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Saturation Behavior of a Doppler Broadened Transition Involving Levels With Closely Spaced Structure*

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ABSTRACT

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The non-linear absorption or gain characteristics of an optical frequency, Doppler broadened atomic resonance involving levels with closely spaced structure is analyzed. The level structures are assumed to be resolved with respect to their natural widths, but not necessarily with respect to the Doppler width at the optical transition. The radiation field consists of two closely spaced monochromatic frequencies lying within the Doppler width of the resonance. It is shown that, due to saturation of level populations and double quantum Raman transitions between levels, appreciable non-linear coupling takes place between the two fields. This coupling shows a resonance behavior when the frequency separation of the two applied fields becomes equal to the frequency splitting between two of the components which form either level structure. The width of this resonance is determined entirely by the natural widths of these two level components and not by the Doppler width of the optical transition or the natural width of other level components. When such a resonance occurs the overall gain or attenuation characteristics of the atomic resonance change drastically. In practice the frequency spacing of the fields may be kept constant while the level structures are tuned, e. g. magnetically.

The effects are analyzed for cases of running wave and standing wave radiation fields. The use of this effect in precise determination of level structures as well as mode or transition coupling of a gas laser is discussed. Portions of the analysis are applicable to resonances with more general form of inhomogeneous broadening.

Author

1. INTRODUCTION

This paper is a detailed theoretical analysis of the non-linear attenuation or gain characteristics of a Doppler broadened atomic (or molecular) resonance involving levels with small splittings. An effect is discussed which manifests itself in a particularly useful way for high resolution studies of the structure of an atomic resonance using a gas laser. Also, the non-linear gain characteristics considered here are applicable to studies of frequency behavior and mode or transition coupling of a gas laser operating on levels with closely spaced structure. This kind of structure, commonly encountered in practice, may arise, for instance, from fine or hyperfine interactions or Zeeman or Stark splitting. The structure of a line may be considerably smaller than its Doppler broadening.

The attenuation or gain characteristics are considered here for a case in which an applied radiation contains two monochromatic optical or infrared frequencies separated by an amount larger than the natural linewidths of the atomic resonance (but not necessarily larger than the corresponding Doppler width). A radiation field of the type discussed can be obtained, for example, from an optical or infrared gas laser oscillating simultaneously in two modes of its Fabry-Perot resonator.

The ordinary linear attenuation or gain factor of the system described above is straightforward.[1] Its details are determined by the Doppler width of the atomic resonance and the actual size of the structure of the line. When the applied field strengths are sufficiently large, non-linear effects become observable. The details of non-linear effects, however, depend primarily on the natural linewidths of the system and not as much on the Doppler width. These non-linear effects arise from Raman type transitions between components of the structure of either energy

level and from the effect of saturation of the populations of the levels. In particular, their effects lead to a coupling between the two applied fields which has a resonance behavior when the frequency separation of the two applied fields becomes equal to the frequency splitting between two of the components which form either level structure. The width of this resonance is determined entirely by the natural widths of the levels and not by the Doppler width. When such a resonance occurs, the overall gain or attenuation characteristics of the atomic resonance change drastically.

In an experimental arrangement, instead of changing the frequency separation of the two applied fields, one may choose a system in which the level splittings are tunable (for example, by a magnetic field). In this case the frequency spacing of the applied fields may be considered constant, and resonances may be observed in the total attenuation or gain as a function of tuning. These resonances, together with the known value of the frequency difference of the two applied fields can be used to obtain high resolution details of the level structures. Where appropriate, these resonances will be referred to as difference frequency crossing.

Under certain general conditions to be described later, the problem for a complex level structure can be reduced to consideration of simply a pair of closely spaced upper levels and a single lower level—leading to two transitions involving one common level. The Doppler width of the two transitions may be allowed to overlap. (The presence of a common level for the two transitions is essential).

The effect discussed here was initially described in an earlier publication involving the present authors and a preliminary experimental observation was given.[2] In that discussion only gross features of the effect were described. In a

later paper, we will present spectroscopic applications of the present detailed theory.

In section 2, the theory of the effect mentioned above is given in its simplest form. The applied radiation is taken as two travelling waves obtained from the output of a gas laser oscillating simultaneously on two of its resonator modes. The atomic system is assumed to be exposed to this radiation field outside of the laser cavity. (In practice, this system may be, for instance, a gaseous discharge tube containing the same atomic species at that forming the active element of the laser oscillator). The level structure is assumed to be of the simplest type; namely, the transitions are assumed to occur between two upper levels and one lower level. Various expressions appearing in different stages of these calculations are unavoidably lengthy. In order to avoid having computational complexities obscure the nature of our discussion, we have, insofar as possible, given only representative manipulations and results in the text, and details in appendices. In spite of all the details, the final result, including averaging over the velocity distribution takes a simple form. An interesting aspect of the result of this calculation is the fact that the width of a difference frequency crossing resonance involves only the natural width of the two upper levels and is entirely independent of the natural width of the lower level. Accordingly, the width of an observed resonance will be narrow for cases where the two upper levels are long lived. This holds regardless of the lifetime of the lower level. It may be an extremely short lived level.

In section 3, we will discuss the applicability of the three level model, used throughout, toward description of transitions between levels of complicated structure. It will be shown that, as long as the structure of the levels are resolved to within their natural widths, (but not necessarily to within their Doppler widths)

the problem involving complex level structure may be reduced, in many instances, to considerations of three level systems.

Section 4 gives detailed theory of the non-linear characteristic for a case where the atomic system is placed within the laser resonator. In this case, the radiation fields will be in the form of two standing waves. In this case, additional resonant terms appear which are related only to the standing wave nature of the radiation fields. These additional terms have origins similar to that responsible for the Lamb dip[3] effect. The two applied standing waves may be decomposed into four traveling wave components. The additional terms may be thought of as a coupling between two components traveling in opposite directions. The frequency dependence of these additional terms will be quite different from that arising from coupling of components traveling in the same direction. Because of this the experimental conditions for observation of these additional terms are considerably different.

When the sample is within the resonator additional complications can arise. Due to the oscillator nature of the system, the output power is determined regeneratively by losses in the system. We will show that, under certain conditions, these effects can be made unimportant and the lineshape of the non-linear absorption or the gain characteristics may be observed directly by observing the behavior of the total output power. The discussion is given in section 4 with details of calculations presented in appendix D.

The discussions of this paper are specifically formulated for Doppler broadened atomic transitions. However, the results obtained in section 2 are directly applicable to any inhomogeneously broadened line with closely spaced structure. This kind of structure is encountered in numerous resonances in solids.

Furthermore, in the case of solids, resonances in the microwave region are also inhomogeneously broadened. Accordingly, the discussions in section 2 suggest a number of interesting applications involving resonances in the microwave region.

2. TRAVELLING WAVE RADIATION FIELDS

2a. Theory

Consider the three level atomic system shown in figure 1 subjected to optical traveling waves at frequencies ν_1 and ν_2. Assume that levels two and three are well resolved with respect to their natural widths, but not necessarily with respect to the Doppler width for transitions between either of them and level one. Assume also that $(\omega_{32}\ u/c)$[4] is negligible compared to the natural width of any level (where u is the average thermal velocity and c is the velocity of light). Finally, assume the Doppler width of the transitions to be considerably larger than their natural width. For the above system, a particular small band of atoms within the atomic velocity distribution will see ν_1 Doppler shifted close to ω_{21} and thus will interact strongly with the radiation at frequency ν_1. For another small band in the velocity distribution the same will be true pertaining to ν_2 and ω_{31}. Assume now that we can tune the spacing ω_{32}(for example with a magnetic field). At some particular value of ω_{32} the two sets of atoms in the velocity distribution will degenerate into each other. In this case the same group of atoms within the velocity distribution will resonate simultaneously with both traveling waves. If we consider the effect of saturation of level populations, we note that the influence of the field at the frequency ν_1 will result in a change of transmission coefficient(gain or attenuation)of the field at frequency ν_2 and vica versa. This will not occur if the frequencies of the applied fields are such that different sets of atoms within the velocity distribution interact most strongly with the two fields. Accordingly, we may expect to find a resonant behavior in the coupling between the two applied fields at that value of ω_{32} at which both fields interact with the same velocity atoms. The frequency condition for the

resonance is readily obtained. We must have $\nu_1(1 - v_R/c) = \omega_{21}$ and for the same atomic velocity, v_R, $\nu_2(1-v_R/c)=\omega_{31}$. Eliminating v_R/c gives the condition. We are interested in the case where ω_{32} is not too large (for example it is a small Zeeman splitting) and the discussion in footnote (4) applies. We then obtain

$$\omega_{32} = \nu_2 - \nu_1 \qquad (1)$$

that is the frequency spacing between levels is tuned to equal the separation between the applied frequencies. It should be noted that in a perturbation treatment of polarization induced by the presence of the applied fields, the saturation effect appears in the non-linear (third order and higher) terms. To the lowest order of perturbation where only linear absorption (or gain) of a medium is considered, the coupling between the two fields is entirely absent.

In the three level system atoms whose velocity is not near v_R may still have an appreciable Raman interaction in the presence of two strong applied fields. A Raman interaction in this case leads to an atom making a transition from level 2 to level 3 by absorbing a photon at frequency ν_2 and emitting a photon at frequency ν_1. This corresponds to a rate of transition proportional to the product of the powers at frequencies ν_1 and ν_2. The frequency condition for such occurence is, however, again given by equation 1. The Raman interaction has an additional resonant form[5] and thus is particularly strong for atoms whose velocity is near v_R. This additional effect is appreciable only for the condition of equation 1.[6] We shall see that the same frequency condition for both saturation effects and Raman processes has a profound influence on the shape of the resonance in the third order polarization. In

calculation of the induced polarization, the Raman terms also appear in third order.

The Hamiltonian for the atom with energy levels of figure 1 is taken to be

$$H = H_o - p \cdot E \tag{2}$$

where p is the dipole moment operator and the electromagnetic field E for two traveling waves is

$$\begin{aligned} E(z,t) &= \sum_{\mu=1}^{2} 2E_\mu \cos(\nu_\mu t - k_\mu z) \\ &= \sum E_\mu \left[e^{i(\nu_\mu t - k_\mu z)} + e^{-i(\nu_\mu t - k_\mu z)} \right] \end{aligned} \tag{3}$$

We wish to calculate the third order polarization P(r, t) in the atomic sample and to exhibit the behavior discussed above. We consider the density matrix $\rho(r, t, v, t_o, \alpha)$ describing the ensemble of atoms which are at position "r" at time "t", are moving with velocity "v", and which were excited to state α during their last collision at "t_o".[7] The equations of motion for the density matrix taken with respect to the three unperturbed states are[8]

(a) $$\left(\frac{d}{dt} - i\omega_{21} + \gamma_{21} \right) \rho_{12} = iV(t)\mu_{12}(\rho_{22} - \rho_{11}) - iV(t)\mu_{13}\rho_{32}$$

(b) $$\left(\frac{d}{dt} + i\omega_{31} + \gamma_{31} \right) \rho_{31} = iV(t)\mu_{31}(\rho_{33} - \rho_{11}) + iV(t)\mu_{21}\rho_{32}$$

(c) $$\left(\frac{d}{dt} + i\omega_{32} + \gamma_{32} \right) \rho_{32} = -iV(t)\mu_{31}\rho_{12} + iV(t)\mu_{12}\rho_{31}$$

(d) $$\left(\frac{d}{dt} + \gamma_{22} \right) \rho_{22} = iV(t)(\mu_{12}\rho_{21} - \mu_{21}\rho_{12})$$

(e) $$\left(\frac{d}{dt} + \gamma_{33}\right)\rho_{33} = \iota V(t)(\mu_{13}\rho_{31} - \mu_{31}\rho_{13})$$

(f) $$\left(\frac{d}{dt} + \gamma_{11}\right)\rho_{11} = -\iota V(t)(\mu_{12}\rho_{21} - \mu_{21}\rho_{12}) - \iota V(t)(\mu_{13}\rho_{31} - \mu_{31}\rho_{13})$$

(g) $$\rho_{21} = \rho_{12}^{*} \quad , \quad \rho_{13} = \rho_{31}^{*} \quad , \quad \rho_{23} = \rho_{32}^{*} \qquad (4)$$

The decay of the density matrix elements is assumed to arise from spontaneous emission. This means of decay is introduced in the above by assuming that the probability amplitude of a given level, α , suffers an exponential decay in the form $\exp(-\gamma_\alpha t/2)$. Accordingly, the decay of $\rho_{\alpha\beta}$ is described by the parameter $\gamma_{\alpha\beta} = (\gamma_\alpha + \gamma_\beta)/2$ with $\gamma_{\alpha\alpha} = \gamma_\alpha$ as the natural width of level α. The parameters μ_{12} and μ_{13} appearing in the above equations are the matrix elements connecting levels and 1, 2 and 1, 3 respectively. We assume there are no other matrix elements. For the ensemble in question

$$V(t') = E\left[r - v(t-t'), t'\right] \qquad (6)$$

for t> t'(since the atom of velocity v arrives at r at t) and the initial conditions on (4) are

$$\rho_{\alpha\alpha}(r, t_0; v, t_0, \alpha) = 1$$

$$\rho_{\gamma\beta}(r, t_0; v, t_0, \alpha) = 0 \quad , \beta, \gamma \neq \alpha \qquad (7)$$

The polarization due to the ensemble is given by

$$P(r,t;v,t_0,\alpha) = \mu_{21}\rho_{12}(r,t;v,t_0,\alpha) + \mu_{13}\rho_{31}(r,t;v,t_0,\alpha) \tag{8}$$

+ complex conjugate

Let the rate of production per unit volume, via collisions, of atoms of velocity v in the state α be $\Lambda_\alpha(v)$. Then the actual polarization in the material is

$$P(r,t) = \sum_\alpha \int_{-\infty}^{\infty} dv\, \Lambda_\alpha(v) \int_{-\infty}^{t} dt_0\, P(r,t;v,t_0,\alpha) \tag{9}$$

(It is assumed Λ_α is independent of time and position).

