MODULATION SPECTROSCOPY IN PARTIALLY COHERENT FIELDS OF ARBITRARY INTENSITY

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C E R T I F I C A T E

Certified that the work described in this thesis has been carried out by Ragini Saxena under my supervision. She has satisfied the requirements for the submission of Ph.D. thesis as laid down in the University Ordinances.

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TO

MY PARENTS

ABSTRACT

The thesis is devoted to the study of the spectral response of atoms or molecules to optical excitation by a weakly modulated source under a wide range of conditions such as those on homogeneous and inhomogeneous line widths, intensity of the field and temporal fluctuations of the field.

Chapter I serves to introduce the problem, tracing its previous history and outlining the need and relevance of the present study.

Chapter II contains a general formulation for obtaining the total intensity of the modulated fluorescence starting from the basic principles, and probes the relationship between the modulated fluorescences and the correlation function of the dipole moments. The type of information obtained by study of the intensity of the modulated fluorescence as a function of the modulation frequency is discussed.

Chapter III is devoted to the study of the modulated fluorescence for near-resonant excitation by a source of arbitrary bandwidth and intensity. This study is carried out within the framework of a general relaxation theory and takes into account the effects of Doppler broadening. The phaseshift of the modulated

fluorescence is shown to be particularly sensitive to the relaxation parameters, while the results of the previous studies are contained as limiting cases of our general theory. The position, width and weight of the resonances in the amplitude of modulation are explicitly discussed for intense, resonant fields. For weak fields, our results show that the behaviour of the velocity-averaged modulated fluorescence for monochromatic excitation is the same as that of broad-band excitation, provided the Doppler width is much larger than the natural linewidth, laser detuning and modulation frequency. Numerical plots of the velocity averaged amplitude of modulation for large atom-laser coupling and various Doppler widths reveal a shift in the resonance at the Rabi frequency for large Doppler widths.

In chapter IV, we investigate the Zeeman structure of the excited state in a $|J=0\rangle \leftrightarrow |J=1\rangle$ transition using modulated sources of arbitrary intensity and bandwidth. The shape as well as the width of the modulated Hanle signals is shown to be critically dependent on the bandwidth of the exciting source and its frequency of modulation. For weak fields, excitation by an incoherent, broad-band pump leads to the results of previous studies,

while excitation by a truly monochromatic source leads to entirely new results that are interpreted physically. For example, the modulation frequency scan of the modulated fluorescence for broad-band (monochromatic) excitation yields resonances at values 0, $\pm$ 2s ($\pm$ s), where 2s is the Zeeman splitting of the excited state. For intense fields, numerical studies for the amplitude of modulation exhibit resonances due to Autler-Townes effect at the modulation frequency values 0, $\pm \alpha_o$, $\pm 2\alpha_o$, where $\alpha_o^2 = s^2 + 2\alpha^2$, 2α being the Rabi frequency characterizing the atom-laser coupling.

Two photon processes are especially useful for studying transitions between two levels of same parity. Hence it would be interesting to study these processes using modulated sources, which is done so in chapter V for the case of stepwise excitation. The total intensity of the spontaneously emitted radiation from the upper (intermediate) level is studied as a function of the modulation frequency. It is shown that the resonances in the amplitude of modulation at the modulation frequency values 0, $\pm \alpha_o$, $\pm 2\alpha_o$ ($\alpha_o^2 = \alpha_1^2 + \alpha_2^2$, $2\alpha_1$ and $2\alpha_2$ being the Rabi frequencies characterising the coupling of the atom with the two laser fields) yield the energy spectra

of the composite system consisting of the atom and the intense, coherent laser field. These resonances are studied analytically using the secular approximation, which shows that the resonance at $\pm\alpha_0$ is the most intense one. In the presence of detuning of the applied field, this resonance splits into two closely-spaced structure, as may be seen by the numerical studies.

In chapter VI, the possibility of obtaining resolution beyond the natural linewidth using modulated sources is examined. It is indeed shown that if the amplitude of the exciting source is weakly modulated at a low frequency ν, then the fluorescence oscillating at 4ν exhibits a narrow resonance which is considerably less than the natural linewidth. This particular component can be detected separately by a phase sensitive detector. In contrast, the fluorescence oscillating at 2ν exhibits resonance having the natural linewidth.

CHAPTER - I

INTRODUCTION

Modulation spectroscopy has been extensively used as a tool for the measurement of atomic and molecular lifetimes by the Phaseshift Method.[1-5] This method is based on the fact that when a broad-band light source, whose amplitude is weakly modulated in time, is used to excite atoms or molecules, then the fluorescence is also modulated at the same frequency but with a phase displacement.[1-5] The phase shift Φ bears a simple relationship with the lifetime τ of the excited state, given by

$$\tan \Phi = - \nu \tau \qquad (1.1)$$

where ν is the modulation frequency, while the amplitude of modulation, A, varies as

$$A = (\nu^2 \tau^2 + 1)^{-\frac{1}{2}} \qquad (1.2)$$

With the advent of tunable, highly monochromatic dye lasers, it became necessary to study the problem in search of new effects that might arise in the laser induced modulated fluorescence. Such a study was first undertaken by Armstrong and Feneuille in 1975.[6] They found that the phaseshift is no longer independent of the strength of the excitation and even in the limit of weak laser intensities, it differed by a factor two i.e.

