = 1 - \frac{c\alpha}{\omega}\frac{2F(x)}{\sqrt{\pi}} \quad , \qquad 6.3.3\text{-}4$$

where $x = (\omega_o - \omega)/k_o u$ as before. We shall also temporarily neglect the unmodulated sine terms in 6.3.3-3, but will add them again later.

In order to evaluate the contribution of 6.3.3-3 to various frequencies present in the polarization, we need the expansion

$$\left(1 + \frac{E_1^2 + E_2^2 + 2E_1E_2\cos\Delta_{12}}{E_o^2}\right)^{-\frac{1}{2}} = \sum_{m=o}^{\infty} C'_m \cos m\Delta_{12} \quad . \qquad 6.3.3\text{-}5$$

6.3.3-5 is considered in Appendix III, and we find for the sine terms of the polarization 6.3.3-3

$$\frac{\omega P(\underline{r},t)}{2\epsilon_o c} = \frac{\alpha e^{-x^2}}{2}\sum_{m=o}^{\infty} C'_m \Big\{ E_1 \sin\big[(\underline{k}_1 + m(\underline{k}_1 - \underline{k}_2))\cdot\underline{r} - (\omega_1 + m(\omega_1 - \omega_2)t$$

$$+ (\varphi_1 + m(\varphi_1 - \varphi_2))\big] + E_1 \sin\big[(\underline{k}_1 - m(\underline{k}_1 - \underline{k}_2))\cdot\underline{r} - (\omega_1 - m(\omega_1 - \omega_2))t$$

$$+ (\varphi_1 - m(\varphi_1 - \varphi_2))\big] + \text{(same with subscripts 1,2 interchanged)}\Big\} , \qquad 6.3.3\text{-}6$$

where

$$C'_m = \frac{(2 - \delta_{om})(-1)^m\sqrt{8/a'_o}}{\pi\left(1 + \frac{E_1^2 + E_2^2}{E_o^2}\right)^{\frac{1}{2}}} \int_o^d \frac{(d^2 - t^2)^{m-\frac{1}{2}}}{(c^2 + t^2)^{\frac{1}{2}}}\,dt \quad , \qquad 6.3.3\text{-}7$$

$$d^2 = \frac{(1 - \sqrt{1 - a_o'^2})}{a_o'} \quad , \qquad 6.3.3\text{-}8$$

$$c^2 = \frac{2\sqrt{1 - a_o'^2}}{a_o'} \quad , \qquad 6.3.3\text{-}9$$

and

$$a_o' = \frac{2E_1E_2/E_o^2}{1 + (E_1^2 + E_2^2)/E_o^2} \quad . \qquad 6.3.3\text{-}10$$

An alternative evaluation of the C_m' , also given in Appendix III, gives an infinite series expression, viz.,

$$C_m' = \frac{(2 - \delta_{om})(- a_o'/4)^m}{\left(1 + \frac{E_1^2 + E_2^2}{E_o^2}\right)^{\frac{1}{2}}} \sum_{p=o}^{\infty} \left(\frac{a_o'^2}{16}\right)^p \frac{(4p + 2m - 1)!!}{p!\ (m + p)!} \qquad 6.3.3\text{-}11$$

where we define $(4p + 2m - 1)!! = 1$ for $p = m = 0$.

Collecting terms in 6.3.3-6, we find for the polarization amplitudes due the fields E_1 and E_2

$$\frac{\omega P_s(\omega_1 \pm m(\omega_1 - \omega_2))}{2\epsilon_o c} = \frac{\alpha e^{-x^2}}{2} \left[E_1 C_m'(1 + \delta_{om}) + E_2 C_{m\pm 1}'(1 + \delta_{o,m\pm 1})\right] \quad .$$

6.3.3-12

For $\omega_{\pm m} = \omega_1 \pm m(\omega_1 - \omega_2) \neq \omega_1$ or ω_2 , we will find a polarization due to the induced field

$$\frac{\omega P_{s(\pm m)}}{2\epsilon_o c} = \alpha \left[e^{-x^2} E_{\pm m} C_o' - \frac{2aE_{\pm m}}{\sqrt{\pi}} (1 - 2xF(x)) \right] . \qquad 6.3.3\text{-}13$$

Using 6.3.3-12 and 13 in 2.3.2-21, and including the term previously neglected in 6.3.3-3, we have

$$\frac{\partial E_1}{\partial z} = \alpha \left[e^{-x^2} \left(E_1 C_o' + \frac{E_2 C_1'}{2} \right) - \frac{2aE_1}{\sqrt{\pi}} (1 - 2xF(x)) \right] , \qquad 6.3.3\text{-}14$$

$$\frac{\partial E_2}{\partial z} = \alpha \left[e^{-x^2} \left(\frac{E_1 C_1'}{2} + E_2 C_o' \right) - \frac{2aE_2}{\sqrt{\pi}} (1 - 2xF(x)) \right] ,$$

$$\frac{\partial E_{\pm m}}{\partial z} = \alpha \left[e^{-x^2} \left(E_{\pm m} C_o' + E_1 C_m' + E_2 C_{m\pm 1}' \right) - \frac{2aE_{\pm m}}{\sqrt{\pi}} (1 - 2xF(x)) \right] ,$$

6.3.3-15

where we have assumed $\Delta k = 0$ for 6.3.3-15.

Some of the C_m' coefficients are plotted in Figure 17 for $E_1 = E_2$, along with the corresponding C_m coefficients from Figure 16 for comparison. The relative amount of "spreading" can be seen to be very similar for the homogeneous and inhomogeneous cases.

For E_1/E_o, $E_2/E_o \ll 1$, Appendix III gives

$$C_o' \approx 1 - \frac{E_1^2 + E_2^2}{2E_o^2} , \qquad 6.3.3\text{-}16$$

and

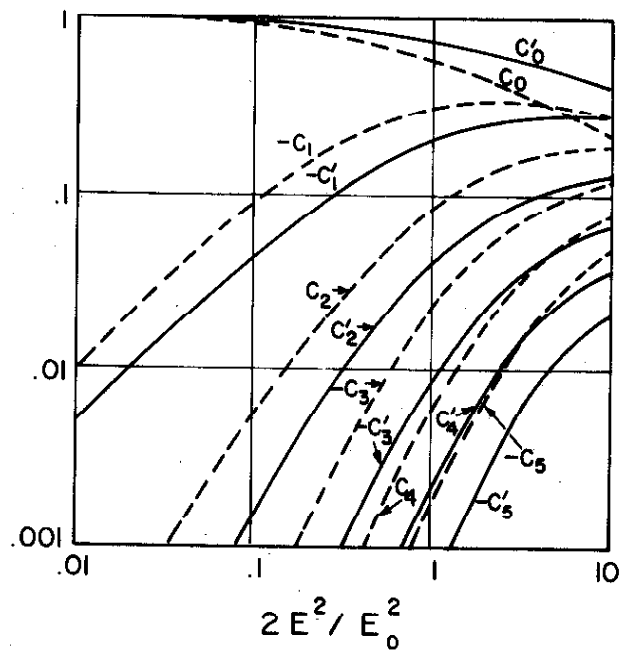


FIGURE 17 THE FOURIER COEFFICIENTS FOR INHOMOGENEOUS BROADENING, COMPARED TO THE HOMOGENEOUS COEFFICIENTS.

$$C_1' \simeq - a_o'/2 \simeq - E_1 E_2/E_o^2 \quad , \qquad 6.3.3\text{-}17$$

so that, for example, 6.3.3-14 becomes

$$\frac{\partial E_1}{\partial z} = \alpha\, E_1 \left[e^{-x^2} \left(1 - \frac{E_1^2 + 2E_2^2}{2E_o^2} \right) - \frac{2a}{\pi} \left(1 - 2xF(x)\right) \right] \quad , \qquad 6.3.3\text{-}18$$

which is the linear gain with lowest order nonlinear corrections, for $|\omega_1 - \omega_2| \ll \gamma_a \gamma_b$.

As before, the above results hold only for $m|\omega_1 - \omega_2| \ll \gamma_a, \gamma_b$ and only as long as none of the induced fields become appreciable compared to E_o . We can obtain an estimate of the induced fields expected by neglecting the saturation effect of the induced fields and fixing the coefficients at their $z = 0$ values. Thus for $E_1 = E_2 = E$ and $2E^2/E_o^2 = 5$, we find from Figure

$$C_o' \simeq .5,\ C_1' \simeq - .3,\ C_2' \simeq .1,\ C_3' \simeq - .05,\ C_4' \simeq .03 \quad .$$

If we take a moderate value of $\alpha = 2$ per meter and an amplifier one half meter long, we find at line center, using 6.3.3-15,

$$E_{+1} \sim .2\ E \quad \text{at} \quad 2\omega_1 - \omega_2 \ ,$$

$$E_{+2} \sim .05\ E \quad \text{at} \quad 3\omega_1 - 2\omega_2 \ , \qquad 6.3.3\text{-}19$$

$$E_{+3} \sim .01\ E \quad \text{at} \quad 4\omega_1 - 3\omega_2 \ .$$

For the 3.4 micron line in neon, this means that for 3 mw/cm^2 input at ω_1 and ω_2 , we might expect an output of roughly 0.1 mw/cm^2 at

$2\omega_1 - \omega_2$, 10 μw/cm^2 at $3\omega_1 - 2\omega_2$ and 0.1 μw/cm^2 at $4\omega_1 - 3\omega_2$.