Since we are interested in the third order polarization, we seek a solution of equations (4) and (7) to third order in perturbation V(t). We use the standard iteration technique which consists of finding a zero order solution by setting V = 0 and then using this solution to obtain successive corrections. To this end it is convenient to introduce

$$\sigma_{ij} = \rho_{ij} \exp\left[(i\omega_{ij} + \gamma_{ij})t\right] \tag{10}$$

It is shown in appendix A that the equations of motion and the initial condition for $\sigma(r,t;v,t_0,\alpha)$ are [9]

(a) $$\frac{d\sigma_{12}}{dt} = -iV(t)e^{-i\omega_{21}t}\mu_{12}\left(\sigma_{22}e^{(\gamma_{12}-\gamma_2)t} - \sigma_{11}e^{(\gamma_{12}-\gamma_1)t}\right) - i\mu_{13}V(t)\sigma_{32}e^{-i\omega_{31}t}e^{(\gamma_{12}-\gamma_{32})}$$

(b) $$\frac{d\sigma_{31}}{dt} = iV(t)e^{i\omega_{31}t}\mu_{31}\left(\sigma_{33}e^{(\gamma_{13}-\gamma_3)t} - \sigma_{11}e^{(\gamma_{13}-\gamma_1)t}\right) + i\mu_{21}V(t)\sigma_{32}e^{i\omega_{21}t}e^{(\gamma_{13}-\gamma_{32})}$$

(c) $$\frac{d\sigma_{32}}{dt} = -i V(t)\mu_{31}\sigma_{12} e^{i\omega_{31}t} e^{(\gamma_{32}-\gamma_{12})t} + i V(t)\mu_{12}\sigma_{31} e^{-i\omega_{21}t} e^{(\gamma_{32}-\gamma_{31})t}$$

(d) $$\frac{d\sigma_{22}}{dt} = -i V(t)\left(\mu_{21}\sigma_{12} e^{i\omega_{21}t} e^{(\gamma_2-\gamma_{12})t} - \mu_{12}\sigma_{21} e^{-i\omega_{21}t} e^{(\gamma_2-\gamma_{12})t}\right)$$

(e) $$\frac{d\sigma_{33}}{dt} = i V(t)\left(\mu_{13}\sigma_{31} e^{-i\omega_{31}t} e^{(\gamma_3-\gamma_{13})t} - \mu_{31}\sigma_{13} e^{i\omega_{31}t} e^{(\gamma_3-\gamma_{13})t}\right)$$

(f) $$\frac{d\sigma_{11}}{dt} = i V(t)\left(\mu_{21}\sigma_{12} e^{i\omega_{21}t} e^{(\gamma_1-\gamma_{12})t} - \mu_{12}\sigma_{21} e^{-i\omega_{21}t} e^{(\gamma_1-\gamma_{12})t}\right) - i V(t)\left(\mu_{13}\sigma_{31} e^{-i\omega_{31}t} e^{(\gamma_1-\gamma_{13})t} - \mu_{31}\sigma_{13} e^{i\omega_{31}t} e^{(\gamma_1-\gamma_{13})t}\right)$$

(11)

(g) $$\sigma_{21} = \sigma_{12}^{*}, \qquad \sigma_{13} = \sigma_{31}^{*}, \qquad \sigma_{23} = \sigma_{32}^{*}$$

$$\sigma_{\alpha\alpha}(r, t_0; v, t_0, \alpha) = e^{\gamma_\alpha t_0} \tag{12}$$

It is shown in appendix B that the solution when the atom starts in state one or three (α = 1 or 3) can be obtained simply from the solution when α = 2. We thus need consider only $\sigma_{22}^{(0)}$. If we set V (t) = 0 we get from (11d) and (12) $\sigma_{22}^{(0)} = e^{\gamma_2 t}$ We integrate this solution in (11a) and (11g) to get first order solutions $\sigma_{12}^{(1)}$ and $\sigma_{21}^{(1)}$. These first order solutions give second order solutions $\sigma_{11}^{(2)}$, $\sigma_{22}^{(2)}$ and $\sigma_{32}^{(2)}$ via equations (11d), (11f) and (11c). Finally we use these in (11a) and (11b) to find the third order $\sigma_{12}^{(3)}$ and $\sigma_{31}^{(3)}$.

The procedure just outlined leads to a sum of triple integrals for $\sigma_{12}^{(3)}(r, t; v, t_0, \alpha)$ and $\sigma_{31}^{(3)}(r, t; v, t_0, \alpha)$. There are eight terms in all. The complete algebraic manipulations are rather lengthy. They are carried out in a shorthand

manner in appendix B. Here we will follow through the typical manipulations for one important term to illustrate the procedure. We will use $\sigma_{32}^{(2)}$ which will lead to one of the Raman[10] terms as is explained later.

The term in $\sigma_{31}^{(3)}(r, t; v, t_o, 2)$ which comes about due to $\sigma_{32}^{(2)}$ is given by

$$\begin{aligned}\sigma_{31\,typ}^{(3)}(r,t;v,t_0,2) = -i\,|\mu_{12}|^2\mu_{31}\exp(\gamma_2 t_0) \\ \times \int_{t_0}^{t} dt'\,V(t')\exp\left[i\omega_{21}+(\gamma_{13}-\gamma_{32})\right]t' \\ \int_{t_0}^{t'} dt''\,V(t'')\exp\left[i\omega_{31}+(\gamma_{32}-\gamma_{12})\right]t'' \\ \int_{t_0}^{t''} dt'''\,V(t''')\exp\left[-i\omega_{21}+(\gamma_{12}-\gamma_2)\right]t'''\end{aligned} \tag{13}$$

We integrate overall possible initial times from $-\infty$ to t. Changing the order of integration three times gives

$$\begin{aligned}\sigma_{31\,typ}^{(3)}(r,t;v,2) = \left(-i\,|\mu_{12}|^2\mu_{31}/\gamma_2\right) \\ \times \int_{-\infty}^{t} dt'\,V(t')\exp\left[i\omega_{21}+(\gamma_{13}-\gamma_{32})\right]t' \\ \int_{-\infty}^{t'} dt''\,V(t'')\exp\left[i\omega_{31}+(\gamma_{32}-\gamma_{12})\right]t'' \\ \int_{-\infty}^{t''} dt'''\,V(t''')\exp\left[-i\omega_{21}+\gamma_{12}\right]t'''\end{aligned} \tag{14}$$

We next substitute for the V's using equations (6) and (3). We make the usual rotating wave approximation, that is we neglect any frequency dependent exponential which cannot have a resonance. We then have

$$\sigma^{(3)}_{31\,typ}(r,t;v,2) = \left(i\,|\mu_{12}|^2\mu_{31}/\gamma_2\right)$$
$$\times \sum_{\mu\rho\sigma} E_\mu E_\rho E_\sigma \exp i\left[(k_\sigma+k_\rho-k_\mu)z-(\nu_\sigma+\nu_\rho-\nu_\mu)t - kvt\right]$$
$$\times \int_{-\infty}^{t} dt' \exp -\left[i(\nu_\sigma-\omega_{21}-kv)-(\gamma_{13}-\gamma_{32})\right]t'$$
$$\int_{-\infty}^{t'} dt'' \exp -\left[i(\nu_\rho-\omega_{31}-kv)-(\gamma_{32}-\gamma_{12})\right]t''$$
$$\int_{-\infty}^{t''} dt''' \exp\left[i(\nu_\mu-\omega_{21}-kv)+\gamma_{12}\right]t''' \tag{15}$$

whre $kv = k_1 v \approx k_2 v^{(4)}$ with $k_1 = \nu_1/c$ and $k_2 = \nu_2/c$. These integrals can be straightforwardly done and on using the relation $\omega_{31}-\omega_{21} = \omega_{32}$ and equation (10) one obtains for the polarization due to the typical term

$$P^{(3)}_{typ}(r,t;v,2) = \left(i\,|\mu_{12}|^2|\mu_{31}|^2\Lambda_2(v)/\gamma_2\right)$$
$$\times \sum E_\mu E_\rho E_\sigma \exp i\left[(k_\sigma+k_\rho-k_\mu)z-(\nu_\sigma+\nu_\rho-\nu_\mu)t\right]$$
$$\times \Big\{ \left[\gamma_{12}+i(\nu_\mu-\omega_{21}-kv)\right]^{-1}$$
$$\left[\gamma_{32}+i(-\nu_\rho+\nu_\mu+\omega_{32})\right]^{-1}$$
$$\left[\gamma_{13}+i(-\nu_\sigma-\nu_\rho+\nu_\mu+\omega_{31}+kv)\right]^{-1}\Big\} \tag{16}$$

+ complex conjugate

Equation (16) is seen to consist of a sum of terms each of which contains a product of three complex Lorentzians.[11] Each complex Lorentzian can have a resonance when the frequency combination appearing in it goes to zero. Two of the resonant frequency terms, $(\nu_\mu - \omega_{21} - kv)$ and $(-\nu_\sigma - \nu_\rho + \nu_\mu + \omega_{31} + kv)$, enter as the difference between an applied optical frequency field and an atomic resonance frequency. The effect of the atomic velocity appears in these expressions simply as a Doppler shift in the applied frequency. The middle complex Lorentzian depends on the detuning of ω_{32} from the difference of two applied optical frequencies. In taking this difference Doppler shifting has cancelled out due to the approximation of footnote (4).

When ω_{32} is close to $\nu_2 - \nu_1 = \Delta$ the middle term will be resonant when the indices $\rho = 2$ and $\mu = 1$. If we restrict attention to polarization which varies at ν_1 or ν_2, then for this choice of ρ and μ, we need $\sigma = 1$. This term in the sum gives

$$
\begin{aligned}
P^{(3)}_{type}(r, t; v, 2) = {} & i\,|\mu_{12}|^2\,|\mu_{31}|^2 N_2(v) \\
& \times E_1^2 E_2\, e^{i(k_2 z - \nu_2 t)} \\
& \times \Big\{ \left[\gamma_{12} + i(\nu_1 - \omega_{21} - kv)\right]^{-1} \left[\gamma_{32} + i(\omega_{32} - \Delta)\right]^{-1} \\
& \quad \left[\gamma_{13} + i(\omega_{31} - \nu_2 + kv)\right]^{-1} \Big\} \\
& + \text{complex conjugate}
\end{aligned}
\tag{17}
$$

where $\Delta = \nu_2 - \nu_1$ and $N_2(v) = \Lambda_2/\gamma_2$ is the steady state population of level 2 as a function of velocity in the absence of any applied radiation. For any other choice of indices μ, ρ, σ the middle complex Lorentzian will not be resonant. For

some particular velocity atom one or the other(but, inspection shows, not both) velocity dependent complex Lorentzians may then be on resonance. Clearly, though, when we sum (16) over the velocity distribution the term written in (17) will be the most important.

The appearance of ω_{32} in (16) is due to the fact that we have obtained our typical term for third order polarization through consideration of $\rho_{32}^{(2)}$. The terms so obtained will be referred to as the Raman terms.[10] In addition to these Raman terms are the terms in third order polarization which come about due to the diagonal density matrix components in second order. Such contributions are the saturation terms arising because population changes due to one applied field change the rate of transition due to the other.

As may be verified in appendix B, the saturation terms in the third order polarization will contain two complex Lorentians resonant when a Doppler shifted applied frequency equals an atomic transition frequency (ω_{21} or ω_{31}). However, the term corresponding to the middle Lorentzian in (16) will contain simply the difference of two optical frequencies. Such a term is not resonant. It is large only when the two optical frequencies are the same, and it has no tunable behavior at all.