$$\tan \Phi = -2 \nu \tau \qquad (1.3)$$

while the amplitude of modulation now varied as

$$A = (4\nu^2 \tau^2 + 1)^{-\frac{1}{2}} \qquad (1.4)$$

For intense lasers, their study revealed that the amplitude of modulation displays a resonant behaviour as the modulation frequency is varied, this resonance being directly related to the Autler - Townes effect.[7] Their calculations were limited to weak modulation of the monochromatic laser, the applied field being exactly on resonance with the atomic transition. The theory was further generalized by including the detuning of the laser from the atomic resonance frequency by McClean and Swain,[8] who showed that even quite small detunings produce appreciable changes in the phaseshift. It is clear from equation (1.1) to (1.4), that the phaseshift and amplitude of the modulated fluorescence are extremely sensitive to the bandwidth of the exciting source. Hence a proper theory of the modulated fluorescence must take into account the finite bandwidth of the exciting source. One would also expect that the statistics of the field will change the modulation characteristics. Since the fluctuations of the exciting source (which give rise to its finite bandwidth) appear as relaxation effects in the spectral response of the atom,[9-14] one would also expect that other relaxation processes, such as collisions etc., would also influence the modulated fluorescence appreciably. Hence a complete theory

for the phenomenon should be evolved within the framework of a general relaxation theory. Such a study would be relevant for experiments where the samples are contained in vapor cells. The effect of Doppler broadening will also have to be considered. If the atoms/molecules are present in the form of a collimated beam, then the only relaxation mechanism to consider is spontaneous emission, and the results in this case may be easily obtained from the general theory. For experiments with atomic beams, the Doppler effect can be suppressed to first order by using mutually orthogonal directions for the atomic beam, excitation and detection.[15-19]

The fluorescence induced by a monochromatic laser whose amplitude is strongly modulated has been studied by Thomann[20] and by Feneuille et al.[21] The studies reveal that the atomic inversion exhibits resonances whenever the Rabi frequency is equal to an integral multiple of the modulation frequency, so that detection of the modulated fluorescence will yield the atom - laser coupling. However, since weak modulation of the excitation source leads to equally interesting and useful information about the atomic system,[1-6,22-24] we will concentrate our study on small modulation depths.

The Zeeman structure and the hyperfine structure of the excited state have been studied by examining the fluorescence in various directions.[25-33] This method is popularly known as Hanle effect (or zero magnetic field level-crossing experiment if a static magnetic field is used to lift the degeneracy of the excited state) or Optical Hanle effect[34-37] (if suitably polarized radiation is used to split the Zeeman sublevels by producing different light shifts). Excitation by modulated optical sources in the usual Hanle effect yields resonances[38] in the fluorescent light when the frequency of modulation is equal to the splitting of the Zeeman sublevels. The width of these resonances was determined by the lifetime of the excited state when the source used for preparing the atoms in a coherent superposition of the Zeeman sublevels was a weak, broad-band source.[22,23] In view of these important results, it is certainly desirable to study the laser-induced modulated Hanle signals for a source of arbitrary bandwidth and intensity.

One question that is of great importance is concerning the width of the structures in modulated fluorescence as generally the resolution of spectral lines of gases in the optical range has been limited by the Doppler effect which gives each line a relative width of a few GHz. Recently, several techniques have been proposed and used to improve the

linewidth in this respect,[39-41] besides the well known methods of atomic beam spectroscopy,[15-19] two photon spectroscopy[42-44] and saturated absorption spectroscopy.[45-48] Modulation spectroscopy can also be used to obtain Doppler free lines in gases, as was demonstrated by Sorem and Schawlow[24]. Two oppositely directed waves, which were intensity modulated at different frequencies, were irradiated on samples in cell. The narrow resonance of excited atoms is detected by recording the fluorescence intensity oscillating at the sum of the modulation frequencies. This method is shown to have a definite advantage when working with very weak transitions (with extremely small transition probabilities), with very low pressures (where the total absorption of the atoms or molecules is small) or with a sparsely populated lower state. In all the techniques discussed above, the resolution is limited by the natural linewidth. However several methods using transient response of atoms to pulsed excitation [49-56] have been discussed to obtain lines narrower than the natural linewidth. In view of the experiments of Sorem and Schawlow for natural-linewidth limited spectroscopy, it would be extremely tempting to explore the possibility of achieving resolution beyond the natural linewidth via fluorescence in modulated fields.

Two photon spectroscopy is a useful tool to investigate transitions between levels of the same parity, and at

the same time provides a highly efficient method to eliminate Doppler broadening.[42-44] In stepwise excitation, the detection of the intensity of the fluorescent light emitted spontaneously from the upper level reveals splitting due to the Autler-Townes effect.[57-61] A natural question that arises is what is the structure of the phase shifts and the amplitude of modulation in the optical double resonance situation and how the Rabi resonances appear in such experiments.