6.4 More Than Two Waves; Super-radiance

For more than two waves, the physical results of section 6.3 are generally unchanged but are more complicated in detail. For example, for $|\omega_i - \omega_j| \gg \gamma_a, \gamma_b$ so that modulation effects may be neglected, 6.3.1-4 can be generalized to give

$$\frac{\partial E_1}{\partial z} = \frac{\alpha a E_1}{\pi} \int_{-\infty}^{\infty} \frac{e^{-y^2}\, dy}{a^2 \left(1 + \frac{E_1^2}{E_o^2} + \sum_{i \neq 1} \frac{E_i^2}{E_o^2} \frac{a^2 + (x_1 + y)^2}{a^2 + (x_i + y)^2}\right) + (x_1 + y)^2} \qquad 6.4\text{-}1$$

For $|\omega_i - \omega_j| \ll \gamma_a, \gamma_b$, we can generalize 6.3.3-1 to find the polarization

$$\frac{\omega P(\underline{r},t)}{2\epsilon_o c} = \frac{\alpha}{\pi} \sum_i \int_{-\infty}^{\infty} \frac{e^{-y^2} dy \left[E_i a \sin(\underline{k}_i \cdot \underline{r} - \omega_i t + \varphi_i) - E_i (x_i + y) \cos(\underline{k}_i \cdot \underline{r} - \omega_i t + \varphi_i)\right]}{a^2 \left(1 + \sum_j \frac{E_j^2}{E_o^2} + \sum_{j \neq k} \frac{E_j E_k}{E_o^2} \cos\Delta_{jk}\right) + (x_i + y)^2} \qquad 6.4\text{-}2$$

We see that 6.4-1 contains the saturation effects of all waves. In addition to these saturation effects, 6.4-2 contains the effects of modulation of the population inversion at all possible combinations of the difference frequencies $\omega_i - \omega_j$. Since no new physical effects are involved, we will not attempt a treatment of these cases here.

There is one interesting generalization of 6.4-2 which we will discuss briefly. If we have a large number of waves within a frequency range γ which have random phases and approximately equal amplitudes, the $\cos\Delta_{jk}$ summation in 6.4-2 will give zero, and 6.4-2 will become

$$\frac{\omega P(\underline{r}\, t)}{2\epsilon_o c} = \frac{\alpha}{\pi} \sum_i \int_{-\infty}^{\infty} \frac{e^{-y^2} dy E_i \left[a \sin(\underline{k}_i \cdot \underline{r} - \omega_i t + \varphi_i) - (x_i + y)\cos(\underline{k}_i \cdot \underline{r} - \omega_i t + \varphi_i) \right]}{a^2(1 + \sum_j E_j^2/E_o^2) + (x_i + y)^2} \qquad 6.4\text{-}3$$

Performing the integration gives

$$\frac{\partial E_i}{\partial z} = \frac{\alpha E_i}{(1 + \sum_j E_j^2/E_o^2)^{\frac{1}{2}}} \operatorname{Re} w(x_i + ib) \quad , \qquad 6.4\text{-}4$$

where

$$b = a(1 + \sum_j E_j^2/E_o^2)^{\frac{1}{2}} \quad .$$

Since x_i does not change appreciably for the different fields, we can replace all x_i by some average value x. Written in terms of the relative intensities

$$\mathcal{I}_i = I_i/I_o = E_i^2/E_o^2 \quad ,$$

6.4-4 becomes

$$\frac{\partial \mathcal{I}_i}{\partial z} = \frac{2\alpha\, \mathcal{I}_i}{(1 + \sum_j \mathcal{I}_j)^{\frac{1}{2}}} \operatorname{Re} w(x + ib) \quad . \qquad 6.4\text{-}5$$

Summing 6.4-5 over all i and using

$$I_{total} = \sum_j I_j \quad ,$$

we find

$$\frac{\partial I_{total}}{\partial z} = \frac{2\alpha \ I_{total}}{(1 + I_{total})^{\frac{1}{2}}} \ \mathrm{Re}\ w(x + ib) \quad . \qquad 6.4\text{-}6$$

In the limit $b \ll 1$, 6.4-6 becomes

$$\frac{\partial I_{total}}{\partial z} = \frac{2\alpha \ I_{total} \ e^{-x^2}}{(1 + I_{total})^{\frac{1}{2}}} - \frac{4\alpha a}{\sqrt{\pi}} (1 - 2xF(x)) \quad , \qquad 6.4\text{-}7$$

which is of the same form as 6.2.2-10 for a single wave. For $x = 0$ 6.4-7 looks exactly like 6.2.2-11, except I is replaced by I_{total} .

The above discussion gives us a means of approximately describing the result of a broad, incoherent input. In particular, we can describe the "super-radiant" emission of a high gain amplifier (16), which arises due to the amplification of spontaneous emission. We see from the above discussion and especially 6.4-7 that there can be no modulation effects and that the amplification is frequency dependent, so that the super-radiant output will have a narrower frequency width than the spontaneous emission line, as is well known.

It is interesting to note that the above results may explain why Gordon, White and Rigden (16), who used a super-radiant source for their experiments, were able to fit their data to a single-frequency saturation curve.

Another interesting implication of the above discussion is that a high gain amplifier will be saturated by the super-radiant emission and for an inhomogeneously broadened line the center of the line will be saturated more strongly than the wings. Thus if one attempts to measure the small signal gain of the amplifier, the gain line will be broadened so that the observed gain line width will be larger than the spontaneous emission line width.

6.5 Discussion

In the preceding sections of this chapter we have discussed some strong nonlinear effects, primarily gain saturation and the behavior and saturation of some modulation effects. Although several assumptions were made which limit the range of validity of this discussion, these assumptions simplified the discussion so that the physical processes involved were readily apparent. For example, the coherent modulations of the population inversion density were made very clear. Also, the loss of generality is mitigated by the availability of the detailed discussion of lowest order nonlinear effects in Chapter 5; in particular the discussion of polarization effects, frequency dependence of modulation effects, and cases with more general wave vectors largely carries over to situations where the strong nonlinear effects are important.

We can qualitatively discuss several aspects of the theory presented above; in particular we will discuss holeburning in 6.5.1, frequency dependence of modulation effects in 6.5.2, and the high-field limit for the inhomogeneously broadened line in 6.5.3.

6.5.1 Holeburning

It is interesting to discuss some of the above results for a Doppler broadened line in terms of the holeburning ideas proposed by Bennett (2,15). Since the total incremental gain, $g = (1/E)\,\partial E/\partial z$, is determined by an integration over velocities, we can consider the contribution due to those atoms with a velocity $\underline{v}$. This contribution is proportional to the population inversion density of atoms with that velocity, multiplied by a frequency dependent factor,

$$g(\underline{v},\omega_1) \propto \frac{\gamma^2\,[\rho_{aa}(\underline{v}) - \rho_{bb}(\underline{v})]}{\gamma^2 + (\omega_o - \omega_1 + \underline{k}\cdot\underline{v})^2} \quad . \qquad 6.5.1\text{-}1$$

For a single wave, the population inversion density (PID) was given by 6.2-1:

$$\rho_{aa} - \rho_{bb} = \frac{NW(\underline{v})}{1 + \frac{E^2}{E_o^2}\,\frac{\gamma^2}{\gamma^2 + (\omega_o - \omega + \underline{k}\cdot\underline{v})^2}} \quad . \qquad 6.5.1\text{-}2$$

Thus the PID as a function of velocity has, relative to $W(\underline{v})$, a Lorentzian-shaped dip or "hole" for velocities $\underline{v}$ such that the interaction frequency, $\omega - \underline{k}\cdot\underline{v}$, is nearly equal to ω_o. The width of this hole in frequency units is

$$\Delta(\underline{k}\cdot\underline{v}) = 2\gamma(1 + E^2/E_o^2)^{\frac{1}{2}} \quad , \qquad 6.5.1\text{-}3$$

and the depth, relative to unity, is

$$\frac{E^2/E_o^2}{1 + E^2/E_o^2} \quad . \tag{6.5.1-4}$$

Let us consider the situation where we have, in addition to the strong field E at ω, a field E_1 at ω_1, where E_1 is so small that it has no effect on the PID. Then the incremental gain at ω_1 is found by substituting 6.5.1-2 into 6.5.1-1, giving

$$g(\underline{v},\omega_1) \propto \frac{\gamma^2 W(\underline{v})}{\gamma^2\left(1 + \frac{E^2}{E_o^2}\frac{\gamma^2 + (\omega_o - \omega_1 + \underline{k}\cdot\underline{v})^2}{\gamma^2 + (\omega_o - \omega + \underline{k}\cdot\underline{v})^2}\right) + (\omega_o - \omega_1 + \underline{k}\cdot\underline{v})^2}$$

6.5.1-5

If we consider atoms with velocity $\underline{v}$ such that $\underline{k}\cdot\underline{v} = \omega_1 - \omega_o$, i.e., which are resonant with the field at ω_1, we find, relative to $W(\underline{v})$,

$$g(\omega_1) \propto \frac{1}{\left(1 + \frac{E^2}{E_o^2}\frac{\gamma^2}{\gamma^2 + (\omega_1 - \omega)^2}\right)} \quad , \tag{6.5.1-6}$$

which implies a hole in the gain at ω_1 as a function of frequency. Since the field E_1 interacts with atoms over a frequency width 2γ (the natural line width), the total frequency width of the hole in the incremental gain curve for E_1 will be 2γ plus the width of 6.5.1-6, or $2\gamma[1 + (1 + E^2/E_o^2)^{\frac{1}{2}}]$. This hole is of course centered at ω. Its depth will be essentially the depth of 6.5.1-6, or

$$\frac{E^2/E_o^2}{1 + E^2/E_o^2} \quad ,$$

relative to unity.