Keeping only the most important terms in the summation over indices, as above, the complete form of third order polarization due to the Raman terms is

$$P^{(3)}_{Ram}(r,t;v) = i(N_2-N_1)|\mu_{12}|^2|\mu_{31}|^2\{E_1^2E_2\exp i(k_2z-\nu_2t)$$
$$\times[\gamma_{32}+i(\omega_{32}-\Delta)]^{-1}[\gamma_{12}+i(\nu_1-\omega_{21}-kv)]^{-1}$$

$$\left[\gamma_{13} + i(\omega_{31} - \nu_2 + kv)\right]^{-1}$$

$$- E_1^2 E_2 \exp - i(k_1 z - \nu_1 t)$$

$$\times \left[\gamma_{32} + i(\omega_{32} - \Delta)\right]^{-1} \left[\gamma_{12} + i(\nu_1 - \omega_{21} - kv)\right]^{-1}$$

$$\left[\gamma_{12} + i(\nu_1 - \omega_{21} - kv)\right]^{-1} \Bigg\}$$

$$+ i(N_3 - N_1) \left\{ \begin{array}{l} \text{above expression interchange 1 and 2 in E,} \\ \text{interchange 2 and 3 in } \omega \text{ and} \\ \quad + \text{ complex conjugate} \end{array} \right\}, \tag{18}$$

and from appendix B, the complete form of the third order polarization due to the saturation term is

$$P_{sat}(r, t; v) = i(N_2 - N_1) \Bigg\{ \frac{2|\mu_{12}|^4 \gamma_{12}}{\gamma_1 \gamma_2} E_1^3 \exp - i(k_1 z - \nu_1 t)$$

$$\times \left[\gamma_{12} + i(\nu_1 - \omega_{21} - kv)\right]^{-1} \left[\gamma_{12} + i(\nu_1 - \omega_{21} - kv)\right]^{-1}$$

$$+ \left[\gamma_{12} + i(\nu_1 - \omega_{21} - kv)\right]^{-1} \left[\gamma_{12} + i(\omega_{21} - \nu_1 + kv)\right]^{-1}$$

$$+ \frac{2|\mu_{12}|^4 \gamma_{12}}{\gamma_1 \gamma_2} E_2^3 \exp - i(k_2 z - \nu_2 t)$$

$$\times \left[\gamma_{12} + i(\nu_2 - \omega_{21} - kv)\right]^{-1} \left[\gamma_{12} + i(\nu_2 - \omega_{21} - kv)\right]^{-1}$$

$$+ \left[\gamma_{12} + i(\nu_2 - \omega_{21} - kv)\right]^{-1} \left[\gamma_{12} + i(\omega_{21} - \nu_2 + kv)\right]^{-1}$$

$$+ \frac{|\mu_{12}|^2 |\mu_{31}|^2}{\gamma_1} E_1^2 E_2 \exp i(k_2 z - \nu_2 t)$$

$$\times \left[\gamma_{12} + i(\nu_1 - \omega_{21} - kv)\right]^{-1} \left[\gamma_{13} + i(\omega_{31} - \nu_2 + kv)\right]^{-1}$$

$$+ \left[\gamma_{12} + i(\omega_{21} - \nu_1 + kv)\right]^{-1} \left[\gamma_{13} + i(\omega_{31} - \nu_2 + kv)\right]^{-1}$$

$+ i(N_3 - N_1)$ (same replacement as in (18))
+ complex conjugate (19)

As previously pointed out the terms in (18) contain a product of three Lorentzians, while those of (19) contain only two. The additional resonance in the Raman terms is the $(\omega_{32} - \Delta)$ resonance which is unaffected by the atomic velocity. It should be noted, however, that some of the other velocity dependent products have an implicit dependence on the difference $(\omega_{32} - \Delta)$ in that they depend on both $\omega_{21} - \nu_1$ and $\omega_{31} - \nu_2$. The above equations are consistent with previous treatments of the interaction of three levels with two fields for stationary atoms.[5]

Let us now consider what happens when we integrate (18) and (19) over the velocity distribution. Let up suppose that the width of the velocity distribution is characterized by u and that $ku \ll \gamma$, i.e. there is large Doppler broadening.

(According to this notation, ku determines the exact Doppler width. The linear polarization terms, not discussed here, depend on ku in the form of $\exp-[(\nu-\omega)/ku]^2$ with ν representing the frequency of one of the applied fields and ω, one of the atomic resonances). Furthermore, for the sake of simplicity, we assume $|\nu_1-\omega_{21}|$ and $|\nu_2-\omega_{31}| \ll ku$, the detuning from resonance is small compared to the Doppler width. (We discuss the results without this latter restriction in appendix C. It is shown that the important conclusions of this section are unchanged.) We now assert that when we sum over the velocity distribution the terms in (18) and (19) in which the velocity has the same sign in each of the product of Lorentzians will be smaller by a factor γ/ku than the terms for which the velocity enters with opposite signs.

A complete discussion of the integrals which arise when a Maxwell velocity distribution is assumed is given in appendix C. Also in appendix C, it is shown that the above assertion is due to the contribution from atoms which are not exactly on resonance.[13] For the larger terms these atoms contribute in phase with each other, while for the smaller terms they tend to cancel out each other.

For a Maxwell distribution

$$N_\alpha(v) = \frac{N_\alpha}{\pi^{1/2}u} \exp - v^2/u^2$$

a term of the form

$$\frac{1}{\gamma+i(x+kv)} \; \frac{1}{\gamma'+i(y-kv)}$$

(where x and y may stand for $\pm(\nu_1-\omega_{21})$ or $\pm(\nu_2-\omega_{31})$)

leads to an integral of the form

$$\frac{1}{\pi^{1/2} u} \int_{-\infty}^{\infty} \frac{dv\, e^{-v^2/u^2}}{[\gamma + i(x + kv)]\,[\gamma' + i(y - kv)]}$$

It is shown in appendix C that such an integral to order γ/ku and for $|x|$ and $|y|$ less than ku is given by

$$\frac{2\pi^{1/2}}{ku} \frac{1}{\gamma + \gamma' + i(x+y)} \tag{20}$$

Using formula (20) in expressions (18) and (19) after some slight algebra we get the complete expression for the third order polarization to order

$$\begin{aligned} P^{(3)}(r,t) = {} & \frac{2\pi^{1/2}}{ku} i\,(N_2 - N_1) \\ & \times \Bigg\{ \frac{|\mu_{12}|^4}{\gamma_1 \gamma_2} \left[E_1^3 e^{-i(k_1 z - \nu_1 t)} + E_2^3 e^{-i(k_2 z - \nu_2 t)} \right] \\ & \quad + \frac{|\mu_{12}|^2 |\mu_{31}|^2}{\gamma_1} E_1^2 E_2 e^{i(k_2 z - \nu_2 t)} \\ & \quad \times \left[2\gamma + i(\omega_{32} - \Delta) \right]^{-1} \left[1 + \gamma_1 \left[\gamma_{23} + i(\omega_{32} - \Delta) \right]^{-1} \right] \Bigg\} \\ & + \frac{2\pi^{1/2}}{ku} i\,(N_3 - N_1) \left\{ \begin{array}{ll} \text{in above,} & \text{interchange 1 and 2} \\ \text{in E,} & \text{interchange 2 and 3} \\ \text{in } \mu,\ \omega & \end{array} \right\} \\ & + \text{complex conjugate} \end{aligned} \tag{21}$$

where we have let $2\gamma = \gamma_{12} + \gamma_{13}$

Expression (21) shows the behavior described earlier in this section, namely, in addition to ordinary saturation terms, there appear cross terms in the mode amplitudes which show a resonant behavior near $\omega_{32} = \Delta$. We discuss the lineshape due these terms in an absorption experiment next. Although at first glance it would appear from (21) that this lineshape would be rather complicated, it will turn out that cancellations occur which make it simple with interesting implications.

2b. Discussion

An important feature of the expression for third order polarization given by (21) is the fact that resonances appear only in terms of the difference frequency $\nu_2 - \nu_1 = \Delta$. According to this expression, the exact frequencies of the two applied fields as referred to the center frequencies of the atomic transitions, ω_{21} and ω_{31} are unimportant.

We shall again point out that (21) is obtained for $|\nu_1 - \omega_{21}|$ and $|\nu_2 - \omega_{31}|$ much less than ku; namely, that ν_1 and ν_2 are assumed to be close to the center frequencies of the Doppler broadened atomic transitions, ω_{21} and ω_{31}. For large ku, (i.e. large Doppler width), this condition may remain valid even for ν_1 and ν_2 appreciably detuned from the peak frequencies of the atomic transitions, and expression (21) will hold. (It is shown in appendix C that for $\nu_1 - \omega_{21}$ comparable to ku but $|\omega_{32} - \Delta| \ll ku$, the resulting polarization, for this case of two travelling waves, is merely multiplied by an overall factor exp $-(\nu_1 - \omega_{21})^2/(ku)^2$). This factor remains nearly constant for changes in ν_1 and $\nu_2 \ll ku$. As a result of this, as long as the frequency $\nu_2 - \nu_1 = \Delta$ remains fixed, the absolute frequencies of the applied fields may be detuned appreciably without influencing the size of the third order

polarization. This fact has an important bearing on the necessary experimental condition for observation of the resonances involving $\Delta - \omega_{32}$ appearing in (21).

In most gas lasers, unless special care is taken, the exact optical frequencies ν_1 and ν_2 do not remain stable. Rather, they fluctuate throughout the entire portion of the Doppler response which has sufficient gain to allow oscillations. These fluctuations in frequency are due to external disturbances which cause fluctuations in the length of the Fabry-Perot resonator. However the magnitude of the fluctuations of the difference frequency, $\nu_2 - \nu_1 = \Delta$, is much less than those of the absolute frequencies, ν_1 and ν_2. Consequently, while ν_1 and ν_2 may suffer appreciable fluctuations, Δ remains nearly fixed. This relaxes considerably experimental requirements of frequency stability for observing the narrow resonances in expression (21).

Let us now consider, as an example, the simplified experimental arrangement shown in figure 2. The output from a Brewster angle gas laser oscillating in two modes passes through a sample gas and then onto a detector. The sample gas is assumed to have an atomic or molecular transition frequency which coincides closely with the laser frequency. It may be in either an absorbing or an emitting state. The sample gas is subjected to a magnetic field, either axial or perpendicular to the plane of the drawing. We will discuss the applicability of the three level model we are using to more general level structure in the next section. For now we assume one can isolate three pertinent levels and that the spacing between the upper two (for example Zeeman sublevels) can be magnetically tuned about the value determined by the laser cavity. We consider the power absorption in the cell as ω_{32} is tuned about Δ.

The absorption in the cell per unit length and time is given by

$$\frac{dW}{dz} = A\dot{P}\cdot E \tag{22}$$

where A is the cross sectional area of the beam, P is the time derivative of the total polarization vector and E is the total electric field. For the two travelling waves we take as before

$$E(z,t) = 2E_1 \cos(\nu_1 t - k_1 z) + 2E_2 \cos(\nu_2 t - k_2 z) \tag{23}$$

Absorption due to linear polarization in the sample will be characterized by the Doppler width ku, and thus will remain approximately constant for small magnetic tuning. If the field strengths are large enough to cause appreciable saturation in the sample then changes in absorption will be observed for small tuning (of the order of the natural linewidth) due the terms in the third order polarization which show the $\omega_{32} - \Delta$ resonance.

Let us write the part of equation (21) which shows this resonance as

$$P^{(3)}(r,t) =$$

$$C_1 E_1 E_2^2 \cos(\nu_1 t - k_1 z) + S_1 E_1 E_2^2 \sin(\nu_1 t - k_1 z)$$

$$+ C_2 E_1^2 E_2 \cos(\nu_2 t - k_2 z) + S_2 E_1^2 E_2 \sin(\nu_2 t - k_2 z)$$

(24)

The quantities $C_1E_2^2$ and $S_1E_2^2$ are respectively the contribution of the third order polarization to the real and imaginary part of the susceptibility at the frequency ν_1. Similarly $C_2E_1^2$ and $S_2E_1^2$ at frequency ν_2. If we substitute this expression and (23) into (22), integrate over the length of the cell and average over a long time compared to $1/(\nu_2-\nu_1)$ we obtain for the total absorbed power:

$$\bar{W} = -\nu \ell A E_1^2 E_2^2 (S_1 + S_2) \tag{25}$$

where ℓ is the length of the sample, $\nu = \nu_1 \approx \nu_2$

The lineshape of this absorption, then, is contained in S_1 and S_2. To put the expression (21) in the form (24) involves a somewhat lengthy algebraic manipulation. When the definitions $2\gamma = \gamma_{12}+\gamma_{13}$ and $\gamma_{ij} = (\gamma_i+\gamma_j)/2$ are substituted, however, the result comes out in a remarkably simple form. The result is:

$$S_1 = -\frac{4\pi^{1/2}}{\hbar 2\pi \gamma_1}(N_3 - N_1)\,|\mu_{12}|^2 |\mu_{13}|^2 \frac{\gamma_{32}}{\gamma_{32}^2 + (\omega_{32}-\Delta)^2} \tag{26}$$

S_2 = same with $(N_2 - N_1)$ replacing $(N_3 - N_1)$ (27)

The lineshape is thus given by the Lorentzian $\left[\gamma_{32}^2 + (\omega_{32}-\Delta)^2\right]^{-1}$. This expression for lineshape has one remarkable feature. The width of the Lorentzian $\gamma_{32} = (\gamma_2+\gamma_3)/2$ is entirely independent of the width of the lower level. This is quite contrary to what one would expect from considering, for instance, only the effect of saturation of the level populations, since the broader the lower level, the greater

is the range one could tune ω_{32} and still be on resonance with both applied fields (for the same velocity atom).

We may make several remarks with regard to the above behavior of the lineshape. First of all there are two terms in expression (21) one of which contains a single complex Lorentzian of width 2γ. This has come about due to the diagonal density matrix element $\rho^{(2)}_{22}$ and is thus strictly a saturation effect. The other term is a product of complex Lorentzians and has come about due to $\rho^{(2)}_{32}$ which is the Raman term. Each of these terms individually depends on the width of the lower level through the width $\gamma=(\gamma_{13}+\gamma_{12})/2$. However, in combining them the width γ_1 has cancelled out of the lineshape.