This thesis is devoted to the development of a general theory of modulated fluorescence[62,70] from a system under various conditions of excitation, environment of the system. In chapter II, we present a general formulation for the modulated fluorescence which will be used in subsequent chapters to study special cases depending on the geometry of the experiment and the nature of transitions. We also show for the first time the relationship between the modulated fluorescence and the correlation function of the atomic system.[cf.73-77]

In chapter III, we study the response of the system to a weakly-modulated light source of arbitrary bandwidth, the applied field being near resonance to a particular atomic transition. This study is made within the framework of a general relaxation theory to allow for non-radiative

transitions, and the effects of Doppler broadening are included. Our study reveals that the phaseshift of the modulated fluorescence is particularly sensitive to the relaxation parameters. The results (1.1) to (1.4) for broad-band and monochromatic excitation are recovered as limiting cases of our general theory, so that the difference of factor two in the phaseshift arises naturally when one takes into account the bandwidth of the exciting source. For intense resonant fields, the weight factor of the central peak at $\nu = 0$ reduces to zero, so that the amplitude of modulation displays only the resonances at the Rabi frequency due to Autler Townes effect.[7] For weak fields, our results show that the behaviour of the velocity averaged modulated fluorescence for monochromatic excitation is the same as that of broad-band excitation, provided the Doppler width is much larger than the natural linewidth, laser detuning and modulation frequency. Numerical plots of the velocity averaged modulated fluorescence for intense fields are presented, which reveal a shift in the resonance at the Rabi frequency for large Doppler widths. In chapter IV, we study modulated Hanle signals in partially coherent fields. The shape as well as the linewidth of the resonances observed in the modulated fluorescence are shown to be critically dependent on the bandwidth of the exciting source and its frequency of modulation. The results of a previous study of

the problem using weak, incoherent broad-band pump[22,23] are obtained as a limiting case of our analytical results for weak fields, while excitation by a truly monochromatic laser leads to entirely new results which are interpreted physically. For intense fields, numerical plots for the amplitude of modulation exhibit resonances due to Autler Townes effect.[7] In chapter V, we study two-pho on stepwise excitation in modulated fields. The total intensity of the spontaneously emitted radiation from the upper (intermediate) level is studied as a function of the modulation frequency. It is shown that the resonances in the amplitude of modulation yield the energy spectra of the composite system consisting of the atom and the coherent laser field. For example, resonances occur at the modulation frequency values $0, \pm \alpha_o$, $\pm 2\alpha_o$ due to dynamical Stark splitting[7] of the various energy levels (α_o is related to the Rabi frequency α_1 and α_2 characterizing the coupling of the atom with the two fields, by $\alpha_0^2 = \alpha_1^2 + \alpha_2^2$). These resonances are studied analytically using the secular approximation. In the presence of detunings of the applied fields, numerical results reveal additional structures. This should be contrasted with the zeroth order intensity of fluorescent light (unmodulated) which does <u>not</u> exhibit dynamic Stark splitting even when both the lasers are very intense, unless one studies the spectral resolution

of the emitted radiation. In chapter VI, we show for the first time how modulation spectroscopy can be used to obtain resolution beyond the natural linewidth. The study is made in the context of atomic beam experiments with orthogonal geometry to suppress Doppler broadening.[15-19] The fluorescence oscillating at four times the frequency of modulation of the amplitude of the exciting source is monitored as a function of the laser frequency. This reveals narrow resonance centred at the atomic frequency with a FWHM of the order of 1.3γ, which is considerably less than the natural linewidth of 2γ.

CHAPTER - II

GENERAL FORMULATION FOR THE MODULATED FLUORESCENCE

We are interested in the study of the fluorescence that is spontaneously emitted by atoms or molecules in vapor form when excited by lasers whose intensity is weakly modulated in time. The atoms (molecules) may be contained in a vapor cell or be present in the form of a collimated beam. At low pressures and small atomic densities, the interaction of the atoms amongst themselves may be neglected so that, in effect, we have to consider the interaction of a single, isolated atom or molecule with the laser beam. The field in the problem would thus be a prescribed field. The radiated field will, however, depend on the dynamics of the system. The free atom is excited by the intensity modulated laser beam and then radiates spontaneously due to its interaction with the vacuum of the radiation field. In this chapter, we give a general formulation for the modulated fluorescence emitted by the atom and will use it in subsequent chapters to study special cases depending on the geometry of the experiment and the nature of transitions. The total Hamiltonian for the atom and the radiation field is given along with the equation of motion for the reduced density operator

corresponding to the atomic system. The intensity of the modulated fluorescence is shown to be related to the atomic density matrix elements. A general method for solving the master equation to obtain the relevant density matrix elements is presented. We also show for the first time the relationship between the modulated fluorescence and the correlation function of the atomic system. The relation is reminiscent of the relationship of the spectrum of the unmodulated fluorescence to the correlation function of the dipole moments.[73-77] Hence the detection of the total intensity of the modulated fluorescence as a function of modulation frequency (say) will give the same type of information about the atomic system as the detection of the spectrum of the fluorescence emitted by the atom in the absence of modulation.