For two waves with $|\omega_1 - \omega_2| \gg \gamma_a, \gamma_b$, the PID was given by 6.3-1:

$$\rho_{aa} - \rho_{bb} = \frac{NW(\underline{v})}{1 + \frac{E_1^2}{E_o^2} \frac{\gamma^2}{\gamma^2 + (\omega_o - \omega_1 + \underline{k} \cdot \underline{v})^2} + \frac{E_2^2}{E_o^2} \frac{\gamma^2}{\gamma^2 + (\omega_o - \omega_2 + \underline{k} \cdot \underline{v})^2}}$$

6.5.1-7

Thus for this case there are two holes in the PID, centered about $(\underline{k} \cdot \underline{v}) = \omega_1 - \omega_o$ and $\omega_2 - \omega_o$, and similar to the single hole for a single wave.

For two waves with $|\omega_1 - \omega_2| \ll \gamma_a, \gamma_b$, the PID was given by 6.3-2:

$$\rho_{aa} - \rho_{bb} = \frac{NW(\underline{v})}{1 + \frac{E_1^2 + E_2^2 + 2E_1E_2 \cos\Delta_{12}}{E_o^2} \frac{\gamma^2}{\gamma^2 + (\omega_o - \omega + \underline{k} \cdot \underline{v})^2}} \quad .$$

6.5.1-8

Thus there is a single hole in the PID, whose width and depth vary harmonically at a frequency $|\omega_1 - \omega_2|$. The maximum and minimum widths are

$$2\gamma\left[1+\frac{(E_1+E_2)^2}{E_o^2}\right]^{\frac{1}{2}} \quad \text{and} \quad 2\gamma\left[1+\frac{(E_1-E_2)^2}{E_o^2}\right]^{\frac{1}{2}}, \qquad 6.5.1\text{-}9$$

respectively, and the corresponding depths are

$$\frac{(E_1+E_2)^2/E_o^2}{1+(E_1+E_2)^2/E_o^2} \quad \text{and} \quad \frac{(E_1-E_2)^2/E_o^2}{1+(E_1-E_2)^2/E_o^2}, \qquad 6.5.1\text{-}10$$

relative to unity. This fluctuating hole clearly shows how the PID follows the beating of the two incident fields, for $|\omega_1 - \omega_2| \ll \gamma_a, \gamma_b$.

6.5.2 Frequency Dependence of Modulation Effects

The treatment of modulation effects due to two waves which was given in this chapter did not allow us to study how these effects changed with the frequency separation of the two waves. From Chapter 5 we know that the frequency dependence is determined by γ_a, γ_b and $(\gamma_a + \gamma_b)/2 = \gamma$. In particular, for the lowest order modulation effect, Chapter 5 gives the detailed frequency dependence. The most important frequency dependence is the Lorentzian variation with $\omega_1 - \omega_2$ which has two components with widths $2\gamma_a$ and $2\gamma_b$ and appears in the results for both stationary and moving atoms. In addition there is a Lorentzian frequency dependence with a width 2γ or 4γ, which depends on the frequency spacing relative to ω_o for stationary atoms, and again on the frequency difference $\omega_1 - \omega_2$ for moving atoms.

If we consider the perturbation expansion of section 6.1, we see that terms contributing to the saturation of a particular effect are characterized by terms in the summation with equal frequencies, so that they do not contribute any further frequency dependence. Thus the frequency dependence of a particular modulation effect is determined by the lowest order of the perturbation expansion which contributes to that effect. Therefore the frequency dependence of the lowest order modulation effect is that given in Chapter 5. Each higher order will multiply the previous order's frequency dependence by two Lorentzian factors, one depending on γ_a, γ_b and the other on γ. If the modulation term is at $\omega_1 + n(\omega_1 - \omega_2)$, for example, the frequency dependence will be roughly

$$\left(\frac{1}{\gamma + i\Delta\omega}\right)^n \left(\frac{1}{\gamma_a + i\Delta\omega} + \frac{1}{\gamma_b + i\Delta\omega}\right)\left(\frac{1}{\gamma_a + i2\Delta\omega} + \frac{1}{\gamma_b + i2\Delta\omega}\right) \times \cdots$$

$$\times \left(\frac{1}{\gamma_a + in\Delta\omega} + \frac{1}{\gamma_b + in\Delta\omega}\right) , \qquad 6.5.2\text{-}1$$

where

$$\Delta\omega = \omega_1 - \omega_2 \quad .$$

It is evident that the higher order modulation effects will decrease rapidly with increasing $\Delta\omega$. We should note here again that the results of this chapter show that the modulation effects decrease rapidly with increasing order, even disregarding any dependence on $\Delta\omega$.

6.5.3 The High-Field Limit For the Doppler Broadened Line

In 6.3.3 we made the assumption

$$a\left(1 + \frac{E_1^2 + E_2^2}{E_o^2}\right)^{\frac{1}{2}} \ll 1 , \qquad 6.5.3\text{-}1$$

which physically means that the frequency width of the interaction between the field and excited atoms remains small compared to the Doppler width, i.e., that the field always sees a strongly inhomogeneously broadened line. Of course for a given system (fixed a) the condition 6.5.3-1 will eventually be violated as the field strength is increased, and it is interesting to discuss the nature of this high-field limit.

Physically, we expect that as the field strength increases, thereby increasing the interaction width, the gain line should appear effectively homogeneously broadened, as far as the interaction between the strong field and excited atoms is concerned. Thus for $a(1 + E^2/E_o^2)^{\frac{1}{2}} \gg 1$, we expect the results to be independent of the Doppler width. In order to see this, we consider the asymptotic form of the complex error function for large arguments (38):

$$\sqrt{\pi}\, z\, e^{z^2}\, \mathrm{erfc}\, z \simeq 1 - \frac{1}{2z^2} \quad . \tag{6.5.3-2}$$

Using I-2, we have

$$w(z) \simeq \frac{i}{\sqrt{\pi}} \left(\frac{1}{z} + \frac{1}{2z^3} \right) \quad , \tag{6.5.3-3}$$

for $z \to \infty$. Keeping only the first term of 6.5.3-3, we have

$$w(x + ib) \simeq \frac{i}{\sqrt{\pi}} \frac{1}{x + ib} \quad . \tag{6.5.3-4}$$

For a single wave, 6.2.2-6 becomes, for $b \gg 1$,

$$\frac{\partial E}{\partial z} = \frac{\alpha a E}{b} \operatorname{Re} w(x + ib)$$

$$\approx \frac{\alpha a E}{b} \frac{b/\sqrt{\pi}}{b^2 + x^2} = \frac{\alpha_o a^2 E}{x^2 + b^2} \qquad 6.5.3\text{-}5$$

$$= \frac{\alpha_o E \gamma^2}{\gamma^2(1 + E^2/E_o^2) + (\omega_o - \omega)^2} \quad ,$$

which is just 6.2.1-1 for stationary atoms. Similarly, 6.2.2-7 becomes

$$n(\omega) = 1 - \frac{c}{\omega} \alpha \operatorname{Im} w(x + ib)$$

$$\approx 1 - \frac{c}{\omega} \frac{\alpha_o a x}{b^2 + x^2} \qquad 6.5.3\text{-}6$$

$$= 1 - \frac{c}{\omega} \frac{\alpha_o \gamma(\omega_o - \omega)}{\gamma^2(1 + E^2/E_o^2) + (\omega_o - \omega)^2} \quad ,$$

which is 6.2.1-2 for stationary atoms.

The same results of course are found for other cases of Doppler broadening. For example, with two waves and $|\omega_1 - \omega_2| \ll \gamma_a, \gamma_b$, 6.3.3-1 gives for one of the sine terms:

$$\frac{\omega P}{2\epsilon_o c} = \frac{E_1 \alpha_a}{\pi} \int_{-\infty}^{\infty} \frac{e^{-y^2} dy \sin(\underline{k}_1 \cdot \underline{r} - \omega_1 t + \varphi_1)}{a^2 \left(1 + \dfrac{E_1^2 + E_2^2 + 2E_1 E_2 \cos\Delta_{12}}{E_o^2}\right) + (x + y)^2} \quad . \qquad 6.5.3\text{-}7$$

For

$$a \left(1 + \frac{E_1^2 + E_2^2 + 2E_1E_2 \cos\Delta_{12}}{E_o^2}\right)^{\frac{1}{2}} \gg 1 \quad , \qquad 6.5.3\text{-}8$$

this becomes, as above,

$$\frac{\omega P}{2\epsilon_o c} \approx \frac{\alpha_o E_1 \gamma^2}{\gamma^2 \left(1 + \frac{E_1^2 + E_2^2 + 2E_1E_2 \cos\Delta_{12}}{E_o^2}\right) + (\omega_o - \omega)^2} \quad , \qquad 6.5.3\text{-}9$$

which is the corresponding term of 6.3.2-1 for stationary atoms (homogeneous broadening).