Second, we note that the above behavior of the lineshape is similar to a behavior involving spontaneous or stimulated Raman scattering in gases. In an ordinary type of Raman experiment, one deals primarily with two energy levels forming the initial and final state of the transition. In these cases, the presence of a third energy level enters only as a virtual state and does not satisfy any resonance condition. The atomic system undergoes transitions by absorbing a photon from a pump field at frequency ν and emitting a photon at a displaced frequency ν'. The frequency difference $\nu-\nu'$ is roughly equal to the level spacing of the initial and final state of the atom.

Consider the case of spontaneous Raman effect in which only the pump field is applied externally. It can readily be shown that the Doppler width of the spontaneous Raman scattering in the forward direction is $(\nu-\nu')u/c$ with u as average velocity of atoms. (In the backward direction the Doppler width is much larger, $(\nu+\nu')u/c$).) For cases where the Raman displaced frequency, $\nu-\nu'$, is

in the far infrared or submillimeter range $(\nu-\nu')u/c$ may be less than the natural or collision widths of the initial or final levels. In that case the Raman scattering in the forward direction will show no Doppler broadening but its width will be determined by the individual widths of the initial and final states. (But it is independent of the width of the virtual state.) The above shows that in the case of stimulated Raman effect in the forward direction, where in addition to the so called pump field at frequency ν there is an additional applied field in the same direction at a frequency close to ν', the width of the Raman resonance will be the same as that in the spontaneous case, namely limited by the width of the initial and the final state only.

In our case, our fields at frequency ν_1 and ν_2 are in the same direction and we assume $\nu_2-\nu_1$ to be close to ω_{32}. We note that atoms whose velocity is <u>not</u> such as to Doppler shift ν_1 close to ω_{21} (or, therefore, to Doppler shift ν_2 close to ω_{31}) will undergo stimulated Raman transitions entirely identical to the case of ordinary Raman effect described above. For these atoms, level one plays the role of a virtual level. Hence, their stimulated Raman response will have a width given only by the natural widths of levels 2 and 3. However, atoms whose velocity is such that the Doppler shifted ν_1 is close to ω_{21} will show a Raman behavior with a different frequency response. And for these atoms, the effect arising from saturation of population should also be considered.

From the above mentioned cancellations and the similarity of linewidth behavior with ordinary stimulated Raman effect, we note that there has been a cancellation of the saturation terms and "resonant Raman"[5] terms for those atoms whose velocity makes level one a resonant level leaving only the contribution

of those atoms which are somewhat off resonance.

It should be noted, however, that our overall result is quite different generally from usual discussions of stimulated Raman effect due to the previously mentioned loss of some terms by destructive interference in integrating over the velocity distribution.

It should also be pointed out that the frequency behavior of the non-linear part of the dispersion of the atomic resonance behaves similarly. This may be shown by inspecting the in phase, $C_1E_1E_2^2$ and $C_2E_1^2E_2$ of expression (24) through the terms appearing in equation (21). The width of these dispersive terms also depend only on the radiative width of the upper levels.

3. MORE GENERAL LEVEL STRUCTURE

Before we undertake the treatment of a more complex form of the radiation field, let us examine the applicability of our treatments in cases which involve more complex level structure. For this, we proceed by examining several specific examples where the level structure is assumed to arise from a Zeeman splitting of simple paramagnetic levels. Suppose first that transitions occur between atomic states of total angular momentum $j = 1$ to $j = 0$. Then in an axial magnetic field the energy levels are split as in figure 3a. Consider the laser fields to be linearly polarized perpendicular to the direction of the magnetic field. In this case only the $m_j = \pm 1$ Zeeman components of the $j = 1$ level have matrix elements connecting to the $j = 0$ level. The $j = 1$, $m = \pm 1$ and $j = 0$, $m = 0$ set constitute a three level system of the type we have analyzed. Accordingly when the frequency splitting of the $m = +1$ and $m = -1$ components becomes very close to the frequency separation $\Delta = \nu_2 - \nu_1$ resonant behavior should occur in the coupling terms of the third order polarization in a manner described in section 2.

Consider now a more complex system, the $j = 2$ to $j = 1$ transition shown in figure 3b. Let m_μ, g_μ and m_ℓ, g_ℓ represent the m and "g" values of the $j = 2$ and $j = 1$ state respectively and suppose $g_\ell > g_\mu$. The applied field will be again considered as linearly polarized and perpendicular to the H-field. Lines are drawn in figure 3b between the components with matrix elements connecting them. As the magnetic field is increased from zero the first matching occurs when $m_\mu = -2$ and $m_\mu = +2$ are separated by Δ. But these two components do not have a common lower level. Furthermore, because of the different g values of

of the upper and lower levels, the frequency spacing of all the allowed transitions indicated in figure 3b are different. As a result, the Doppler effect will lead to a situation where a given atom can interact with the applied fields through coupling via only one of these transitions. Thus nothing interesting happens. As the magnetic field is further increased the $m_\ell = \pm 1$ components are separated by Δ. These both are connected to $m_\mu = 0$ and resonant behavior is observed in the output, from which g_ℓ can be measured. Finally the separations between $m_\mu = -2$ and $m_\mu = 0$, between $m_\mu = -1$ and $m_\mu = +1$ and between $m_\mu = 0$ and $m_\mu = +2$ are separated by Δ. The $m_\mu = \pm 1$, $m_\ell = 0$ clearly form a distinct 3 level subsystem. Furthermore since the "g" values are different it will always be different atoms in the velocity distribution which are coupled to the two fields via the three levels ($m_\mu = -2, 0$; $m_\ell = -1$) or via the levels ($m_\mu = 0, +2$, $m_\ell = +1$). In other words, transitions are induced within the ($m_\mu = -2, 0$, $m_\ell = -1$) set of levels separately from those induced within the ($m_\mu = 0, +2$, $m_\ell = +1$) set. These two sets of levels are not coupled together insofar as the Zeeman splitting of the lower level is appreciably different than that of the upper level.

Similar arguments to the above may be advanced to show that our analysis may be directly applicable to resonances with more complex structures such as those arising from fine or hyperfine interactions. The conditions for interactions involving three levels at a time may be readily inspected in each case as was done in the above.

4. STANDING WAVE RADIATION FIELDS

4a. Theory

In this section we treat a case where the two monochromatic fields are in the form of standing waves. The atomic resonance will be assumed, as before, to consist of two transitions centered at frequencies ω_{21} and ω_{31}. (See fig. 1). The Doppler width of these two transitions may be overlapping. However, as before, we consider the transitions to be well resolved with respect to their natural widths. This problem is, in particular, applicable to a case where the atomic system is placed within the resonator of a laser oscillating in two Fabry-Perot modes at frequencies ν_1 and ν_2.

In cases where an active laser material itself consists of two closely spaced transitions, the calculated polarization may be used to obtain details of operation of such a laser. Such details would be obtained by self consistency arguments, as in the treatment of Lamb.[3]

Each of the standing waves in the applied radiation may be decomposed into two traveling waves of half the original amplitude in opposite directions. Therefore, the third order polarization will again show resonances of the form we have just discussed for traveling waves. In addition, a number of new resonances will appear.

These additional resonances are of similar origin to the effects described by Lamb[3] for a two level laser. An atom of velocity, v, sees the right traveling component and the left traveling component of a standing wave at different frequencies due to the Doppler effect. Therefore, in general, different atoms within

the velocity distribution will interact resonantly with the right and left traveling components. For our three level system there are two atomic transition frequencies ω_{21} and ω_{31}. For two standing waves, at frequencies ν_1 and ν_2, generally, there will be eight different sets of atoms within the velocity distribution which can interact resonantly. Each set is centered about one of the velocities v_R determined by $\nu_1(1 \pm v_R/c) = \omega_{21}$, $\nu_1(1 \pm v_R/c) = \omega_{31}$ or $\nu_2(1 \pm v_R/c) = \omega_{21}$ $\nu_2(1 \pm v_R/c) = \omega_{31}$ When ν_1 and ν_2 satisfy one of several easily determined relationships with respect to ω_{21} and ω_{31}, (we will point out these relationships in part (b) of this section) two bands in the velocity distribution will overlap. In this case the same atoms will interact resonantly with two traveling wave components in <u>opposite directions</u> (except for the one case described in the previous sections). Due to the saturation of level populations each overlap condition, as previously discussed, is characterized by a resonance in the non-linear polarization of the atomic medium.

We may take over the treatment of section (2a) up to the typical form given in equation 14. At this point instead of expression (3) for a travelling wave, we must substitute the standing wave

$$E(z,t) = \sum_{\mu=1}^{2} 2E_\mu \cos \nu_\mu t \sin k_\mu z$$
$$= \sum E_\mu \left(e^{i\nu_\mu t} + e^{-i\nu_\mu t} \right) \sin k_\mu z \qquad (28)$$

into perturbation $V(t)$ given by equation (6). Here $k_\mu = \pi n_\mu / L$ with L the length of the optical resonator and n_μ a large integer. The functions $\sin k_\mu z$ are taken to represent approximate normal modes of the Fabry-Perot resonator.[3]

On making the rotating wave approximation we have from (14)

$$\sigma^{(3)}_{31\,typ}(r,t;\nu,\Omega) = \left(i|\mu_{12}|^2\mu_{31}/\gamma_2\right) \sum E_\mu E_\rho E_\sigma$$
$$\int_{-\infty}^{t} dt' \exp -\left[i(\gamma_\sigma-\omega_{21})-(\gamma_{13}-\gamma_{32})\right]t'$$
$$\sin k_\mu(z-\nu t-\nu t')$$
$$\int_{-\infty}^{t'} dt'' \exp -\left[i(\gamma_\rho-\omega_{31})-(\gamma_{32}-\gamma_{12})\right]t''$$
$$\sin k_\rho(z-\nu t-\nu t'')$$
$$\int_{-\infty}^{t''} dt''' \exp \left[i(\gamma_\mu-\omega_{21})+\gamma_{12}\right]t'''$$
$$\sin k_\mu(z-\nu t-\nu t''')$$

(29)

It is convenient to remove the variable limits in equation (29) by introducing

$$\tau' = t-t', \quad \tau'' = t'-t'', \quad \tau''' = t''-t'''$$

Making this substitution and using (8) and (10) we have for the polarization (after slight algebra) due to the typical term

$$P^{(3)}_{\sigma\rho\mu}(z,t;v,\alpha) = i\,|\mu_{12}|^2\mu_{31}N_2(v)$$
$$\sum E_\sigma E_\rho E_\mu \exp i(-\nu_\sigma-\nu_\rho+\nu_\mu)t$$
$$\times\int_0^\infty d\tau'\int_0^\infty d\tau''\int_0^\infty d\tau'''\exp-[\gamma_{13}+i(-\nu_\sigma-\nu_\rho+\nu_\mu+\omega_{31})]\tau'$$
$$\exp-[\gamma_{32}+i(\nu_\mu-\nu_\rho+\omega_{32})]\tau''$$
$$\exp[\gamma_{12}+i(\nu_\mu-\omega_{21})]\tau'''\,[\sin k_\sigma(z-v\tau')$$
$$\sin k_\rho(z-v\tau'-v\tau'')\sin k_\mu(z-v\tau'-v\tau''-v\tau''')] \tag{30}$$

By making use of the identities for products of trigonometric functions the product of three sine functions in (30) is equal to

$$\frac{1}{4}\Big\{-\sin[(k_\sigma+k_\rho+k_\mu)z-kv(3\tau'+2\tau''+\tau''')]$$
$$+\sin[(-k_\sigma+k_\rho+k_\mu)z-kv(\tau'+2\tau''+\tau''')]$$
$$+\sin[(k_\sigma+k_\rho-k_\mu)z-kv(\tau'-\tau''')]$$
$$+\sin[(k_\sigma-k_\rho+k_\mu)z-kv(\tau'+\tau''')]\Big\} \tag{31}$$

The first term in (31) varies spatially as three times the optical wavelength λ. Therefore the contribution of this term to absorption will be of order λ/ℓ (ℓ= length of cell) compared to the other terms.

We assume λ/ℓ is small and neglect the first term of (31). The remaining terms can be written

$$\frac{1}{4}\Big\{ \sin(-k_\sigma + k_\rho + k_\mu) z \, e^{ikv(\tau' + 2\tau'' + \tau''')} + \sin(k_\sigma + k_\rho - k_\mu) z \, e^{ikv(\tau' - \tau''')} + \sin(k_\sigma - k_\rho + k_\mu) z \, e^{ikv(\tau' + \tau''')} + \text{terms odd in } v \tag{32}$$

The terms odd in v will clearly not contribute to overall polarization if the atomic velocity distribution is symmetric.

Consider the result of substituting (32) back into (30). It is easily seen that each τ integral gives a complex Lorentzian and thus each term in (32) leads to a product of three complex Lorentzians. In particular the τ'' integral applies to the resonance involving ω_{32} and differences in applied frequencies, $\nu_2 - \nu_1 = \Delta$. The three terms of (32) may be related to different Doppler effects in the individual Lorentzians due to the presence of standing waves. Thus, for our typical term the $e^{ikv(\tau' - \tau''')}$ leads to the terms we have already encountered in the traveling wave treatment. They thus represent the effect of travelling wave components in the same direction. The resulting expressions are identical to those obtained in previous sections. The $e^{ikv(\tau' + \tau''')}$ changes the sign of one velocity term thus making it represent an

interaction with a travelling wave in the opposite direction. The $\exp i k v (\tau' + 2\tau'' + \tau''')$ terms represent, in additions, the occurrence of the $(\omega_{32} - \Delta)$ resonance for travelling components in opposite directions.