Section-2.1 : Hamiltonian

The total Hamiltonian for the atom and the radiation field in the presence of an external laser beam is

$$H = H_A + H_R + H_{AR} + H_{ext}(t) \qquad (2.1.1)$$

where H_A (H_R) is the unperturbed Hamiltonian of the atom (radiation field), H_{AR} ($H_{ext}(t)$) is the interaction Hamiltonia between the atom and the radiation field (external field). If we label the energy levels of the atom with energy E_k by

the energy eigenstate $|K\rangle$ and denote an atomic operator by a subscripted A, then we have

$$H_A = \sum_K E_K A_{KK} \quad , \quad A_{KL} = |K\rangle\langle L| \tag{2.1.2}$$

The unperturbed Hamiltonian of the quantized radiation field is

$$H_R = \sum_{\bar{K}S} \omega_{\bar{K}S} a^{\dagger}_{\bar{K}S} a_{\bar{K}S} \quad , \quad \hbar = 1 \tag{2.1.3}$$

where we have used the following mode expansion for the quantized electric field

$$\vec{E}_R(\vec{r}) = i \sum_{\bar{K}S} \left(\frac{2\pi cK}{L^3}\right)^{\frac{1}{2}} a_{\bar{K}S} \hat{\varepsilon}_{\bar{K}S} e^{i\vec{K}.\vec{r}} + H.c. \tag{2.1.4}$$

In the above equation, L^3 is the volume in which the field is quantized, $\hat{\varepsilon}_{\bar{K}S}$ is the polarization vector and $\vec{K}$ the wave vector, while $a_{\bar{K}S}$ and $a^{\dagger}_{\bar{K}S}$ are the annihilation and creation operators respectively. The interaction Hamiltonian has the following form in the dipole approximation

$$H_{AR}\ [H_{ext}(t)] = -\hat{\vec{d}}.\ \vec{E}_R\ [\vec{E}(t)] \tag{2.1.5}$$

where $\hat{\vec{d}}$ is the dipole moment operator of the atom having only off-diagonal elements :

$$\hat{\vec{d}} = \sum_{K,L} \vec{d}_{KL} A_{KL} + H.c.$$

$$\vec{d}_{KL} = 0 \qquad \text{if } K = L \qquad (2.1.6)$$

and $\vec{E}$ is the electric field at the point $\vec{r}$ where the atom is located. Using a classical description for the laser field, we may write the electric field of the modulated source at the position of the atom (chosen as the origin) as

$$\vec{E}(t) = \frac{1}{2} \sum_j \vec{\mathcal{E}}_{oj}(t) [1+a_j \cos(\upsilon_j t+\phi_j)] e^{-i\Omega_j t} + c.c. \qquad (2.1.7)$$

where $\vec{\mathcal{E}}_{oj}(t)$ is the amplitude of the j^{th} component oscillating at mean frequency Ω_j. The function $\vec{\mathcal{E}}_{oj}$ is taken to be a slowly varying function of time. For a strictly monochromatic field $\vec{\mathcal{E}}_{oj}(t)$ is independent of time t and is not a random variable, but for a fluctuating field, $\vec{\mathcal{E}}_{oj}(t)$ is a stochastic function. The amplitude of the j^{th} component of the laser beam is modulated in time at a frequency υ_j, and the modulation is assumed to be weak i.e. the modulation ratio $a_j \ll 1$. The time-averaged intensity of the incident laser field is

$$I_{inc}(t) = \frac{1}{2} \sum_j \mathcal{E}_{oj}^2 [1+a_j \cos(\upsilon_j t+\phi_j)]^2$$

$$\cong \frac{1}{2} \sum_j \mathcal{E}_{oj}^2 [1+2a_j \cos(\upsilon_j t+\phi_j)] \qquad (2.1.8)$$

where we have assumed the modulation frequencies to be much smaller than the optical frequencies i.e. $\nu_j \ll \Omega_j$. Equation (2.1.8) shows that as long as we ignore terms of order a_j^2, the modulation of the intensity and the amplitude of the laser beam are equivalent. In all the subsequent chapters, except chapter VI, we study the intensity of the fluorescent light to first order in modulation. However, in chapter VI, we examine the fluorescence to higher orders in modulation with some very remarkable consequences. The interaction Hamiltonian has the form given by (2.1.5) in the dipole approximation. We will transform the Hamiltonian to the rotating frame and neglect terms oscillating at twice the optical frequencies in the rotating wave approximation.

Section - 2.2 : Equations of Motion for the Atomic System :

Since we are ultimately interested in the properties of the atomic system, we must eliminate the degrees of freedom corresponding to relaxation and spontaneous emission. We first consider the case when there is no external field i.e. $H_{ext}(t) = 0$. Let $\rho_{A+R}(t)$ be the density operator characterizing the statistical state of the combined system of the atom and the radiation field, or rather a general reservoir responsible for all types of relaxation mechanism.