It is rather interesting to note that if $a \ll 1$ for the latter case, and $E_1 \sim E_2$, we will have

$$a \left(1 + \frac{(E_1 + E_2)^2}{E_o^2}\right)^{\frac{1}{2}} \gg 1$$

for $\cos\Delta_{12} \sim +1$, and

$$a \left(1 + \frac{(E_1 - E_2)^2}{E_o^2}\right)^{\frac{1}{2}} \ll 1$$

for $\cos\Delta_{12} \sim -1$. Therefore the total field will in effect be interacting alternately with a homogeneously broadened line and an inhomogeneously broadened line. This type of effect is not covered by the above theory, which assume that a was small enough so that the line always appeared inhomogeneously broadened. For the 3.4 micron line in neon, $a \sim .2$, so that for $E^2/E_o^2 \sim 10$, $b \sim .6$, an intermediate case.

CHAPTER SEVEN

SUMMARY AND DISCUSSION

In this chapter we first give a brief summary in section 7.1. Section 7.2 discusses several applications of the theory. The relationship to other kinds of nonlinear processes which have been recently observed is discussed in section 7.3. Possible experiments to test the theory and use it to gain useful information are discussed in section 7.4, and extensions of the theory are discussed in section 7.5.

7.1 Summary

The results of the preceding calculations are contained primarily in Chapters 5 and 6, where various nonlinear effects are studied and compared to the well-known linear effects of Chapter 4. The nonlinear effects are studied in terms of the corrections they make to the linear gain per unit length and index of refraction, and the gain induced at different polarizations and new frequencies. The nonlinear processes are compared for stationary atoms and for a Maxwellian velocity distribution of excited atoms, with emphasis on the latter, which should apply to practical gaseous laser devices. The two most important characteristics of the Doppler broadened nonlinear effects are: first, the inhomogeneous nature of the broadening, and second, the strong dependence on the relative propagation directions of the interacting waves. The latter characteristic would not be expected of a treatment of Doppler broadening in terms of a distribution of atomic resonance frequencies, and is thus unique for Doppler broadening. One interesting aspect of the inhomogeneous nature of the broadening is that as the interaction width increases with increasing field strength, the interaction effectively becomes more homogeneous.

The lowest order nonlinear effects are studied in considerable detail. Strong nonlinear effects are studied in a more approximate way and related to the perturbation expansion approach to studying nonlinear effects. It is found that the strong nonlinear effects may be thought of as representing saturation of the various processes derived from the perturbation technique. Some of these saturated processes are considered in detail and found to be very similar for stationary atoms, i.e., homogeneous broadening, and for a wide enough velocity distribution so that the broadening is strongly inhomogeneous, as long as the interacting frequencies are within the natural linewidth. The only large difference is in the magnitude of the effects, which are much smaller for the inhomogeneously broadened line, as is the case for linear amplification.

7.2 Applications of the Theory

Aside from their use in calculating the frequency and amplitude dependence of various processes in the nonlinear interaction of waves, the above results could be utilized to study the characteristics of several devices based on laser action. These applications would use both the space dependent and time dependent field amplitudes of Chapter 2. Several of these possible applications will now be listed. First, for the space-dependent amplitudes, some useful applications would be:

(1) The most obvious application would be for studying traveling wave laser amplifiers as a component of a communications system operating at optical frequencies. Such an amplifier would certainly be used as a preamplifier at the receiver, but might well also be used as a power amplifier at the transmitter. The study of nonlinear effects would of course be most useful in the latter case, but some other aspects of the theory would apply to both. For example, we have briefly discussed the effects of "superradiance" in saturating and broadening the small signal gain. Also, the fact that some nonlinear interactions of waves traveling in opposite directions are relatively weak would be of interest in determining the effect of waves reflected back through the amplifier.

(2) The modes of a laser oscillator with mirrors of different reflectivities will not be true cavity modes, but will have traveling wave components. The above theory would be useful in studying the effects of such a situation.

Some cases where the time dependent field amplitudes would be used are:

(3) A study of the effects of nonlinear interactions on the behavior of the cavity modes of a laser oscillator (12) could be carried out using the above theory. Although the theory is not very well suited for such a study, the treatment in terms of traveling waves does give some useful insight into the problem. The cavity modes can be broken down into traveling waves in different directions, and we have

seen that some of the nonlinear processes are strongly dependent on wave directions. Thus only the traveling wave components traveling in the same direction will interact strongly. It is interesting to study the various processes affecting each wave for a given number of modes excited, however, this will not be done here.

Other aspects of laser oscillators are also of interest in terms of the above theory: Laser oscillators are often observed to oscillate in two perpendicularly polarized modes (39,40), and these effects can be studied. Mode locking has been induced in laser oscillators by proper modulation of the cavity medium (41). It is interesting to note that such mode locking will tend to occur naturally, due to the polarizations induced at "combination frequencies" like $2\omega_2 - \omega_1$. For cases where the cavity mode spacing is not much larger than the natural linewidth, this effect should become pronounced.

(4) The effects of nonlinear interactions on the operation of the "ring laser rotation rate sensor" (42) would have to be studied in terms of traveling waves, since this device is characterized by oppositely-directed traveling waves. Again, the fact that some nonlinear interactions for such waves are relatively much weaker is interesting.

7.3 Relationship to Other Nonlinear Effects

With the attainment of very high field strengths from pulsed laser oscillators, many nonlinear effects have been observed (43). Regarding their relationship to the nonlinear effects discussed above, we are primarily interested in effects like Stimulated Raman Scattering (44-46). In this effect, a strong field incident on a Raman active medium results in the production of a field at the first Stokes frequency (47). The incident and Stokes fields then can coherently interact to produce fields at other Stokes and Anti-Stokes frequencies. These effects have been extensively studied theoretically, using perturbation expansion techniques (43,48,49), i.e., in terms of a polarization cubic in the field strengths, and experimentally (43,44,50) using "giant pulse" ruby lasers (51). The experimental results in general agree with theory, with some exceptions (52). One very interesting experimental result which has also been discussed theoretically (53) is the effect of two or more spectral components in the input field. In this case the various Stokes and Anti-Stokes lines are observed to consist of many components or a relatively broad spectrum (44,52).

The similarity between the theory of these effects, as outlined above, and the theory of the lowest order nonlinear effects of Chapter 5 is immediately evident. The only essential difference is that the effects of Chapter 5 are more resonant than the effects of SRS, so that they should be observable at much lower intensities (see section 7.4). The analogy with the effects of multiple spectral components is even more striking.

It is interesting to inquire as to whether some of the effects observed in SRS could be associated with coherent waves in the populations and/or higher order nonlinear effects such as have been discussed in connection with the above theory. It would seem that modulation of the population would be especially likely when multiple spectral components are present in the input wave. Also, the field attained with giant pulse ruby lasers are so intense (51) that higher order effects may become important even for these off-resonance processes.

7.4 Some Possible Experiments

As indicated above, it should be quite feasible to observe and measure not only some lowest order nonlinear effects, but also some of the higher order effects. Among possible interesting observations, we note the following:

(1) Polarization effects. With properly oriented polarizers and adequate detection, it should be possible to observe the perpendicularly polarized field, and perhaps also the polarization dependence of saturation and the waves at "combination" frequencies like $2\omega_2 - \omega_1$.

(2) The waves induced at new frequencies, e.g., $2\omega_2 - \omega_1$, are very interesting. As noted in Chapter 6, it should be possible to observe some higher order waves in addition to the waves due to lowest order nonlinear effects. The frequency and amplitude dependence of these waves could be studied; in addition it might be possible to make measurements on the linewidth of the induced waves and its dependence on amplitude and medium gain.

(3) The above theory could be used to measure some of the parameters characterizing the medium. From small signal (linear) gain

measurements we can determine α and a . From single frequency saturation measurements we can determine E_o^2 . Assuming the Doppler width known, a gives a value for $2\gamma = \gamma_a + \gamma_b$ and E_o^2 gives a value for $\gamma_a\gamma_b/P_o^2$. Thus we can measure the natural linewidth, and by assuming a value for P_o^2 , get approximate values for γ_a and γ_b and thus the lifetimes of the upper and lower levels of the laser transition. It should be noted that for $\gamma_b \gg \gamma_a$, the value of γ_b obtained in this way is essentially independent of the value chosen for P_o^2 , over a wide range. The values of γ_a and γ_b obtained in this way should contain any contributions due to resonance trapping of radiation or pressure broadening, i.e., they are the values arising from the actual operating conditions of the amplifier.

(4) It would be interesting to observe the characteristics of the spontaneous mode locking which should occur in laser oscillators (see section 7.2) and their dependence on the cavity mode spacing relative to the natural linewidth.

(5) It would be very interesting to observe some of the effects which are strongly analogous to the effects observed in stimulated Raman scattering (see section 7.3). Of particular interest in this regard are: index matching or momentum conservation conditions; intensity dependent index of refraction, linewidths and angular widths of new fields, relative to laser linewidths and angular widths; effects of focusing; effects of multiple laser modes. It may also be possible to infer what processes or combination of processes are responsible for the introduction of new fields in SRS.

There are several advantages of gaseous lasers over pulsed ruby lasers for doing work on nonlinear effects, particularly such as those in (5) above. First, since the effects are resonant, they can be observed at lower intensities than would otherwise be required. This in turn allows the experiments to be done on a CW rather than a pulsed basis, so that there is less need for bandwidth in the detection equipment, adjustment of the apparatus and data recording are greatly simplified and beating techniques can easily be applied. Also, the use of gaseous lasers results in a much higher degree of control over the experimental conditions. It is relatively easy to obtain and control a single frequency and spatial mode, and linewidths are very narrow. A very important advantage is that due to the presence of gain there will be no "thresholds" for observing nonlinear effects, and the various responses will be strongly amplified to facilitate observation.