We now consider integrating over the velocity distribution. The interpretation of the terms in (32) as to directions of standing waves is, of course, not unique. When we consider other than our typical Raman term, the interpretation will depend on how the applied frequencies enter into the expressions (see appendix B). However, the $\exp i k v (\tau' - \tau''')$ will always involve two Lorentzians featuring Doppler shifts and the sign of v will always be different in the two. The $\exp i k v (\tau' + \tau''')$ likewise involve two Doppler shifts but the sign of v is the same. We have already mentioned (see appendix C) that the former (opposite sign of v) terms contribute more than the latter term by an order $\gamma / k u$ after integrating over the velocity distribution. It is shown in appendix C that the terms arising from $\exp i k v (\tau' + 2\tau'' + \tau''')$ may also be ignored under the same conditions (and to the same order $\gamma / k u$). We may evaluate the only important contribution, then, using formula (20).

When we apply formula (20) to all the third order contributions of appendix B, and keep only those values of the indices μ, ρ, σ which lead to possible resonant forms at frequency ν_1 or ν_2 we get the following expression for the third order polarization.

$$P^{(3)}(z,t) = \exp i \nu_1 t \left[P_1 \sin k_1 z + P_1' \sin k_+ z \right] + \exp i \nu_2 t \left[P_2 \sin k_2 z + P_2' \sin k_- z \right]$$

+ complex conjugate

where

$$k_1 = \pi n_1/L \;,\; k_2 = \pi n_2/L \;,\; k_+ = 2k_2 - k_1$$
$$k_- = 2k_1 - k_2$$

and with $P_1 = i\pi^{1/2}/(2k\kappa)$

(a) $$\times \Big\{ \left[|\mu_{12}|^4 (N_2 - N_1)/(\gamma_1 \gamma_2) \right] E_1^3 \left[1 + \gamma_{12} \left[\gamma_{12} + i(\omega_{21} - \nu_1) \right]^{-1} \right]$$

(b) $$+ \left(|\mu_{12}|^4/\gamma_1 \right) E_1 E_2^2 \Big[(\gamma_{12}/\gamma_1)(N_2 - N_1) \left[\gamma_{12} + i(\omega_{21} - \nu_B) \right]^{-1}$$
$$+ (\gamma_{13}/\gamma_3)(N_3 - N_1) \left[\gamma_{13} + i(\omega_{31} - \nu_B) \right]^{-1} \Big]$$

(c) $$+ \left(|\mu_{12}\mu_{31}|^2/2\gamma_1 \right)(N_3 + N_2 - 2N_1) E_1^3 \left[\gamma + i(\omega_B - \nu_1) \right]^{-1}$$

(d) $$+ \left(|\mu_{12}\mu_{31}|^2/\gamma_1 \right)(N_3 + N_2 - 2N_1) E_1 E_2^2 \left[2\gamma + i(\omega_B - \nu_B) \right]^{-1}$$

(e) $$+ \left(|\mu_{12}\mu_{31}|^2/\gamma_1 \right)(N_3 - N_1) E_1 E_2 \left[2\gamma + i(\omega_{32} - \Delta) \right]^{-1}$$
$$\times \left[1 + \gamma_1 \left[\gamma_{32} + i(\omega_{32} - \Delta) \right]^{-1} \right] \Big\}$$

(f) $$P_1' = \left[i\pi^{1/2}/(4k\kappa) \right] E_1^2 E_2 \left[\gamma_{12} + i(\omega_{21} - \nu_1) \right]^{-1} \left[\gamma_{32} + i(\omega_{32} - \Delta) \right]^{-1}$$

$$P_2 = P_1 \left\{ \begin{array}{l} \text{interchange 2, 3 in N, } \omega, \mu, \\ \text{interchange 1, 2 in E, } \nu \end{array} \right\} ; \quad P_2' = P_1' \left\{ \begin{array}{l} \text{same} \\ \text{interchange} \end{array} \right\}$$

$$\nu_B = (\nu_1 + \nu_2)/2 \;, \quad \omega_B = (\omega_{21} + \omega_{31})/2 \qquad (33)$$

4b. Discussion

The resonant terms in the expression (33) may, as discussed previously, be divided into two classes. The first class are the terms which do not depend on the standing wave nature of the field. These are the terms in P_1 and P_2 represented by line (e). They are identical in form with the resonant terms derived in 2a. In particular, their resonant form does not depend on the exact optical frequencies ν_1 and ν_2, only on the difference frequency $\Delta = \nu_2 - \nu_1$

The other class of resonant terms consists of the ones which arise because of the standing wave nature of the field. It can be seen in (33) that all of those resonant forms do depend on the exact optical frequency ν_1 or ν_2. The distinction between the frequency dependencies of the two classes is of experimental importance for the reasons discussed at the beginning of section 2b.. Observation of the class which depends on standing wave features would require a more elaborate laser system to achieve necessary frequency stability of ν_1 and ν_2. A more conventional laser with less frequency stability averages out the standing wave class of effects but is generally quite adequate for observing effects of the other class (those appearing in line (e) of eqn. (33)).

It should also be noted that, in contrast to the other class, the width of the standing wave class of resonances depends on the natural width of the lower, as well as the upper levels.

The standing wave resonance terms may be described, with reference to the line designations (and their counterparts in the interchange instructions), in (33) as follows :

(a) The resonance is the usual Lamb dip effect. e. g. when $\nu_1 = \omega_{21}$ atoms of zero velocity resonate with both travelling wave components at ν_1.

(b) The resonance occurs when the frequencies of the two applied fields are symmetrically located with respect to ω_{21} (or ω_{31}). In this case the same atoms can resonate on ω_{21} (or ω_{31}) with the right travelling component at ν_2 and the left at ν_1.

(c) One of the applied frequencies (e. g. ν_1) is halfway between ω_{21} and ω_{31}. In this case the left travelling component at ν_1 is Doppler shifted to ω_{31} for atoms of proper velocity, and for the same atoms the right travelling component at ν_1 is Doppler shifted to ω_{21}.

(d) The frequency midway between ν_1 and ν_2 is also midway between ω_{21} and ω_{31}. In this case, the right travelling component at, for example, ν_1 is Doppler shifted to ω_{31} and the left travelling component of ν_2 is Doppler shifted to ω_{21}.

(f) This term is a complicated term representing a combination of Lamb dip and Raman effects. When $\nu_1 = \omega_{21}$ and $\nu_2 = \omega_{32}$ (which implies $\Delta = \omega_{32}$), then several bands in the atomic velocity distribution overlap. The Lamb dip effect is due to travelling components in different directions at the same frequency, while the Raman effect is due to travelling components in the same direction at different frequencies. Note that these terms vary at a frequency of one of the applied fields (ν_1 or ν_2) but their spatial variation is characterized by an allowed k vector which is not that of an applied field. Thus the absorption or emission due to this term in the polarization depends on where the material is placed within the resonator. Also the effect of this term becomes small when the length of the sample becomes an appreciable part of the cavity length.

The power amplified or absorbed due to the polarization (33) is found from (22), with the electric field given by (28). If, as in the travelling wave case, we consider the average power absorbed in a time long compared to $1/(\nu_2 - \nu_1)$, then we may consider the effect of the fields at frequencies ν_1 and ν_2 separately. In particular we again need the component of the polarization at frequency ν_1 (or ν_2) which is out of phase with the electric field at that frequency. Let us take for these out of phase parts of the third order polarization

$$S(z,t) = \sin \nu_1 t \left\{ \hat{S}_1 \sin k_1 z + \hat{S}_1' \sin k_+ z \right\} + \sin \nu_2 t \left\{ \hat{S}_2 \sin k_2 z + \hat{S}_2' \sin k_- z \right\}$$

as before $k_1 = \pi n_1/L$, $k_2 = \pi n_2/L$, $k_+ = 2k_2 - k_1$, $k_- = 2k_1 - k_2$ (34)

where $\hat{S}_1, \hat{S}_1', \hat{S}_2, \hat{S}_2'$ are obtained from (33). The general form of these expressions will not be needed in the following arguments. Explicit reference will be made only to the resonant terms involving $(\omega_{32} - \Delta)$ and for these $\hat{S}_1 = S_1 E_1 E_2^2/4$; $\hat{S}_2 = S_2 E_1^2 E_2/4$ where S_1, S_2 were calculated for travelling wave case and given by (26) and (27).

The amplification or absorption at frequency ν_1 (aside from the linear part) is thus given by

$$\overline{W}_1 = \nu_1 E_1 A \int_{a-\ell/2}^{a+\ell/2} (\hat{S}_1 \sin k_1 z + \hat{S}_1' \sin k_3 z) \sin k_1 z \, dz \tag{35}$$

where a is the location of the center of the sample with respect to the end of the resonator, ℓ is the length of the sample, L is the length of the cavity. A similar

expression may be obtained for the power emitted or absorbed, $\overline{W}_2$, at frequency ν_2. It is convenient for later reference to rewrite (35) and the similar equation for frequency ν_2 symbolically as

$$\overline{W}_1 = f_1(\omega)E_1^4 + \xi_{12}(\omega)E_1^2E_2^2$$

$$\overline{W}_2 = \xi_{21}(\omega)E_1^2E_2^2 + f_2(\omega)E_2^4 \tag{36}$$

Identification of the terms in (36) with (35) and its counterpart in ν_2 defines the quantities $f_1(\omega)$, $f_2(\omega)$, $\xi_{12}(\omega)$, $\xi_{21}(\omega)$. Again, the explicit form of the expressions will not be required here. We have written in the ω dependence to emphasize that the quantities are sharp functions of ω_{21}, ω_{31}, ω_{32}, for given ν_1 and ν_2.

First consider the case that an active laser material itself is, for instance, placed in a magnetic field which tunes the level spacings. Assume that a suitable set of three tunable energy levels can be isolated.

The nonlinear polarization of the active medium governs the steady state power emitted from the laser.[3] The resonances in the third order polarization will therefore manifest similar resonances in the output power. From an observation of the output power[14] as a function of magnetic field strength, then, one can, using the general theory of two mode laser behavior, work back to obtain values of parameters appearing in eqn. (33).

In practice the above experiment is a good one to observe the resonant effects qualitatively. Its details, however, involve additional complications because the laser output is not directly proportional to the non-linear polarization parameters. Instead, the functional dependence of the output power on the non-linear

polarization terms appears through a set of straightforward but algebraically complicated expressions[3] For this reason, a simple Lorentzian dependence appearing in the polarization terms, manifests itself in the output power, in a way which is highly distorted and non-Lorentzian. This fact can make working out the parameters of the polarization difficult. The procedure described below, however, avoids these complexities.

Consider, a case in which a cell containing a sample gas is placed within the laser resonator in addition to the active laser material. Suppose the sample gas but not the laser material, to be subjected to a magnetic field. It is shown in appendix D that under certain conditions the sample may be considered a small perturbation on the operating conditions of the laser. In particular this is the case when the sample saturates less readily than the active laser material. It is also true if the sample saturates comparable with the laser material, providing only a small quantity of the sample is used. Under these conditions it is shown in appendix D that the change in power output due to the non-linear response of the sample is given by

$$\delta P_{out} = B\left[f_1(\omega)E_{10}^2 + \xi_{12}(\omega)E_{20}^2\right] + D\left[\xi_{21}(\omega)E_{10}^2 + f_2 E_{20}^2\right] \tag{37}$$

where E_{10} and E_{20} are the unperturbed field strengths in the cavity ← obtained in the absence of non-linear coupling to the sample. D and B are parameters which relate only to the behavior of the laser under completely general operating conditions (that is, even when the laser operates well above threshold and the active material is no longer desirable by standard perturbation theories). Equation (37) shows that, for small tuning of the sample, the resonant behavior in the polarization will be directly observable as a change in laser output power.

It is interesting and, for reasons we have discussed, experimentally important to consider a case in which the frequencies ν_1 and ν_2 fluctuate slowly by amounts greater than the natural linewidths of the sample gas. As we have repeatedly emphasized, in this case only the resonance in $\omega_{32} - \Delta$, which does not depend on the exact value of ν_1 and ν_2, will be observable.

Suppose that the fluctuations in ν_1 and ν_2 due to thermal effects or microphonics are stationary (in the statistical sense). Let the highest frequency of modulation of ν_1 or ν_2 be $\delta\nu_f$. While we do not restrict the modulation amplitude (except insofar as the laser should keep oscillating in two modes), we shall require that $\delta\nu_f$ be much less than the three quantities, γ_a, $\nu_2 - \nu_1$ and $1/\tau_L$. Here γ_a is the smallest of the three natural linewidths in the sample gas and τ_L is the characteristic time for the laser to reach a steady state. Clearly an atom will see a "fluctuating single frequency" rather than a broad spectrum only if $\delta\nu_f \ll \gamma_a$. This will usually be the smallest of the three quantities. If ν_1 and ν_2 are closely spaced modes of the Fabry-Perot cavity then $\nu_2 - \nu_1$ and $1/\tau_L$ will be about the same order of magnitude. Both are related to the transit time of light across the cavity. Under these conditions, we may still apply all of the considerations leading up to

equation (37) and that equation will still hold but all quantities appearing in it will be random functions of time.