It satisfies the Schrödinger equation

$$\dot{\rho}_{A+R}(t) = -i\,[H,\ \rho_{A+R}(t)] = \mathcal{L}\ \rho_{A+R}(t) \qquad (2.2.1)$$

where $\mathcal{L}$ is the Liouville operator defined by

$$\mathcal{L}\ \ldots = -i\,[H,\ldots] \qquad (2.2.2)$$

Let $\rho_A(t)$ be the reduced density operator corresponding to the atomic system alone. $\rho_A(t)$ and $\rho_{A+R}(t)$ are related by

$$\rho_A(t) = T_r'\ \rho_{A+R}(t) \qquad (2.2.3)$$

where T_r' denotes the trace over the radiation field variables and any other relaxation process. The master equation for the reduced density operator corresponding to the atomic system in the Born, Markov and rotating wave approximation[78-80] is

$$\frac{\partial \rho_{mn}}{\partial t} = -i\omega_{mn}\ \rho_{mn} + \delta_{mn} \sum_{k\neq m} 2\gamma_{mk}\ \rho_{kk} - \Big(\sum_{k\neq m} \gamma_{km} + \sum_{k\neq n} \gamma_{kn} \Big) \rho_{mn} \qquad (2.2.4)$$

where ω_{mn} is the renormalised transition frequency between the atomic levels $|m\rangle$ and $|n\rangle$, and $2\gamma_{mk}$ represents the transition probability per unit time that the atom undergoes

a transition from state $|k\rangle$ to $|m\rangle$ due to spontaneous emission, collisional relaxation and any other incoherent process. If the transition $|k\rangle \rightarrow |m\rangle$ results from spontaneous emission only, then $2\gamma_{mk}$ is equal to the Einstein A coefficient and is given by

$$\gamma_{mk} = \frac{2}{3} |d_{mk}|^2 \quad c^{-3} \omega_{km}^3 \qquad \text{if } E_k > E_m \ ,$$
$$= 0 \qquad \text{if } E_k < E_m \qquad (2.2.5)$$

In equation (2.2.4), we have suppressed the subscript A from ρ. We may write equation (2.2.4) as

$$\frac{\partial \rho}{\partial t} = -i\,[H_A, \rho] + \mathcal{L}_o \rho \qquad (2.2.6)$$

where $\mathcal{L}_o$ represents the contribution from all the incoherent terms arising due to spontaneous emission, collisions etc.

In the presence of external fields ($H_{ext}(t) = 0$), equation (2.2.6) is modified to[81]

$$\frac{\partial \rho}{\partial t} = -i\,[H_A + H_{ext}(t),\ \rho] + \mathcal{L}_o \rho \qquad (2.2.7)$$

which simply implies that one can superimpose the effects of the external field and incoherent interactions. We now remove the fast time dependences from $\rho(t)$ to give the

slowly varying quantities $\tilde{\rho}(t)$ (tilde denotes a quantity in the rotating frame) and neglect terms oscillating at twice the optical frequencies (RWA). The equation of motion for $\tilde{\rho}(t)$ is

$$\frac{\partial \tilde{\rho}}{\partial t} = \mathcal{L}_o \tilde{\rho} - i [\tilde{H}_o, \tilde{\rho}] - i [\tilde{H}_{mod}(t), \tilde{\rho}]$$

$$\equiv L_o \tilde{\rho} + L_{mod}(t) \tilde{\rho} \qquad (2.2.8)$$

where we have explicitly separated the modulated and the unmodulated parts in $H_{ext}(t)$ so that H_o is the sum of the unperturbed atomic Hamiltonian and the interaction Hamiltonian between the atom and the unmodulated laser field. $\tilde{H}_{mod}(t)$ is still time dependent in the rotating frame due to modulation. L_o and $L_{mod}(t)$ are defined by

$$L_o \ldots = \mathcal{L}_o \ldots - i [H_o, \ldots]$$

$$L_{mod}(t) \ldots = -i [\tilde{H}_{mod}(t), \ldots] \quad . \qquad (2.2.9)$$

We will show in subsequent chapters that for an atomic system with discrete number of energy levels involved in the interaction with the laser beams, the equations for the evolution of the density matrix elements as obtained from (2.2.8) form a closed set of coupled, linear differential equations with constant coefficients. In matrix notation, they can be simply written as

$$\frac{\partial\tilde{\psi}(t)}{\partial t}=B\tilde{\psi}(t)+I+\sum_j a_j \cos(\upsilon_j t+\phi_j)[M_j\tilde{\psi}(t)+J_j] \qquad (2.2.10)$$

where $\tilde{\psi}(t)$ is a column matrix with elements $\tilde{\rho}_{mn}(t)$, m,n=1,2....N where N is the number of relevant energy levels of the atom under consideration. The order of $\tilde{\psi}(t)$, I and J_j is (N^2-1), since we use the normalisation condition $\sum_{m=1}^{N}\rho_{mm}(t)=1$ to eliminate the ground state density matrix element. This is done in order to avoid the difficulty that would arise in computing the solutions by Laplace transform techniques were the matrix B to possess a zero eigenvalue. B (M_j) is a square matrix of the same rank and is time independent due to RWA. The structure of the matrix B depends on the nature of the atomic/molecular transitions ; hence the eigenvalues of B will give information about the complex transition frequencies of the system.