7.5 Extensions

It would be fairly easy to extend the above theory to deal directly with cavity modes. This would constitute an extension of Lamb's work (12) to wider applicability, covering more general situations of polarization, frequency spacing and Doppler broadening.

A more fruitful extension would be to deal with an atomic system where more than two levels are important. This would allow direct treatment of Raman effects, along the lines of the calculations performed by Tang (48). The information to be gained from such calculations would include: the frequency dependence of such processes, the effect of multiple spectral components in the input, and knowledge regarding the importance of population modulation and higher order nonlinear effects.

Another interesting extension would be to study the interaction of traveling waves with a medium which has an applied magnetic field. Some work has been done along these lines (54,55), but a detailed analysis for many situations of practical interest is not available. In particular, the effects of Doppler broadening have not been ascertained.

A study should be made of the changes to the above theory due to the effects of collisions. The latter will affect the homogeneous linewidth and, more importantly, make the line asymmetrical (56). Probably the most important effect of collisions will be in introducing a mechanism connecting various parts of the inhomogeneously broadened line. Such a mechanism would change the characteristics of "holes" burned into the line.

APPENDIX I

THE DOPPLER BROADENING INTEGRAL

The purpose of this appendix is to express the integral 4.3-2,

$$I = \frac{1}{\sqrt{\pi}u} \int_{-\infty}^{\infty} \frac{dV\, e^{-V^2/u^2}}{\gamma + i(\omega_o - \omega + kV)} \quad , \tag{I-1}$$

in a convenient form for referral to tabulated functions and for approximation in the limiting case of interest. We first express I-1 in terms of the (tabulated) Error Function for Complex Arguments (25),

$$w(z) = e^{-z^2} \operatorname{erfc}(-iz) = \frac{2}{\sqrt{\pi}} e^{-z^2} \int_z^{\infty} e^{-t^2} dt \quad , \tag{I-2}$$

and then evaluate I-1 to first order in a for $a = \gamma/ka \ll 1$, the limit of strong Doppler broadening.

Defining

$$a = \frac{\gamma}{ku} \, , \quad x = \frac{\omega_o - \omega}{ku} \quad , \tag{I-3}$$

I-1 becomes

$$I = \frac{1}{\sqrt{\pi}ku^2} \int_{-\infty}^{\infty} \frac{dV\, e^{-V^2/u^2}}{a + i(x + V/u)} \quad , \tag{I-4}$$

and with $y = V/u$ this becomes

$$I = \frac{1}{\sqrt{\pi}ku} \int_{-\infty}^{\infty} \frac{dy\, e^{-y^2}}{a + i(x + y)} \quad . \tag{I-5}$$

Using the fact that

$$\int_0^\infty dt' \, e^{-t'[a + i(x + y)]} = \frac{1}{a + i(x + y)} \quad , \qquad \text{I-6}$$

we can write I-5 as

$$I = \frac{1}{\sqrt{\pi}ku} \int_{-\infty}^{\infty} dy \, e^{-y^2} \int_0^\infty dt' \, e^{-t'[a + i(x + y)]} \quad . \qquad \text{I-7}$$

Exchanging the order of integration and performing the y-integration gives

$$I = \frac{1}{ku} \int_0^\infty dt' \, e^{-t'^2/4 - (a + ix)t'} \quad . \qquad \text{I-8}$$

With a change of variable to $t = t'/2 + (a + ix)$, I-8 becomes

$$I = \frac{2}{ku} \, e^{(a + ix)^2} \int_{a+ix}^{\infty+ix} dt \, e^{-t^2} \quad , \qquad \text{I-9}$$

which is 4.3-4. In terms of the definition I-2, we have 4.3-5:

$$I = \frac{\sqrt{\pi}}{ku} \, w(- x + ia) = \frac{\sqrt{\pi}}{ku} \, w^*(x + ia) \quad , \qquad \text{I-10}$$

which is tabulated in reference 25 for the range of values

$$x = 0(.1)\,3.9$$

$$a = 0(.1)\,3 \quad .$$

For evaluation of I-9 in the interesting limit $a \ll 1$, we consider the integral

$$I' = \int_{a+ix}^{\infty+ix} dt \, e^{-t^2} \quad . \qquad \text{I-11}$$

Figure 18 shows an integration contour C which includes I' . The theory of complex integration gives

$$\oint_C dt\ e^{-t^2} = 0 = I' + I_1 + I_2 + I_3 + I_4 \quad , \qquad \text{I-12}$$

where the subscripts refer to the portions of C indicated in Figure 18. We see immediately that $I_1 = 0$ and that

$$I_2 = -\int_0^\infty e^{-t^2}\ dt = -\frac{\sqrt{\pi}}{2} \quad . \qquad \text{I-13}$$

Also, we have

$$I_3 = \int_0^{ix} dt\ e^{-t^2} = i \int_0^x dt'\ e^{t'^2} \quad . \qquad \text{I-14}$$

The remaining portion of the contour is

$$I_4 = \int_{ix}^{a+ix} dt\ e^{-t^2} = e^{x^2} \int_0^a dt'\ e^{-(t'^2 + 2ixt')} \quad , \qquad \text{I-15}$$

where $t' = t - ix$. To first order in a we have

$$I_4 \cong ae^{x^2} \quad , \qquad \text{I-16}$$

and

$$e^{(a + ix)} \simeq e^{-x^2} (1 + 2iax) \quad . \qquad \text{I-17}$$

Using I-12 and the above results, we find to first order in a ,

$$\text{Re } I = \frac{\sqrt{\pi}}{ku} \left[e^{-x^2} - \frac{2a}{\sqrt{\pi}} (1 - 2xF(x)) \right] \qquad \text{I-18}$$

and

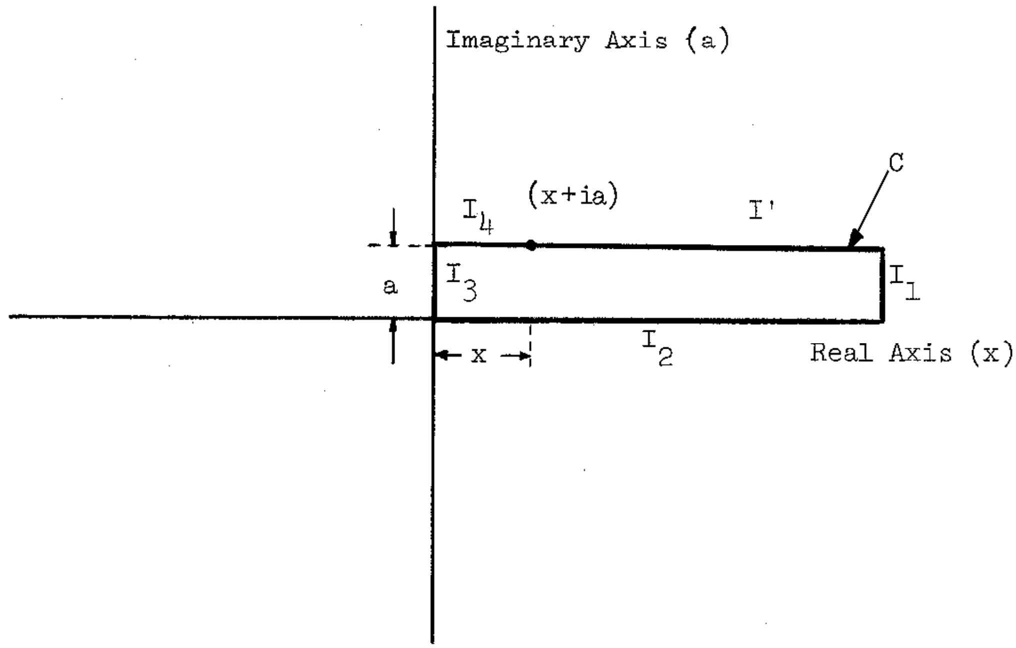


FIGURE 18

The Contour of Integration for Calculating $w(x + ia)$ for $a \ll 1$.

$$\mathrm{Im}\ I = -\frac{\sqrt{\pi}}{ku}\left[\frac{2}{\sqrt{\pi}} F(x) - 2ax\ e^{-x^2}\right] , \qquad \text{I-19}$$

where

$$F(x) = e^{-x^2} \int_0^x e^{t^2}\, dt \quad . \qquad \text{I-20}$$

Reference 25 tabulates $F(x)$ for

$$x = 0(.02)\ 2 \quad ,$$

and $x\ F(x)$ for

$$x^{-2} = .25\ (-\ .005)\ 0 \quad .$$

APPENDIX II

EVALUATION OF THIRD ORDER DOPPLER BROADENING INTEGRALS

The purpose of this appendix is to evaluate the integral $I_{a,b\mp}$ of section 5.3 in terms of the Doppler broadening integral which was studied in Appendix I. This is done below for two cases. In addition, we indicate the techniques to be used for studying limiting cases.