Let us now consider that some standard sort of noise smoothing circuitry is associated with the detector. The dependence of detected power on ω (magnetic field) can then be obtained by taking the time average of both sides of equation (34) over a time, τ, long compared to important fluctuation periods and of the order of the (adjustable) integration time of the detector.

We will now make our only assumption in this sample cell case about the nature of the active laser material. Let us assume that it has a symmetric atomic lineshape (remember it is not in the magnetic field). Then, there should be no distinction in the behavior of the laser at the frequencies ν_1 and ν_2 on the average. In particular, we must have in eqn. (37) $AE_{20}^2 = BE_{10}^2$.[15] We thus arrive at a simple expression for the overall resonant behavior of the sample with magnetic field. We have, from (38), for the field dependent part of the output power

$$\delta P_{out} = Const\left[\xi_{12}(\omega) + \xi_{21}(\omega)\right] \tag{38}$$

where now $\xi_{12}(\omega)$ and $\xi_{21}(\omega)$ refer only to the terms in $(\omega_{32} - \Delta)$, the only terms which retain a tunable behavior after averaging. In fact, for our present result we may set $\xi_{12} + \xi_{21} = (A\ell\nu/3)(S_1 + S_2)$ where S_1 and S_2 are the quantities given by eqns (26) and (27) That is to say the lineshape in the present case is identical to that in the case of two travelling waves.

This result enables one to take advantage of high field intensities within the resonator of a conventional gas laser in order to induce sizable non-linear

polarization. But at the same time, the simplicity of the frequency behavior exhibited in the travelling wave case is retained.

It should also be pointed out that the terms involving the standing wave nature of the field as described above are also of considerable interest. They could sizably influence the behavior of a laser operating between energy levels involving splittings. Their observation, however, requires frequency stability of the type needed in previous experiments with the Lamb dip effect in a single mode laser.[16]

APPENDIX A.

The general form of the density matrix equations with respect to the unperturbed states is

$$\left(\frac{d}{dt} + i\omega_{kj} + \gamma_{kj}\right)\rho_{kj} = -\sum_m iV(t)\left(\mu_{km}\rho_{mj} - \rho_{km}\mu_{mj}\right) \tag{A1}$$

Multiplying both sides by $\exp(i\omega_{kj} + \gamma_{kj})t$, the left side becomes the total derivative $(d/dt)\exp(i\omega_{kj} + \gamma_{kj})t$ On defining $\sigma_{kj} = \exp(i\omega_{kj} + \gamma_{kj})t$ and using $\omega_{kj} = \omega_k - \omega_j$ (A1) gives

$$\frac{d\sigma_{kj}}{dt} = -\sum_m iV(t)\left[\mu_{km}\sigma_{mj}e^{i\omega_{km}t}e^{(\gamma_{kj}-\gamma_{mj})t} - \sigma_{km}\mu_{mj}e^{i\omega_{mj}t}e^{(\gamma_{kj}-\gamma_{km})t}\right] \tag{A2}$$

which leads to eqns (11).

APPENDIX B.

To show that it is sufficient to consider atoms starting in state 2, we first note that equations (11a-11f) are invariant if we change all twos to threes, threes to twos, and take complex conjugates. Therefore, we can obtain the solution of (11) for an atom which starts in state 3(α = 3) from the solution for an atom which starts in state 2 by the same interchanging and complex conjugation.

Next we note that if we had let the atom start in state 1 in the calculation of our typical term, there could be an overall change in sign and the factor exp $\gamma_2(t_0-t''')$ in eqn. (13) would be replaced by exp γ_1 (t_0-t'''). This is due to the different factors multiplying σ_{11} and σ_{22} in (11a) and the different initial condition (12). In integrating over initial times we would then get (15) except with γ_1 in the denominator and an overall sign change. On multiplying by $\Lambda_1(\nu)$, this would give $-N_1(v)$ replacing $N_2(v)$ in (17). On considering effects of atoms starting in both states we may, therefore, simply let $N_2(v)$ go to $N_2(v) - N_1(v)$. It is clear this result depends only on the initial condition and first order approximation and therefore holds for any third order term. Further, from the symmetry of state 2 and 3 described above or from considering (11b) and (12) we get the complete effect of level one by also letting $N_3(v) \rightarrow N_3(v) - N_1(v)$.

We will solve (11) and (12) for all third order terms in ρ_{12} and ρ_{31} in table 1 in the following shorthand notation. In the table below: we write $\sigma_{ij}^{(3)}\sigma_{lm}^{(2)}\sigma_{st}^{(1)}$ to represent the third order term in σ_{ij} which comes about because a first order σ_{st} produces a second order σ_{lm}. (In all cases the atom is assumed to start in state 2). Underneath this term designation we write on three successive lines (with reference to the typical calculation in the text, eqn. (13)) the factors multiplying t''', t'' and t' respectively, in exponents appearing in the triple integrals. Underneath, on the next three successive lines,

we write the denominator of the three complex Lorentzians which will occur after performing the integrals over t', t'', t'''. We leave the Doppler shift understood in writing these—that is, we write γ_μ to mean $\gamma_\mu - kv$. Finally on the next line, (with reference to eqn. (16) we write the multiplicative factor and the frequency dependence of the polarization. In every case, the product of three E's, the factor $iN_2(v)$, summation over indices and spatial variation will be left understood.

It can be noted from the signs of the frequency terms on lines 4 and 6, where the velocity enters with different sign in the travelling wave case. These are the important contributions to overall polarization, as discussed in the text. Contributions come from saturations terms $\sigma_{12}^{(3)}\sigma_{22}^{(2)}\sigma_{21}^{(1)}$, $\sigma_{12}^{(3)}\sigma_{11}^{(2)}\sigma_{21}^{(1)}$, $\sigma_{31}^{(3)}\sigma_{11}^{(2)}\sigma_{12}^{(1)}$ and from Raman term $\sigma_{31}^{(3)}\sigma_{32}^{(2)}\sigma_{12}^{(1)}$ leading to eqn. (21).

In the case of standing waves all terms may contribute, as discussed in the text.

APPENDIX C.

We are interested in integrals of the form

$$I_+ = \frac{1}{\pi^{1/2} u} \int_{-\infty}^{\infty} \frac{dv \exp - v^2/u^2}{(\gamma_a + \iota x + \iota k v)(\gamma_b + \iota y + \iota k v)} \tag{C1}$$

and

$$I_- = \frac{1}{\pi^{1/2} u} \int_{-\infty}^{\infty} \frac{dv \exp - v^2/u^2}{(\gamma_a + \iota x + \iota k v)(\gamma_b + \iota y - \iota k v)} \tag{C2}$$

where γ_a and γ_b stand for any $\gamma_{\iota j}$ in the text, and where x and y stand for any of $\pm(\omega_{21} - \nu_1)$ or $\pm(\omega_{31} - \nu_2)$.

It is asserted in the text that if $|x|$, $|y| \ll ku$, then I_+ is less I_- by a factor of order γ/ku. Before going into detailed calculations (in which we will not place the above restriction on $|x|$ and $|y|$) let us consider the reason for the result in the text. For simplicity let us consider the special case $x = y = 0$ and $\gamma_a = \gamma_b$ Then we have

$$I_+ = \frac{1}{\pi^{1/2} u} \int_{-\infty}^{\infty} \frac{dv \exp - v^2/u^2}{(\gamma_a + \iota k v)^2} \tag{C3}$$

$$I_- = \frac{1}{\pi^{1/2} u} \int_{-\infty}^{\infty} \frac{dv \exp - v^2/u^2}{\gamma_a^2 + k^2 v^2} \tag{C4}$$

We can write the denominator in the integrand of I_+ as $(\exp - 2\iota\varphi(v))/(\gamma_a^2 + k^2 v^2)$ where $\varphi(v) = \pm \tan^{-1}(kv/\gamma_a)$. The integrands of I_+ and I_-, thus, have the same

magnitude and differ only by the phase factor exp-($2\iota\varphi$ (v)). We may, therefore, say that atoms of different velocity contribute in phase with each other to I_- but in varying phase to I+. For example when kv= γ_a (and since ku $>> \gamma_a$) the magnitudes of the integrands have been reduced by 1/2 but the varying phase has pulled the real part of the integrand of I_+ to zero. As kv increases from γ_a, I_- builds up further (albeit slowly), but the phase in I_+ has reversed and it decreases slowly.

We will need to consider an integral of the form

$$H(\eta, \xi) = \frac{1}{\pi^{1/2}} \int_{-\infty}^{\infty} \frac{\exp -t^2}{\eta + \iota\xi + \iota t} \tag{C5}$$

for small values of η. The function H as defined is equal to - Z(z) where $z = -\xi + \iota\eta$ and Z is the tabulated plasma dispersion function. To evaluate H for small η we write

$$\frac{1}{\eta + \iota\xi + \iota t} = \int_0^{\infty} d\lambda \, e^{-(\eta + \iota\xi)\lambda} e^{-\iota t \lambda} \tag{C6}$$

and interchange the order of the t and λ integrations. We have, then

$$H(\eta, \xi) = \int_0^{\infty} d\lambda \, e^{-(\eta + \iota\xi)\lambda} \exp -\lambda^2/4 \tag{C7}$$

which to zero order in η gives

$$H(0,\xi) = \int_0^\infty d\lambda \exp(i\xi\lambda - \lambda^2/4)$$

$$= \exp(-\xi^2)\left[\pi^{1/2} - 2i\int_0^\xi \exp(\beta^2)\,d\beta\right. \tag{C8}$$

We consider I_- first. We expand the denominator in partial fractions

$$\frac{1}{(\gamma_a + ix + ikv)}\,\frac{1}{(\gamma_b + iy - ikv)} = \frac{1}{\gamma_a + \gamma_b - i(x+y)}$$

$$\times\left[\frac{1}{\gamma_a + ix + ikv} + \frac{1}{\gamma_b + iy - ikv}\right] \tag{C9}$$

Then

$$I_- = \frac{1}{\gamma_a + \gamma_b + i(x+y)}\left[\frac{1}{\pi^{1/2}u}\int_{-\infty}^{\infty}\frac{dv\,\exp(-v^2/u^2)}{(\gamma_a + ix + ikv)} + \frac{1}{\pi^{1/2}u}\int_{-\infty}^{\infty}\frac{dv\,\exp(-v^2/u^2)}{(\gamma_b + ix - ikv)}\right.$$

(C10)

In the first integral in (C10) we let $t = v/u$, and identify γ_a/ku with η and x/ku with ξ in (C8). In the second integral in (C10) we let $t = -v/u$ and identify γ_b/ku and y/ku with η and ξ respectively in (C8). We, thus, arrive at an expression to lowest order in γ_a/ku and γ_b/ku.

$$I_- = \frac{1}{ku\left[\gamma_a+\gamma_b+i(x+y)\right]}\left[\pi^{1/2}\,\mathrm{erfc}\,-(x/ku)^2 + \pi^{1/2}\,\mathrm{erfc}\,-(y/ku)^2 - 2i\int_0^{x/ku}\mathrm{erfc}(\beta^2)\,d\beta - 2i\int_0^{y/ku}\mathrm{erfc}(\beta^2)\,d\beta\right] \tag{C11}$$

This expression clearly reduces to eqn. (20) in the text for $|x| \ll ku$, $|y| \ll ku$.

In the travelling wave case, it may be noted (from eqns. (18) and (19) in the text) that in all terms which lead to an integral of the I_- type we have either $x = -y$ or $x + y = \omega_{32} - \Delta \ll ku$. In either of these cases we may write C(11) simply as

$$I_- = \frac{2\pi^{1/2}}{ku}\,\frac{\mathrm{erfc}\,-(x/ku)^2}{\left[\gamma_a+\gamma_b+i(x+y)\right]} \tag{C12}$$

Now we consider I_+. Suppose first $\gamma_a \neq \gamma_b$ or $x \neq y$. We again expand the denominator of the integrand in partial fractions. We again let $t = v/u$ and use the definition (C5) to arrive at

$$I_+ = \frac{1/(ku)^2}{\left[(\gamma_a-\gamma_b)+i(x-y)\right]/ku}\left[H\left(\frac{\gamma_a}{ku},\frac{x}{ku}\right)-H\left(\frac{\gamma_b}{ku},\frac{y}{ku}\right)\right] \tag{C13}$$

Suppose $|x - y|/ku \ll 1$. It may may be verified in eqns. (18) and (19) that for $\omega_{32}-\Delta \ll ku$, this is always true in the travelling wave case for terms which lead to an I+ type integral. (It is also true in any case, of course, if $|x|/ku$ and $|y|/ku$ themselves are small.)