Section - 2.3 : Solution of Master Equation :

Equation (2.2.10) can not be solved exactly so that a perturbation approach must be used, since $a_j \ll 1$, a perturbation expansion may be made with a_j as the expansion parameter, while the interaction with the laser field will be treated exactly. We expand $\tilde{\psi}(t)$ in the following power series :

$$\tilde{\psi}(t) = \tilde{\psi}^{(0)}(t) + \tilde{\psi}^{(1)}(t) + \tilde{\psi}^{(2)}(t) + \cdots \qquad (2.3.1)$$

and on substituting in (2.2.10) we obtain the expressions for different orders by equating the coefficients of similar powers of a_j.

(a) Solutions to zeroth order in Modulation :

When $a_j=0$, equation (2.2.10) reduces to

$$\frac{\partial \tilde{\psi}^{(o)}(t)}{\partial t} = B\tilde{\psi}^{(o)}(t) + I \qquad (2.3.2)$$

On taking the Laplace transform, we obtain

$$\hat{\tilde{\psi}}^{(o)}(z) = (z-B)^{-1}\,\tilde{\psi}^{(o)}(0) + \frac{(z-B)^{-1}}{z} I \qquad (2.3.3)$$

where we have used the notation

$$\hat{\tilde{\psi}}(z) = \int_o^\infty e^{-zt}\,\tilde{\psi}(t)\,dt \quad , \quad \mathrm{Re}\ z > o \qquad (2.3.4)$$

From (2.3.3), we obtain the steady-state solution as

$$\tilde{\psi}^{(o)}(\infty) = \mathcal{L}t_{\,z\to o+}\, z\hat{\tilde{\psi}}^{(o)}(z) = (-B)^{-1} I \qquad (2.3.5)$$

(b) Solutions to First Order in Modulation :

The first order equation in a_j is obtained from (2.2.10) as

$$\frac{\partial \tilde{\psi}^{(1)}(t)}{\partial t} = B\tilde{\psi}^{(1)}(t) + \sum_j a_j \cos(\upsilon_j t+\phi_j)[M_j\tilde{\psi}^{(o)}(t) + J_j] \qquad (2.3.6)$$

On taking the Laplace transform, we obtain

$$\hat{\tilde{\psi}}^{(1)}(z) = \sum_j \tfrac{1}{2} a_j (z-B)^{-1}\Big\{ M_j\,[e^{i\phi_j}\hat{\tilde{\psi}}^{(o)}(z-i\upsilon_j) + e^{-i\phi_j}\hat{\tilde{\psi}}^{(o)}(z+i\upsilon_j)]$$

$$+ J_j\,\Big[\frac{e^{i\phi_j}}{z-i\upsilon_j} + \frac{e^{-i\phi_j}}{z+i\upsilon_j}\Big]\Big\} \qquad (2.3.7)$$

It is clear that the first-order steady-state solution for any element of $\tilde{\psi}$ has the form

$$\psi^{(1)} = \sum_j \left(\eta^{(+)}_{j,\ell} \, e^{i\nu_j t} + \eta^{(-)}_{j,\ell} \, e^{-i\nu_j t} \right) \qquad (2.3.8)$$

where

$$\psi^{(1)} = \mathcal{L}t_{t\to\infty} \tilde{\psi}^{(1)}_{\ell}(t) \quad , \quad \ell=1,2\ldots(N^2-1) \qquad (2.3.9)$$

and $\eta^{(\pm)}_{j,\ell}$ are time independent and one can show that

$$\eta^{(\pm)}_{j,\ell} = \mathcal{L}t_{z\to o} \; z \, \hat{\tilde{\psi}}^{(1)}_{\ell}(z \pm i\nu_j) \qquad (2.3.10)$$

Hence, the first-order steady-state solution is given by

$$\psi^{(1)} = \tfrac{1}{2} \sum_j a_j \left\{ \left[(i\nu_j - B)^{-1} \, e^{i(\nu_j t + \phi_j)} + (-i\nu_j - B)^{-1} e^{-i(\nu_j t + \phi} \right.\right.$$

$$\left.\left. \times (M_j \tilde{\psi}^{(o)}(\infty) + J_j) \right\} \ell^{\text{th}} \text{ element} \qquad (2.3.11)\right.$$

Note the presence of denominators like determinant of $(\pm i\nu_j - B)$ which arise in inverting the matrices. It follows that $\psi^{(1)}$ as a function of ν_j will exhibit resonances whenever ν_j happen to be equal to one of the transition frequency of the system (the latter being characterised by the eigenvalues of matrix B). Some of these peaks may not appear, either because their weight factor is zero, or they are not clearly resolved.