As the basic expression we wish to evaluate, we take

$$I_{a\mp} = \frac{1}{\sqrt{\pi}u}\int_{-\infty}^{\infty} \frac{dV\, e^{-V^2/u^2}}{[\gamma + i(\omega_o - \nu + \underline{K}\cdot\underline{V})][\gamma_a - i(\omega' - \omega'' - (\underline{k}' - \underline{k}'')\cdot\underline{V})]}$$

$$\times \frac{1}{\gamma \mp i(\omega_o - \omega' + \underline{k}'\cdot\underline{V})} \quad , \qquad \text{II-1}$$

where for I_{a-} we must replace ω' and $\underline{k}'$ in the last factor by ω'' and $\underline{k}''$. The notation is that of section 5.3. To the sum of I_{a-} and I_{a+} as given by II-1, we must add the same expressions with γ_a replaced by γ_b , in order to get $I(\omega\omega'\omega'')$ of 5.3-5.

The important parameter in this calculation is u/c , and we typically have

$$u/c \lesssim 10^{-6} \quad . \qquad \text{II-2}$$

If we let each $\underline{k}$ have the magnitude

$$k_o = \omega_o/c \quad ,$$

we obtain the interaction frequencies correct to order u/c , which is completely adequate for these problems.

II.1 $\underline{k}' \sim \underline{k}''$

With $\underline{k}' \sim \underline{k}''$, we have

$$\underline{K} = \underline{k} + \underline{k}' - \underline{k}'' \simeq \underline{k}$$

and

$$|\underline{k}' - \underline{k}''| \simeq \frac{\Delta\omega}{c} \quad ,$$

where $\Delta\omega = \omega' - \omega''$. This gives

$$\gamma_a - i(\omega' - \omega'' - (\underline{k}' - \underline{k}'') \cdot \underline{V}) \simeq \gamma_a - i\Delta\omega(1 - \frac{V}{c}) \qquad \text{II.1-2}$$

Since for all appreciable contribution to the integral we have

$$\frac{V}{c} \lesssim \frac{u}{c} \lesssim 10^{-6} \quad ,$$

we can neglect the contribution of this term to the broadening and replace II.1-2 by

$$\gamma_a - i\Delta\omega \quad .$$

For this case we then have

$$I_{a\mp} = \frac{1}{(\gamma_a - i\Delta\omega)\sqrt{\pi}u} \int_{-\infty}^{\infty} \frac{dV\, e^{-V^2/u^2}}{[\gamma + i(\omega_o - \nu + k_o V)]}$$

$$\times \frac{1}{\gamma \mp i(\omega_o - \omega' + k_o V \cos(k,k'))} \quad , \qquad \text{II.1-3}$$

where, in accordance with the above discussion, we have replaced the $|\underline{k}|$'s by k_o. The $\cos(k,k')$ is $+1$ or -1, depending on whether $\underline{k} \sim \underline{k}'$ or $\underline{k} \sim -\underline{k}'$.

If we define

$$a = \gamma/k_o u$$

$$X = (\omega_o - \nu)/k_o u$$

$$x' = (\omega_o - \omega')/k_o u \qquad \text{II.1-4}$$

$$x'' = (\omega_o - \omega'')/k_o u \quad ,$$

II.1-3 can be written

$$I_{a\mp} = \frac{1}{(\gamma_a - i\Delta\omega)\sqrt{\pi}\, k_o^2 u^2} \int_{-\infty}^{\infty} \frac{dy\, e^{-y^2}}{[a + i(X + y)][a \mp i(x' + y\cos(kk'))]} \quad ,$$

II.1-5

where $y = V/u$. The denominator of the integral can be expanded in a partial fraction expansion, giving

$$\frac{1}{[a + i(X + y)][a \mp i(x' + y\cos(kk'))]}$$

II.1-6

$$= \frac{C_{1\mp}}{a + i(X + y)} + \frac{C_{2\mp}}{a \mp i(x' + y\cos(kk'))} \quad ,$$

where

$$C_{1\mp} = \frac{1}{(a \pm \cos(kk')a) \mp i(x' - \cos(kk')X)} \quad , \qquad \text{II.1-7}$$

$$C_{2\mp} = \frac{\pm\cos(kk')}{(a \pm \cos(kk')a) \mp i(x' - \cos(k,k')X)} \quad . \qquad \text{II.1-8}$$

Using II.1-6 in II.1-5, we then find, using the results of Appendix I,

$$I_{a-} = \frac{\sqrt{\pi}}{(\gamma_a - i\Delta\omega)\, k_o^2 u^2} \left[C_{1-}\ w^*(X + ia) + C_{2-}\ w(x'' + ia) \right] , \qquad \text{II.1-9}$$

$$I_{a+} = \frac{\sqrt{\pi}}{(\gamma_a - i\Delta\omega)\, k_o^2 u^2} \left[C_{1+}\ w^*(X + ia) + C_{2+}\ w^*(x' + ia) \right] . \qquad \text{II.1-10}$$

II.1-9 and 10 of course include the special case $\omega' = \omega''$. If the limit discussed in section II.3 is taken, they also include the case $\omega = \omega' = \omega''$.

II.2 $\underline{k}' \sim - \underline{k}''$

For this case the γ_a, γ_b terms contribute to the Doppler broadening as much as the γ terms. We have to evaluate

$$I_{\mp} = \frac{1}{\sqrt{\pi}u} \int_{-\infty}^{\infty} \frac{dV\ e^{-V^2/u^2}}{[\gamma + i(\omega_o - \nu + KV][\gamma_a - i(\Delta\omega - 2k_o V)]} \qquad \text{II.2-1}$$

$$\frac{1}{\gamma \mp i(\omega_o - {\omega'' \atop \omega'} \mp k_o V)} ,$$

where

$$K = (2 + \cos(kk'))\, k_o \ . \qquad \text{II.2-2}$$

With the definitions

$$\begin{aligned} A &= \gamma_a / 2k_o u \\ a' &= \gamma / Ku \\ X' &= (\omega_o - \nu)/Ku \\ x &= \Delta\omega / 2k_o u \end{aligned} \qquad \text{II.2-3}$$

and II.1-4, II.2-1 becomes

$$I_{\mp} = \frac{1}{2\sqrt{\pi}\, Kk_o^2 u^3} \int_{-\infty}^{\infty} \frac{dy\, e^{-y^2}}{[a' + i(X' + y)][A - i(x - y)][a \mp i(x' \mp y)]} \quad , \qquad \text{II.2-4}$$

where for the upper sign, x' should be replaced by x'' . Although the results are more cumbersome, the denominator of the integrand can again be expanded with a partial fraction expansion of the form

$$\frac{K_{1\mp}}{a' + i(X' + y)} + \frac{K_{2\mp}}{a \mp i(x' \mp y)} + \frac{K_{3\mp}}{A - i(x - y)} \quad , \qquad \text{II.2-5}$$

where

$$\Delta_{\mp} K_{1\mp} = (A - a) - i(x \mp x') \quad , \qquad \text{II.2-6}$$

$$\Delta_{\mp} K_{2\mp} = (a' - A) + i(X' + x) \quad , \qquad \text{II.2-7}$$

$$\Delta_{\mp} K_{3\mp} = (a - a') - i(X' \pm x') \quad , \qquad \text{II.2-8}$$

and

$$\begin{aligned} \Delta_{\mp} = {} & [a' + A + i(X' - x)][A - a' - i(X' + x)](a \mp ix') \\ & + [a' + a + i(X' \mp x')][a' - a + i(X' \pm x')](A - ix) \\ & + [a + A - i(x \pm x')][a - A + i(x \mp x')](a' + iX') \quad . \end{aligned} \qquad \text{II.2-9}$$

As before, x' should be replaced by x'' for the upper sign in these expressions.

Using II.2-5 and the results of Appendix I, II.2-4 becomes

$$I_{a-} = \frac{\sqrt{\pi}/2}{Kk_o^2u^3}\left[K_{1-}\ w^*(X' + ia') + K_{2-}\ w(x'' + ia) + K_{3-}\ w(x + iA)\right] ,$$

II.2-10

$$I_{a+} = \frac{\sqrt{\pi}/2}{Kk_o^2u^3}\left[K_{1+}\ w^*(X' + ia') + K_{2+}\ w^*(x' + ia) + K_{3+}\ w(x + iA)\right] .$$

II.2-11

$I_{b\mp}$ are obtained by replacing A by

$$B = \gamma_b/2k_o u$$ II.2-12

in all the above equations.

II.3 Limiting Cases

The above analysis breaks down when two of the factors in the denominator of the integrand become equal. For example, this happens in II.1 when $\cos(k,k') = 1$ and $X = x'$ for I_+ ; and also when $\cos(k,k'') = -1$ and $X = -x''$ for I_- . These cases could be treated separately, but it will be more useful here to treat them as limiting cases of the above results. It of course always happens that the singularity in the coefficients is balanced by cancellation of terms. Then what is important is the limit as the special case is approached. For the cases mentioned above, this takes the form

$$\lim_{\delta x \to o}\left\{\frac{w^*(x + \delta x + ia) - w^*(x + ia)}{\delta x}\right\} = \left.\frac{\partial w^*}{\partial x}\right|_{x + ia} ,$$

II.3-1

by the definition of the derivative. For the cases arising from II.2, we will find the same behavior with two of the coefficients, and the third coefficient will be of order δx, so that we again have a definite limit existing.