H is an analytic function of the complex variable $p = \eta + i\xi$ (as may be easily verified from the form (C7) in particular) for all positive η. We may write (C13) to order γ_a/ku, γ_b/ku and $(x - y)/ku$ as

$$I_+ = \frac{1}{(ku)^2}\left(\frac{dH}{dp}\right)_{(\gamma_a/ku)+i(x/ku)} \tag{C14}$$

If we handle the special case $\gamma_a = \gamma_b$, $x = y$ separately we clearly get the same result. It is well known in the theory of complex variables $dH/dp = -i\,\partial H/\partial\xi$. To zero order in γ_a/ku we can, therefore, simply differentiate the expression (C8) and the result is

$$I_+ = \frac{2}{(ku)^2}\left\{1 + 2\xi\exp(-\xi^2)\int_0^{\xi}\exp(\beta^2)\,d\beta + i\pi^{1/2}\xi\exp(-\xi^2)\right\}$$

where $\xi = x/ku$ (C15)

The expression in brackets in (C15) is of order one for ξ of order one. In the travelling wave case, comparing (C12) and (C15) (noting x + y in (C12) is small) we see that I_+ is less than I_- by order γ/ku even if $|x|$ and $|y|$ are comparable to ku. Thus, in the travelling wave case we may more generally replace (20) in the text by (C12), the result on (21) in the text being the overall multiplicative factor $\exp -(x/ku)^2$.

In the standing wave experiment since (C15) is valid for $|x|$ and $|y| \ll ku$ we see that in that case I_- is less than I_+ by order γ/ku. For standing waves if $|x|$ and $|y|$ are comparable to ku, it is necessary, for some terms, to use the more general form (C11) of I_- and to re-evaluate I_+ for $x \neq y$. However, it may be easily verified that this is not necessary for those terms whose resonant form depends only on $\omega_{32} - \Delta$ and not on ν_1 or ν_2 directly. Those resonant forms are again the same as in the case of two travelling waves.

Finally we discuss the third type of integral which came up in the standing wave case.

This integral was of the form

$$I_3 = \frac{1}{\pi^{1/2} u} \int_{-\infty}^{\infty} dv \, \exp -v^2/u^2 \int_0^{\infty} d\tau' \int_0^{\infty} d\tau'' \int_0^{\infty} d\tau''' \, \exp -(\gamma_a + i x)\tau'$$

$$\exp -(\gamma_b + i\theta)\tau'' \exp -(\gamma_c + i y)\tau''' \exp i k v (\tau' + 2\tau'' + \tau''') \tag{C16}$$

We will point out how this is evaluated without giving the details. As before we will assume $|x - y| \ll ku$. We also assume $|\theta| \ll ku$ (θ is either 0 or near ω_{32} in every important case). First one changes variables in (C16) to

$$\beta_1 = \tau' + \tau''', \quad \beta_2 = \tau' + 2\tau'' + \tau''', \quad \beta_3 = \tau' - \tau'''$$

Then

$$\int_0^\infty \int_0^\infty \int_0^\infty d\tau' \, d\tau'' \, d\tau''' \longrightarrow \frac{1}{4} \int_0^\infty d\beta_1 \int_{-\beta_1}^{\beta_1} d\beta_3 \int_{\beta_1}^\infty d\beta_2$$

The velocity integration can be done in terms of β_2. One then changes the order of integration and does the integrals involving β_1 and β_3. Next, advantage is taken of the smallness of $|x - y|/ku$ and $|\theta|/ku$ to write the results in terms of derivatives of the H function, similar to the procedure used for I_+. The result, just as the I_+ case, is proportional to $1/(ku)^2$ and is thus less than I_- by order γ/ku.

APPENDIX D.

We discuss,here, the calculation of observed output power when, in addition, to the active laser material, a sample cell of material is placed within the laser cavity as per the discussion of the later part of section 4.

The steady state amplitude of the electric field E_μ in the mode μ can be determined by setting equal the net power gained due to polarization currents in the atomic media and the power lost within the optical cavity—both gain and loss averaged over a time long compared to $1/(\nu_2 - \nu_1)$.

The components of the polarization in the sample gas which time vary out of phase with the electric field are taken as

$$S_1 = \left[2\eta_1 E_1 + f_1(\omega) E_1^3 + \xi_{12}(\omega) E_1 E_2^2 \right] \sin \nu_1 t$$

$$S_2 = \left[2\eta_2 E_2 + \xi_{21}(\omega) E_1^2 E_2 + f_1(\omega) E_2^3 \right] \sin \nu_2 t \tag{D1}$$

for frequencies ν_1 and ν_2 respectively. Here $2\eta_\mu$ is the linear part of the susceptibility at frequency ν_μ, and the quantities $f(\omega)$ and $\xi(\omega)$ are defined in the text. Note that the frequency behavior of η_μ is characterized by the Doppler width.

For the laser material we take the relevant out of phase components of the polarization as

$$S_{L1} = E_1 \chi_1(E_1^2, E_2^2) \sin \nu_1 t$$

$$S_{L2} = E_2 \chi_2(E_1^2, E_2^2) \sin \nu_2 t \tag{D2}$$

We leave the form of the χ's unspecified. (The dependence on E_1^2 and E_2^2 follows from the symmetry of the material and the fact that both fields are in the same direction.) In other words, while we imply that a perturbation expansion in the field strengths is valid for the polarization of the sample gas, we make no such demand of the laser material. The laser could be operating well above its threshold.

The cavity power losses at frequency μ, from the definition of the quality factor, Q_μ, are given by

$$\text{loss}_\mu = \frac{\nu_\mu}{8\pi Q_u} E_\mu^2 LA \tag{D3}$$

Calculating the gain from (D2) and (D1) via (22) in the text, and at each frequency setting that equal to the loss (D3), we arrive at the following two equations.

$$\chi_1(E_1^2, E_2^2) + \xi_1(\omega)E_1^2 + \xi_{12}(\omega)E_2^2 = 2\left(\frac{1}{8\pi Q_1} - \eta_1\right)$$

$$\chi_2(E_1^2, E_2^2) + \xi_{21}(\omega)E_1^2 + \xi_2(\omega)E_2^2 = 2\left(\frac{1}{8\pi Q_2} - \eta_2\right) \tag{D4}$$

Equations (D4) implicitly determine E_1^2 and E_2^2. The power observed at the detector is the sum of the power coupled out at each frequency and is given by

$$P_{out} = \frac{TC}{8\pi}(E_1^2 + E_2^2) \tag{D5}$$

where T is the transmission coefficient of the mirror. (for an otherwise lossless cavity $Q_\mu = (\nu_\mu L/cT)$) The dependence of this power on magnetic tuning of the sample (for small tuning) is given by the ω dependence in (D4). We may always relate

detected power to stored energy in the cavity through equation (D5) and we will consider the two interchangably from now on. General solution of (D4) for the output power versus tuning would necessitate a knowledge of the laser χ's (a good representation of which is available only for laser operation near threshold) and would require considerable analysis to go from an observed power versus field behavior to derive the $\rho(\omega)$ and $\xi(\omega)$ from which natural linewidths and structure could be inferred. These complications are the regenerative effects referred to in the text.

Consider the case, however, that the last two terms on the left of equation (D4) are much smaller than the first term. Then we may consider the non-linear behavior of the sample as a small perturbation on the rest of the system. The conditions for validity of this are clearly those described in the text. In this case, we linearize equations (D4) in the small corrections, δE_1^2 and δE_2^2, to the result obtained by ignoring the small terms.

We have, then, to zero order in eqns. (41)

$$\chi_1(E_{10}^2, E_{20}^2) = 2\left(\frac{1}{8\pi Q_1} - \eta_1\right)$$

$$\chi_2(E_{10}^2, E_{20}^2) = 2\left(\frac{1}{8\pi Q_2} - \eta_2\right)$$

which determine the output power, $E_{10}^2 + E_{20}^2$ in the absence of saturation in the cell. To first order

$$\chi_1(E_{10}^2, E_{20}^2) + \left(\frac{\partial \chi_1}{\partial E_1^2}\right)_0 \delta E_1^2 + \left(\frac{\partial \chi_1}{\partial E_2^2}\right)_0 \delta E_2^2 + f_1(\omega) E_{10}^2 + \xi_{12}(\omega) E_{20}^2 = 2\left(\frac{1}{8\pi Q_1} - \eta_1\right)$$

$$\chi_2(E_{10}^2, E_{20}^2) + \left(\frac{\partial \chi_2}{\partial E_1^2}\right)_0 \delta E_1^2 + \left(\frac{\partial \chi_2}{\partial E_2^2}\right)_0 + \xi_{21}(\omega) E_{10}^2 + f_2(\omega) E_{20}^2 = 2\left(\frac{1}{8\pi Q_2} - \eta_2\right)$$

(D6)

giving two linear equations for the change in power as $\delta E_1^2 + \delta E_2^2$.

$$\left(\frac{\partial \chi_1}{\partial E_1^2}\right)_0 \delta E_1^2 + \left(\frac{\partial \chi_1}{\partial E_2^2}\right)_0 \delta E_2^2 = -\left[f_1(\omega) E_{10}^2 + \xi_{12}(\omega) E_{20}^2\right]$$

$$\left(\frac{\partial \chi_2}{\partial E_1^2}\right)_0 \delta E_1^2 + \left(\frac{\partial \chi_2}{\partial E_2^2}\right) \delta E_2^2 = -\left[\xi_{21}(\omega) E_{10}^2 + f_2(\omega) E_{20}^2\right]$$

(D7)

The solution of these equations for $\delta E_1^2 + \delta E_2^2)$ is given by eqn. (37) in the text, namely

$$\delta(E_1^2 + E_2^2) = B\left[f_1(\omega) E_{10}^2 + \xi_{12}(\omega) E_{20}^2\right] + D\left[\xi_{21} E_{10}^2 + f_2(\omega) E_{20}^2\right]$$

with B =

$$-\frac{\left[\left(\frac{\partial \chi_2}{\partial E_2^2}\right)_0 - \left(\frac{\partial \chi_2}{\partial E_1^2}\right)_0\right]}{\left[\left(\frac{\partial \chi_1}{\partial E_1^2}\right)_0\left(\frac{\partial \chi_2}{\partial E_2^2}\right)_0 - \left(\frac{\partial \chi_1}{\partial E_2^2}\right)_0\left(\frac{\partial \chi_2}{\partial E_1^2}\right)_0\right]}$$

and D =

$$-\frac{\left[\left(\frac{\partial \chi_1}{\partial E_1^2}\right)_0 - \left(\frac{\partial \chi_1}{\partial E_2^2}\right)_0\right]}{\left[\left(\frac{\partial \chi_1}{\partial E_1^2}\right)_0\left(\frac{\partial \chi_2}{\partial E_2^2}\right)_0 - \left(\frac{\partial \chi_1}{\partial E_2^2}\right)_0\left(\frac{\partial \chi_2}{\partial E_1^2}\right)_0\right]}$$

FOOTNOTES

1. See, for example, A. C. G. Mitchell and M. W. Zemansky, Resonance Radiation and Excited Atoms (reprinted by Cambridge Press, 1961).
2. M. S. Feld, J. H. Parks, H. R. Schlossberg and A. Javan in Proceedings of the Conference on Quantum Electronics, Puerto Rico (McGraw-Hill, New York, 1965).
3. W. E. Lamb, Jr. Phys. Rev., 134, A1429 (1965).
4. This condition implies that the difference in the Doppler widths of the 3-1 and 2-1 transitions is negligible. We will always consider the applied frequencies, ν_1 and ν_2, to be somewhere within the common Doppler width of ω_{21} and ω_{31} respectively. This means that those atoms, whose center of mass velocity, v, is such as to make them resonant with one of the applied fields, will Doppler shift ν_1 and ν_2 very nearly the same amount; i.e. $\nu_1 v/c \approx \nu_2 v/c$
5. A. Javan "Quantum Electronics and Coherent Light", (Proceedings of Int. School of Physics, "Enrico Fermi", 1963) ed. P. A. Miles and C. H. Townes (Academic Press, 1964).
6. At extremely high fields there can be some significant frequency pulling effects. A. Javan, Phys. Rev., 107 1579 (1957). Such high field strengths probably invalidate a perturbation calculation of the atomic response and we will not consider them here.
7. More precisely, of course, atoms in the ensemble are excited to mixed states at t_o and we are making the usual random phase assumption of the excitation mechanism in describing the states one at a time.

8. For convenience we take $\hbar = 1$ throughout.

9. The terms in $\gamma_{\ell f}$ in these equations could, of course, be simplified using the $\gamma_{\ell f} = (\gamma_\ell + \gamma_f)/2$ relation. We chose not to make this substitution at this point. In this way our equations may be taken over to cases where equations 10 holds but $\gamma_{\ell f}$ is arbitrary. In particular, this is true in many cases in solids.

10. This terminology may be somewhat confusing and requires explanation. If the frequencies of the two applied fields are off resonance (with respect to the natural width, for a particular atom) then the terms due to $\rho^{(2)}_{32}$ represent the direct transitions from level two to level three via the two quantum process discussed above. These effects are always called Raman effects. If, however, both applied frequencies are near resonance at the two optical transition frequencies, the $\rho^{(2)}_{32}$ terms are also responsible for a change of direct single quantum transition probability. That is, the probability of the field at frequency ν_1 causing a transition from level two to level one is altered by the presence of the field at frequency ν_2. This latter effect is like a usual Raman effect both in its frequency dependence and in its dependence on the product of the two applied intensities. With this in mind, we will continue, for convenience, to refer to terms due to $\rho^{(2)}_{32}$ as Raman terms.

11. We will refer to a term of the form $[\gamma + i x]^{-1}$ as a complex Lorentzian since its real part represents a Lorentzian lineshape. The term is resonant when the combination of frequencies represented by x equals zero.

12. We have written only those saturation terms here in which both complex Lorentzians are resonant for the same velocity atom.