Section - 2.4 : Total Intensity of the Modulated Fluorescence :

We are interested in the total intensity of the fluorescence which is spontaneously emitted by the atom/molecule. Let $\vec{E}^{(+)}$ be the positive frequency part of the radiation field emitted by the atom. In the radiation zone, it can be written as[74]

$$\vec{E}^{(+)}(\vec{r},t) = \vec{E}_o^{(+)}(\vec{r},t) - \sum_{k,l}' \frac{\omega_{kl}^2}{rc^2} [\hat{r}x(\hat{r} \times \vec{d}_{kl})] A_{lk}(t-\frac{r}{c}) \quad (2.4.1)$$

where $\vec{E}_o^{(+)}$ is just the free-field part, while the remaining terms relate the far-field-zone behaviour of the emitted radiation to the properties of the atomic system. The prime on the summation sign indicates that the sum on the energy levels is only for those terms for which $\omega_{kl} > 0$ and the transition $|k\rangle \rightarrow |l\rangle$ is a radiative decay. The intensity of the emitted radiation, which is equal to the normally ordered correlation function $\langle \vec{E}^{(-)}(\vec{r},t).\vec{E}^{(+)}(\vec{r},t) \rangle$, is given by

$$I_{scat} = \sum_{k,l,m}' \frac{\omega_{kl}^2 \omega_{ml}^2}{r^2 c^4} [\hat{r}x(\hat{r}x\vec{d}_{ml}^{*})].[\hat{r}x(\hat{r}x\vec{d}_{kl})] \langle A_{mk}(t-\frac{r}{c}) \rangle \quad (2.4.2)$$

where we have made use of the orthogonalisation condition $A_{mn}(t)\, A_{lk}(t) = \delta_{n,l}\, A_{mk}^{(t)}$. The free field term does not

contribute to (2.4.2) because the radiation field is initially in the vacuum state. $\langle A_{mk}(t) \rangle$ which appears in (2.4.2) is to be obtained from the solution of the master equation because $\langle A_{mk}(t) \rangle = \rho_{km}(t)$. Since the intensity of the fluorescent light is detected at times much larger than the natural lifetime or the decay constants involved in (2.2.9), the relevant density matrix elements ρ_{km} have to be evaluated in the steady-state limit. Hence, the total intensity of the fluorescent light to zeroth - and first-order in modulation is proportional to

$$I^{(o)}_{scat} = \sum'_{k,l,m} \frac{\omega^2_{kl}\ \omega^2_{ml}}{r^2 c^4} [\hat{r}x(\hat{r}x\vec{d}^{\,*}_{ml})].[\hat{r}x(\hat{r}x\vec{d}_{kl})]\ \rho^{(o)}_{km}(\infty) \quad (2.4.3)$$

$$I^{(1)}_{scat} = \sum'_{k,l,m} \frac{\omega^2_{kl}\ \omega^2_{ml}}{r^2 c^4} [\hat{r}x(\hat{r}x\vec{d}^{\,*}_{ml})].[\hat{r}x(\hat{r}x\vec{d}_{kl})]\ \rho^{(1)}_{km}(\infty)$$

$$= \sum'_{k,l,m} \sum_{j} \frac{\omega^2_{kl}\ \omega^2_{ml}}{r^2 c^4} [\hat{r}x(\hat{r}x\vec{d}^{\,*}_{ml})].[\hat{r}x(\hat{r}x\vec{d}_{kl})](\eta^{(+)}_{km,j} e^{i\nu_j t} + \eta^{(-)}_{km,j} e^{-i\nu_j t}) \quad (2.4.4)$$

where $\eta^{(\pm)}_{km,j}$ are time independent constants, so that the first order intensity has terms oscillating at the modulation frequencies. Thus, once we have obtained the solution of the master equation for the relevant atomic density matrix elements at steady states, we have complete knowledge of the total intensity of the modulated fluorescence, which in turn will contain interesting information about the atomic system, such as the lifetime

of the excited state, its Zeeman and hyperfine splittings or the dynamic Stark splittings in an intense laser beam, the study of which will be carried out in subsequent chapters.

Section - 2.5 : Relationship between Modulated Fluorescence and the Correlation functions of the atomic system :

In this section, we relate the intensity of the modulated fluorescence to the linear response of the system[44-47] and to the two time correlation functions of the atomic system. If we separate the modulated and the unmodulated part of $H_{ext}(t)$, then we have seen in section 2 that the density matrix for the atomic system in the rotating frame satisfies equation (2.2.8) which had the form

$$\frac{\partial \tilde{\rho}(t)}{\partial t} = L_o \tilde{\rho}(t) + L_{mod}(t) \tilde{\rho}(t)$$

where L_o is the unperturbed Liouville operator giving all the coherent (external field) and incoherent (spontaneous emission, collisions etc.) interactions. Since the modulation is weak, we can study the response of $\tilde{\rho}$ to lowest order in a_j. From (2.2.8), we have the following equation for the first-order density matrix :

$$\frac{\partial \tilde{\rho}^{(1)}(t)}{\partial t} = L_o \tilde{\rho}^{(1)}(t) + L_{mod}(t) \tilde{\rho}^{(o)}(t) \qquad (2.5.1)$$

which can be easily solved to give

$$\tilde{\rho}^{(1)}(t) = \int_0^\infty d\tau \; e^{L_0 \tau} \, L_{mod}(t-\tau) \; \tilde{\rho}^{(o)}_{st} \tag{2.5.2}$$

where $\tilde{\rho}^{(o)}_{st}$ is the zeroth-order steady-state solution given by

$$L_o \, \tilde{\rho}^{(o)}_{st} = 0 \tag{2.5.3}$$

For any general quantum mechanical operator $\tilde{Q}$, the change in its expectation value to first order in perturbation can be written as

$$\langle \tilde{Q}(t) \rangle = \mathrm{Tr} \, \tilde{Q}(o) \; \tilde{\rho}^{(1)}_{(t)} \tag{2.5.4}$$