We can calculate the derivative as follows:

We have

$$\frac{\sqrt{\pi}}{2} w^*(x + ia) = e^{(a + ix)^2} \int_{a+ix}^{\infty+ix} dt\, e^{-t^2} \qquad \text{II.3-2}$$

Taking the partial derivative with respect to x gives

$$\frac{\sqrt{\pi}}{2} \frac{\partial w^*(x + ia)}{\partial x} = 2i(a + ix)\, e^{(a + ix)^2} \int_{a+ix}^{\infty+ix} dt\, e^{-t^2}$$

$$+ e^{(a + ix)^2} \left[i\, e^{-t^2}\Big|_{\infty+ix} - i\, e^{-t^2}\Big|_{a+ix} \right] \qquad \text{II.3-3}$$

$$= \sqrt{\pi}\, i(a + ix)\, w^*(x + ia) - i \quad .$$

Since

$$\frac{\partial w^*(x + ia)}{\partial a} = - i \frac{\partial w^*(x + ia)}{\partial x} \quad ,$$

we also have

$$\frac{\sqrt{\pi}}{2} \frac{\partial w^*(x + ia)}{\partial a} = \sqrt{\pi}\, (a + ix)\, w^*(x + ia) - 1 \qquad \text{II.3-4}$$

Similarly, for use when $a \ll 1$, we find

$$\frac{\partial F(x)}{\partial x} = 1 - 2xF(x) \quad , \qquad \text{II.3-5}$$

where

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

For the two limiting cases of II.1, we find

$$I_{a+}\ (X = x',\ \cos(k,k') = 1) = I_{a-}\ (X = -x'',\ \cos(k,k'') = -1)$$

$$= \frac{\sqrt{\pi}}{(\gamma_a - i\Delta\omega)(k_o u)^2}\left[1 - 2(a + iX)\ w^*(X + ia)\right] \quad . \qquad \text{II.3-6}$$

For II.2, we see that we obtain equal factors when $\cos(k,k') = -1$ so that $K = k_o$ and $a' = a$; with $X' = x'$ for I_- , and with $X' = -x''$ for I_+ . Evaluation of these cases is rather tedious, with the result

$$I_{a+}\ (\cos(k,k') = -1\ ,\ X' = x') = I_{a-}\ (\cos(k,k') = -1\ ,\ X' = -x'')$$

$$= \frac{\sqrt{\pi}}{2(k_o u)^3}\left\{\frac{-2/\sqrt{\pi} + 2(a + iX')\ w^*(X' + ia)}{B + i(X' + x)} + \frac{w(x + iA) - w^*(X' + ia)}{\left[B + i(X' + x)\right]^2}\right\} \quad . \qquad \text{II.3-7}$$

For $\cos(k,k') = 1$ in II.2, we cannot obtain equal factors because $a' = a/3$. However, for $a \ll 1$ and $X' = x'$ for I_- ; $X' = -x''$ for I_+ , we have approximately equal terms, and obtain, to within terms of relative order a ,

$$I_{a-}\ (\cos(k,k') = 1\ ,\ X' = x'') = I_{a+}\ (\cos(k,k') = 1\ ,\ X' = -x') =$$

$$= \frac{\sqrt{\pi}/2}{3(k_o u)^3}\left\{\frac{2/\sqrt{\pi} - 2(a + iX')\ w^*(X' + ia)}{A - a - i(X' + x)} + \frac{w(x + iA) - w^*(X' + ia)}{[A - a - i(X' + x)]^2}\right\} \quad . \qquad \text{II.3-8}$$

It should be noted that $X' = (\omega_o - \nu)/3k_o u$ here, compared to $X' = (\omega_o - \nu)/k_o u$ above.

APPENDIX III

THE FOURIER COEFFICIENTS

The purpose of this appendix is to derive expressions for the Fourier coefficients used in section 6.3. We need the Fourier series expansion in the form

$$f(\cos u) = \sum_{m=0}^{\infty} c_m \cos mu \quad , \qquad \text{III-1}$$

where

$$C_m = \frac{(2 - \delta_{om})}{\pi} \int_0^{\pi} f(\cos u) \cos mudu \quad . \qquad \text{III-2}$$

In section III.1 we consider the homogeneous coefficients, C_m , and in section III.2 the inhomogeneous coefficients, C'_m .

III.1 The Homogeneous Coefficients

For 6.3.2 we require the expansion

$$\frac{1}{1 + \dfrac{E_1^2 + E_2^2 + 2E_1E_2 \cos u}{E_o^2} + y^2} = \sum_{m=o}^{\infty} C_m \cos mu \ . \qquad \text{III.1-1}$$

From III-1 and 2, we have

$$C_m = \frac{(2 - \delta_{om})\, I_m}{\pi(1 + y^2 + (E_1^2 + E_2^2)/E_o^2)} \quad , \qquad \text{III.1-2}$$

where

$$I_m = \int_0^{\pi} \frac{\cos mu \, du}{1 + a_o \cos u} \quad , \qquad \text{III.1-3}$$

and

$$a_o = \frac{2E_1E_2/E_o^2}{1 + y^2 + (E_1^2 + E_2^2)/E_o^2} \quad . \qquad \text{III.1-4}$$

Although III.1-3 is a tabulated integral (57), we shall evaluate it here because the same technique will be used for evaluating the inhomogeneous coefficients in III.2. With

$$v = e^{iu} \quad , \qquad \text{III.1-5}$$

we have

$$I_m = \frac{1}{2} \int_{-\pi}^{\pi} \frac{\cos mu \, du}{1 + a_o \cos u}$$

$$= \text{Re} \, \frac{1}{2i} \oint \frac{v^{m-1} \, dv}{1 + \frac{a_o}{2}(v + 1/v)} \qquad \text{III.1-6}$$

$$= \text{Re} \, \frac{1}{ia_o} \oint \frac{v^m \, dv}{v^2 + 2v/a_o + 1}$$

$$= \text{Re} \, \frac{1}{ia_o} \oint \frac{v^m \, dv}{(v - v_1)(v - v_2)} \quad ,$$

where

$$v_2 = -\frac{(1 + \sqrt{1 - a_o^2})}{a_o}$$

$$v_1 = -\frac{(1 - \sqrt{1 - a_o^2})}{a_o} \qquad \text{III.1-7}$$

and the integral is around the unit circle in the complex v-plane, as indicated in Figure 19a. We note that the integrand has two first order poles, at v_2 outside the unit circle, and v_1, inside the unit circle. Therefore,

$$I_m = \text{Re } 2\pi i \text{ (residue at } v_1)$$

$$= \frac{\pi\left[\sqrt{1 - a_o^2} - 1\right]^m}{a_o^m \sqrt{1 - a_o^2}} \quad . \qquad \text{III.1-8}$$

In the above we have implicitly used the fact that $a_o < 1$. Therefore we have

$$C_m = \frac{(2 - \delta_{om})}{(1 + y^2 + (E_1^2 + E_2^2)/E_o^2)} \frac{\left[\sqrt{1 - a_o^2} - 1\right]^m}{a_o^m \sqrt{1 - a_o^2}} \quad , \qquad \text{III.1-9}$$

as used in 6.3.2.

III.2 The Inhomogeneous Coefficients

For 6.3.3, we need the expansion

$$\frac{1}{\left(1 + \frac{E_1^2 + E_2^2 + 2E_1E_2 \cos u}{E_o^2}\right)^{\frac{1}{2}}} = \sum_{m=o}^{\infty} C'_m \cos mu \quad . \qquad \text{III.2-1}$$

We have

$$C'_m = \frac{(2 - \delta_{om}) I'_m}{\pi(1 + (E_1^2 + E_2^2)/E_o^2)^{\frac{1}{2}}} \quad , \qquad \text{III.2-2}$$

where

$$I'_m = \int_0^{\pi} \frac{\cos mu \, du}{(1 + a'_o \cos u)^{\frac{1}{2}}} , \qquad \text{III.2-3}$$

and

$$a'_o = \frac{2E_1E_2/E_o^2}{1 + (E_1^2 + E_2^2)/E_o^2} . \qquad \text{III.2-4}$$

Introducing III.1-5, III.2-3 becomes

$$I'_m = \text{Re } I$$

$$= \text{Re} \sqrt{\frac{2}{a'_o}} \frac{1}{i} \int_{c_1} \frac{v^{m-\frac{1}{2}} \, dv}{(v - v_1)^{\frac{1}{2}} (v - v_2)^{\frac{1}{2}}} , \qquad \text{III.2-5}$$

where the contour c_1 is the upper half unit circle in the complex v-plane. Note that we now have a cut in the v-plane, which we take joining v_1 and v_2 along the real axis. To evaluate I, we form the closed contour c, shown in Figure 19b. Since c contains no singularities, the integral around c must equal zero. Splitting the contour up into workable intervals, we have

$$\oint_c = 0 = \int_{c_1} + \int_{-1}^{v_1-r} + \int_{c_2} + \int_{v_1+r}^{0} + \int_0^1 , \qquad \text{III.2-6}$$

where c_2 is the small semicircle of radius r around v_1. For the various contributions we have

$$\int_{-1}^{v_1-r} = \frac{(-1)^{m+1}}{i} \sqrt{\frac{2}{a'_o}} \int_{-1}^{v_1-r} \frac{(-v)^{m-\frac{1}{2}} \, dv}{(v_1 - v)^{\frac{1}{2}} (v - v_2)^{\frac{1}{2}}} ,$$