13. This assertion is similar to the δ function approximation made by Lamb in ref. 3.

14. One could also observe these effects by looking from the side of the tube at spontaneous emission which involves one of the laser levels. Effects of this kind have been discussed by R. L. Fork, L. E. Hargrove and M. A. Pollack, Phys. Rev. Lett., 12 705 (1964).

15. This may be somewhat clearer from the expressions in appendix D.

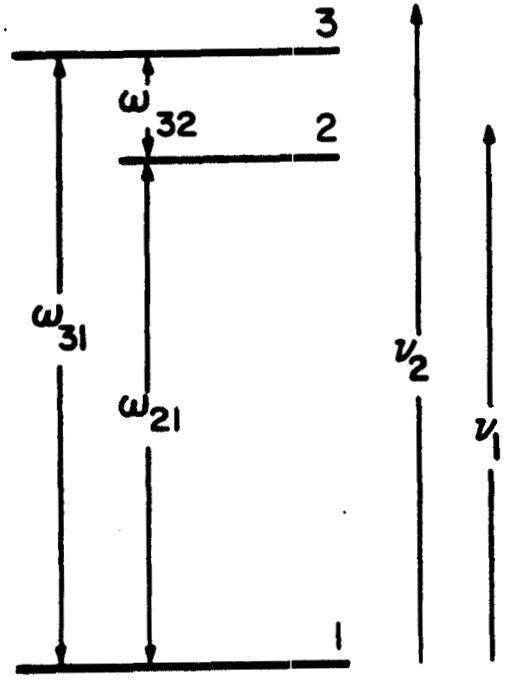
16. A. Szöke and A. Javan, Phys. Rev., to be published.

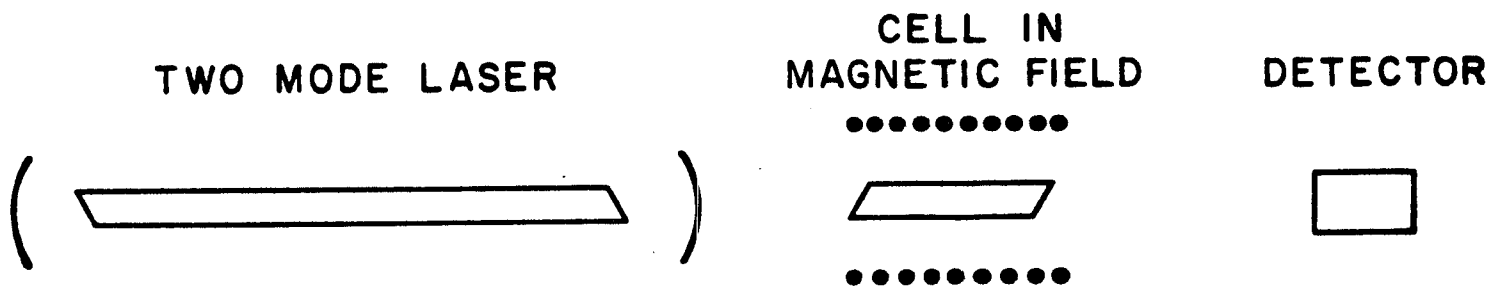
FIGURE CAPTIONS

Figure 1: Frequency spacings of the three atomic energy levels and two applied radiation fields--the system on which is based most of the discussion of this paper.

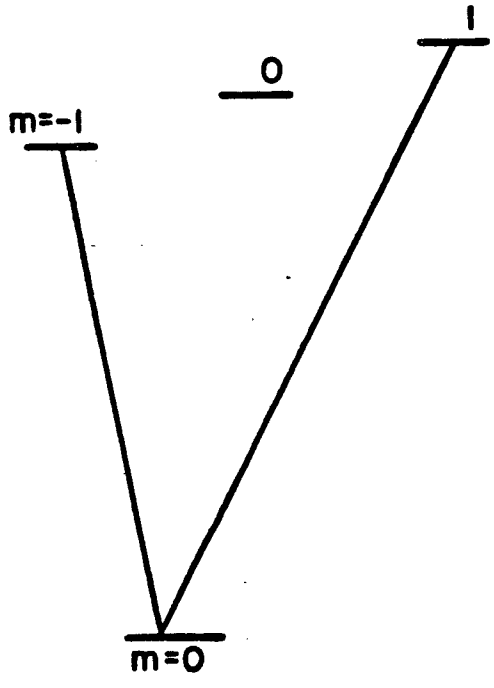
Figure 2: Simplified experimental arrangement for observation of effects discussed in section 2.

Figure 3: Energy level scheme showing Zeeman levels which are connected by matrix elements in an experiment as in figure 2. (a) J = 1 to 0 transition (b) J = 2 to J = 1 transition.

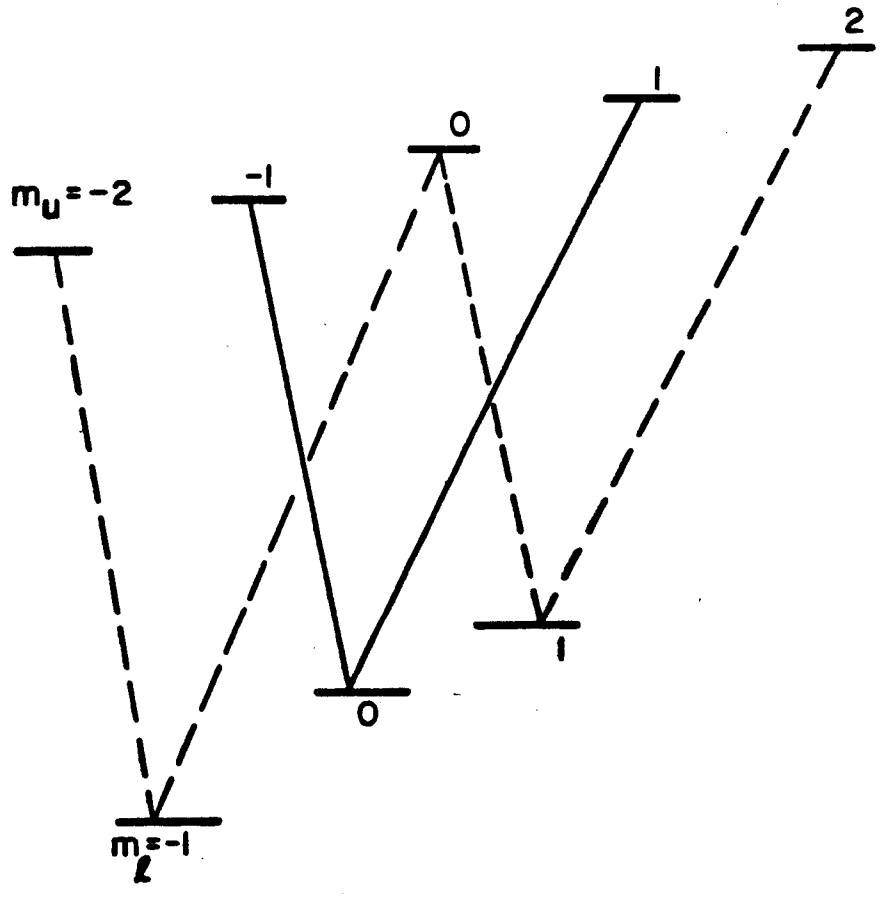

3
2
1
ω_{32}
ω_{31}
ω_{21}
ν_2
ν_1


TWO MODE LASER
CELL IN
MAGNETIC FIELD
DETECTOR

J = 0 TO J = 1



J = 2 TO J = 1



	$\sigma_{12}^{(3)}\sigma_{22}^{(1)}\sigma_{12}^{(1)}$	$\sigma_{12}^{(3)}\sigma_{22}^{(2)}\sigma_{21}^{(1)}$	$\sigma_{12}^{(3)}\sigma_{11}^{(2)}\sigma_{12}^{(1)}$	$\sigma_{12}^{(3)}\sigma_{11}^{(2)}\sigma_{21}^{(1)}$
(1)	$i(\nu_m-\omega_{21})+\gamma_{12}$	$i(-\nu_m+\omega_{21})+\gamma_{12}$	$i(\nu_m-\omega_{21})+\gamma_{12}$	$i(-\nu_m+\omega_{21})+\gamma_{12}$
(2)	$i(-\nu_p+\omega_{21})+\gamma_2-\gamma_{12}$	$i(\nu_p-\omega_{21})+\gamma_2-\gamma_{12}$	$i(-\nu_p+\omega_{21})+\gamma_1-\gamma_{12}$	$i(\nu_p-\omega_{21})+\gamma_1-\gamma_{12}$
(3)	$i(\nu_\sigma-\omega_{21})+\gamma_{12}-\gamma_2$	$i(\nu_\sigma-\omega_{21})+\gamma_{12}-\gamma_2$	$i(\nu_\sigma-\omega_{21})+\gamma_{12}-\gamma_1$	$i(\nu_\sigma-\omega_{21})+\gamma_{12}-\gamma_1$
(4)	$i(\nu_m-\omega_{21})+\gamma_{12}$	$i(-\nu_m+\omega_{21})+\gamma_{12}$	$i(\nu_m-\omega_{21})+\gamma_{12}$	$i(-\nu_m+\omega_{21})+\gamma_{12}$
(5)	$i(-\nu_p+\nu_m)+\gamma_2$	$i(\nu_p-\nu_m)+\gamma_2$	$i(-\nu_p+\nu_m)+\gamma_1$	$i(\nu_p-\nu_m)+\gamma_1$
(6)	$i(\nu_\sigma-\nu_p+\nu_m-\omega_{21})+\gamma_{12}$	$i(\nu_\sigma+\nu_p-\nu_m-\omega_{21})+\gamma_{12}$	$i(\nu_\sigma-\nu_p+\nu_m-\omega_{21})+\gamma_{12}$	$i(\nu_\sigma+\nu_p-\nu_m-\omega_{21})+\gamma_{12}$
(7)	$\lvert\mu_{12}\rvert^4\angle i(\nu_\sigma-\nu_p+\nu_m)t$	$\lvert\mu_{12}\rvert^4\angle i(\nu_\sigma+\nu_p-\nu_m)t$	$\lvert\mu_{12}\rvert^4\angle i(\nu_\sigma-\nu_p+\nu_m)t$	$\lvert\mu_{12}\rvert^4\angle i(\nu_\sigma+\nu_p-\nu_m)t$

	$\sigma_{12}^{(3)}\sigma_{32}^{(2)}\sigma_{12}^{(1)}$	$\sigma_{31}^{(3)}\sigma_{11}^{(2)}\sigma_{12}^{(1)}$	$\sigma_{31}^{(3)}\sigma_{11}^{(2)}\sigma_{21}^{(1)}$	$\sigma_{31}^{(3)}\sigma_{32}^{(2)}\sigma_{12}^{(1)}$
(1)	$i(\nu_m-\omega_{21})+\gamma_{12}$	$i(\nu_m-\omega_{21})+\gamma_{12}$	$i(-\nu_m+\omega_{21})+\gamma_{12}$	$i(\nu_m-\omega_{21})+\gamma_{12}$
(2)	$i(-\nu_p+\omega_{31})+\gamma_{32}-\gamma_{12}$	$i(-\nu_p+\omega_{21})+\gamma_1-\gamma_{12}$	$i(\nu_p-\omega_{21})+\gamma_1-\gamma_{12}$	$i(-\nu_p+\omega_{31})+\gamma_{32}-\gamma_{12}$
(3)	$i(\nu_\sigma-\omega_{31})+\gamma_{12}-\gamma_{32}$	$i(-\nu_\sigma+\omega_{31})+\gamma_{13}-\gamma_1$	$i(-\nu_\sigma+\omega_{31})+\gamma_{13}-\gamma_1$	$i(-\nu_\sigma+\omega_{21})+\gamma_{13}-\gamma_{12}$
(4)	$i(\nu_m-\omega_{21})+\gamma_{12}$	$i(\nu_m-\omega_{21})+\gamma_{12}$	$i(-\nu_m+\omega_{21})+\gamma_{12}$	$i(\nu_m-\omega_{21})+\gamma_{12}$
(5)	$i(-\nu_p+\nu_m+\omega_{32})+\gamma_{32}$	$i(-\nu_p+\nu_m)+\gamma_1$	$i(\nu_p-\nu_m)+\gamma_1$	$i(-\nu_p+\nu_m+\omega_{32})+\gamma_{32}$
(6)	$i(\nu_\sigma-\nu_p+\nu_m-\omega_{21})+\gamma_{12}$	$i(-\nu_\sigma-\nu_p+\nu_m+\omega_{31})+\gamma_{13}$	$i(-\nu_\sigma+\nu_p-\nu_m+\omega_{31})+\gamma_{13}$	$i(-\nu_\sigma-\nu_p+\nu_m+\omega_{31})+\gamma_{13}$
(7)	$\lvert\mu_{12}\rvert^4\angle i(\nu_\sigma-\nu_p+\nu_m)t$	$-\lvert\mu_{12}\mu_{31}\rvert^2\angle i(-\nu_\sigma-\nu_p+\nu_m)t$	$-\lvert\mu_{12}\mu_{31}\rvert^2\angle i(-\nu_\sigma+\nu_p-\nu_m)t$	$-\lvert\mu_{12}\mu_{31}\rvert^2\angle i(-\nu_\sigma-\nu_p+\nu_m)t$

TABLE I Third order terms