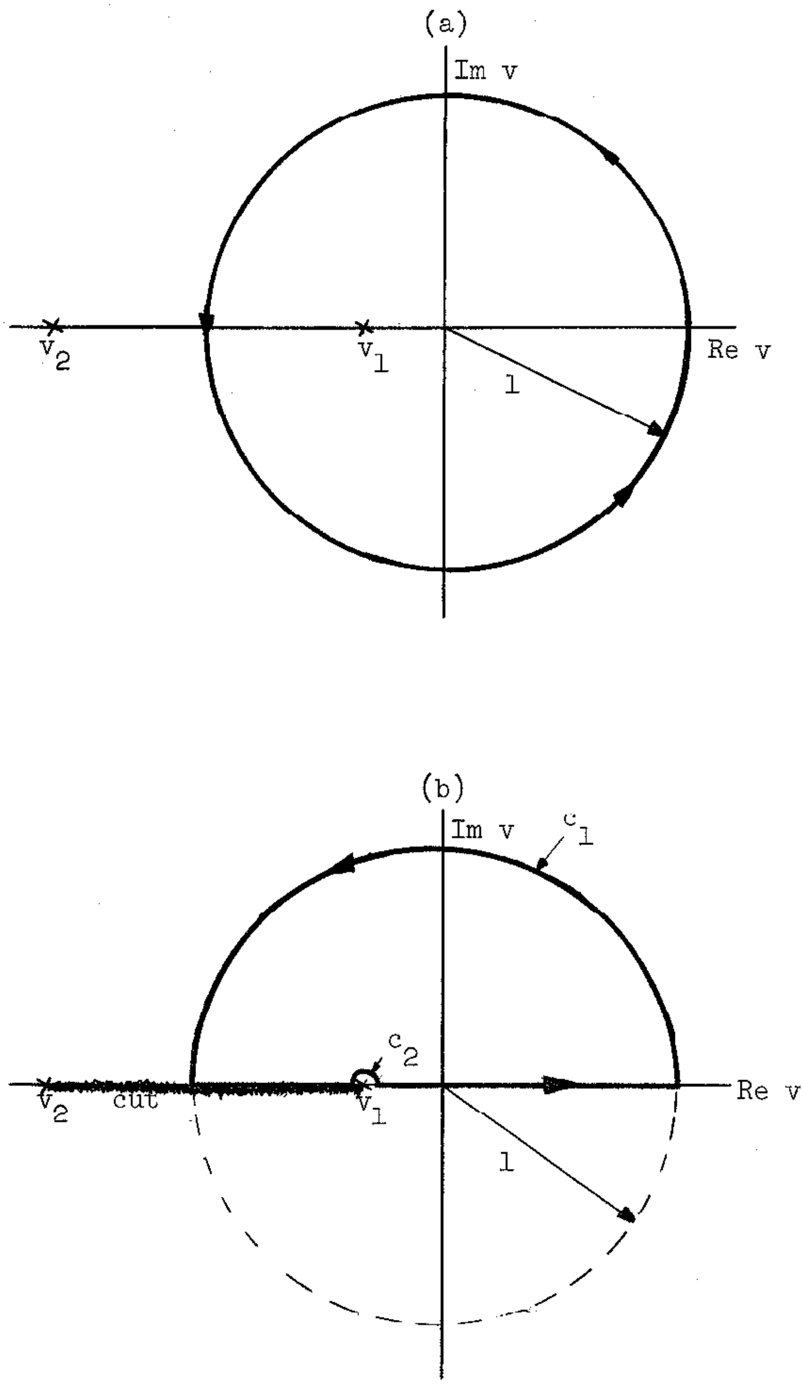


FIGURE 19

Integration Contours for Calculating Fourier Coefficients
(a) Homogeneous (b) Inhomogeneous

which is pure imaginary, since the integrand is real.

$$\int_{c_2} \sim \int \frac{d\theta \; ir \; e^{i\theta} \; v_1^{m-\frac{1}{2}}}{(v_1 - v_2)^{\frac{1}{2}} \; r^{\frac{1}{2}} \; e^{i\theta/2}} \rightarrow 0$$

as $r \rightarrow 0$.

$$\int_{v_1+r}^{0} = (-1)^{m+1} \sqrt{\frac{2}{a_0'}} \int_{v_1+r}^{0} \frac{(-v)^{m-\frac{1}{2}} \, dv}{(v - v_1)^{\frac{1}{2}} \, (v - v_2)^{\frac{1}{2}}} = \text{pure real} \; .$$

$$\int_{0}^{1} = \frac{1}{i} \sqrt{\frac{2}{a_0'}} \int_{0}^{1} \frac{v^{m-\frac{1}{2}} \, dv}{(v - v_1)^{\frac{1}{2}} \, (v - v_2)^{\frac{1}{2}}} = \text{pure imaginary} \; .$$

Thus

$$I_m' = \text{Re} \int_{c_1} = - \int_{v_1+r} = (-1)^m \; \frac{2}{a_0'} \int_{v_1+r}^{0} \frac{(-v)^{m-\frac{1}{2}} \, dv}{(v - v_1)^{\frac{1}{2}} \, (v - v_2)^{\frac{1}{2}}} \; . \qquad \text{III.2-7}$$

Letting

$$t = (v - v_1)^{\frac{1}{2}} \; ,$$

III.2-7 becomes

$$I_m' = \sqrt{\frac{8}{a_0'}} \; (-1)^m \int_{0}^{d} \frac{(d^2 - t^2)^{m-\frac{1}{2}}}{(c^2 + t^2)^{\frac{1}{2}}} \, dt \quad , \qquad \text{III.2-8}$$

where

$$d^2 = - v_1 = \frac{1 - \sqrt{1 - a_o'^2}}{a_o'} \quad ,$$

$$c^2 = v_1 - v_2 = \frac{2\sqrt{1 - a_o'^2}}{a_o'} \quad . \qquad \text{III.2-9}$$

Substituting III.2-8 into III.2-2 gives

$$C_m' = \frac{(2 - \delta_{om})(-1)^m \sqrt{8/a_o'}}{\pi(1 + (E_1^2 + E_2^2)/E_o^2)^{\frac{1}{2}}} \int_0^d \frac{(d^2 - t^2)^{m-\frac{1}{2}}}{(c^2 + t^2)^{\frac{1}{2}}} \, dt \quad . \qquad \text{III.2-10}$$

For the small field limit, $a_o' \ll 1$, we find

$$b^2 \simeq a_o'/2 \quad ,$$

$$c^2 \simeq 2/a_o' \qquad \text{III.2-11}$$

Neglecting t^2 compared to c^2 , I_m' becomes

$$I_m' \simeq 2(-1)^m \left(\frac{a_o'}{2}\right)^m \int_0^1 (1 - z^2)^{m-\frac{1}{2}} \, dz \quad , \qquad \text{III.2-12}$$

where

$$t = \sqrt{\frac{a_o'}{2}} \, z \quad .$$

Thus (58)

$$I_o' \simeq \pi \quad ,$$

$$I_1' \simeq - \frac{a_o'\pi}{4} \quad ,$$

and

$$C_o' \simeq \frac{1}{\left(1 + \frac{E_1^2 + E_2^2}{E_o^2}\right)^{\frac{1}{2}}} \simeq 1 - \frac{E_1^2 + E_2^2}{2E_o^2} \quad , \qquad \text{III.2-13}$$

$$C_1' \simeq \frac{-a_o'}{2\left(1 + \frac{E_1^2 + E_2^2}{E_o^2}\right)^{\frac{1}{2}}} \simeq - \frac{E_1 E_2}{E_o^2} \quad . \qquad \text{III.2-14}$$

An alternative approach leads to an expression for C_m' as an infinite series. Expanding

$$(1 + a_o' \cos u)^{-\frac{1}{2}}$$

by the binomial theorem, we find the coefficient of $(\cos u)^m$ to be

$$\frac{(-1)^m \, (2m - 1)!! \, a_o'^m}{2^m \, m!} \qquad \text{III.2-15}$$

This expansion converges for $a_o' < 1$, which is always true for our case. For $m = 0$ in III.2-15 and elsewhere in this derivation, the double factorial is defined to be equal to unity. From Dwight (59), the coefficient of $\cos(m - 2p)u$ in the expansion of $\cos^m u$ is

$$\frac{1}{2^{m-1}} C_p^m \quad ,$$

unless m is even and $p = m/2$, when it is

$$\frac{1}{2^m} C_{\frac{m}{2}}^m \quad ,$$

where the C_p^m are the binomial coefficients

$$C_p^m = \frac{m!}{p!(m-p)!} \quad .$$

Thus the coefficient of $\cos mu$ in the expansion

$$(1 + a_o' \cos u)^{-\frac{1}{2}} = \sum_{m=o}^{\infty} b_m \cos mu \qquad \text{III.2-16}$$

is

$$b_m = (2 - \delta_{om}) \sum_{q-2p=m} -\left(\frac{a_o'}{4}\right)^q \frac{(2q-1)!!}{p!\,(q-p)!} \quad , \qquad \text{III.2-17}$$

where q, p, m are positive or zero. From III.2-1, 16 and 17 we thus have

$$C_m' = \frac{(2-\delta_{om})(-a_o'/4)^m}{\left(1 + \frac{E_1^2 + E_2^2}{E_o^2}\right)^{\frac{1}{2}}} \sum_{p=o}^{\infty} \left(\frac{a_o'^2}{16}\right)^p \frac{(4p + 2m - 1)!!}{p!\,(m+p)!} \quad , \qquad \text{III.2-18}$$

where we define

$$(4p + 2m - 1)!! = 1$$

for $m = p = 0$. The series converges absolutely if $a_o' < 1$. The convergence is very slow for $a_o' \sim 1$, however it is still useful and was used to calculate the C_m' for Figure 17.

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