From (2.2.8) and (2.5.2) and using simple properties of quantum mechanical operators, we have

$$\langle \tilde{Q}(t) \rangle = -i \int_0^\infty d\tau \; \langle [\tilde{Q}(\tau), \tilde{H}_{mod}(t-\tau)] \rangle_{st} \, d\tau \tag{2.5.5}$$

where the two time correlation function $\langle [\tilde{Q}(\tau), \tilde{H}_{mod}(t-\tau)] \rangle$ has to be evaluated in the absence of the modulating field. It is clear that if the Hamiltonian for the modulated part has the form

$$H_{mod}(t) = \sum_j (H_j \, e^{-i\nu_j t} + H.c.) \tag{2.5.6}$$

Then the linear response of $\langle \tilde{Q}(t) \rangle$ may be written as

$$\langle \tilde{Q}(t) \rangle = \sum_j (Q_j^{(+)} \, e^{-i\nu_j t} + Q_j^{(-)} \, e^{i\nu_j t}) \tag{2.5.7}$$

where

$$Q_j^{(+)} = -i \int_0^\infty d\tau \; e^{i\nu_j \tau} \left\langle [Q(\tau), H_j] \right\rangle_{st} \qquad (2.5.8)$$

The two time correlation function appearing in (2.5.8) can be calculated by using the Quantum Regression Theorem[82,83] and the solutions of the master equation in the absence of modulation. We have already seen that the intensity of the modulated fluorescence, as given by (2.4.2), is proportional to the expectation value of the atomic operators A_{mk}^t From (2.5.8), we have

$$A_{mk,j}^{(+)} = i \int_0^\infty d\tau \; e^{i\nu_j \tau} \left\langle [A_{mk}^t{}_{,j} \quad H_j] \right\rangle_{st} \qquad (2.5.9)$$

Hence, the first order intensity is proportional to

$$I_{scat}^{(1)} = \sum_{k,l,m}{}' \sum_j \frac{\omega_{kl}^2 \omega_{ml}^2}{r^2 c^4} [\hat{r}x(\hat{r}x\vec{d}_{ml}^{\,*})].[\hat{r}x(\hat{r}x\vec{d}_{kl})] \; x$$

$$\left\{ -ie^{-i\nu_j t} \int_0^\infty d\tau \; e^{i\nu_j \tau} \left\langle [A_{mk,j}(\tau) \; , \; H_j] \right\rangle_{st} \right.$$

$$\left. + ie^{i\nu_j t} \int_0^\infty d\tau \; e^{-i\nu_j \tau} \left\langle [H_j^\dagger, \; A_{km,j}(\tau)] \right\rangle_{st} \right\} \qquad (2.5.10)$$

so that the intensity of the modulated fluorescence can be related to the linear response of the system and to the two time correlation functions of the atomic system evaluated in the absence of modulation.

The relation (2.5.10) is to be compared with the following result giving the spectrum of the spontaneously emitted radiation in the absence of modulation

$$S(\omega) = \int_{-\infty}^{\infty} d\tau \; e^{-i\omega\tau} \left\langle \vec{E}^{(-)}_{(t+\tau)} \cdot \vec{E}^{(+)}_{(t)} \right\rangle$$

$$= \sum_{k,l,m,n}{}' (\text{constants}) \int_{-\infty}^{\infty} d\tau \; e^{-i\omega\tau} \left\langle A_{mn}(t+\tau)\, A_{kl}(t) \right\rangle \qquad (2.5.11)$$

where we have made use of the linear dependence of the radiation source-field operators and the atomic operators as given by equation (2.4.1). The prime on the summation sign indicates that only those terms are to be considered for which $\omega_{mn}, \omega_{kl} > 0$ and that the transition $|m\rangle \rightarrow |n\rangle$, $|k\rangle \rightarrow |l\rangle$ are radiatively allowed. Equation (2.5.11) is similar in structure to (2.5.10). Hence, detection of the modulated fluorescence will yield information similar in nature as the spectrum of the unmodulated fluorescence about the atomic system.

CHAPTER - III

RESPONSE OF AN ATOM TO A NEAR-RESONANT EXCITATION BY A WEAKLY MODULATED LASER

As emphasized in chapter I, it would be quite interesting to study the modulated fluorescence within the framework of a general relaxation theory, when collisions and other non-radiative relaxations play a crucial role in determining the phaseshift and amplitude of modulation. Such a study would be relevant for experiments where the samples are contained in vapor cells, which are at times preferable because of the relative ease with which they can be performed. Also a cell, in general, requires less material than in the use of atomic beams and since it can be used for a long time without refilling it is often desirable to monitor the fluorescence emitted from samples in cell. The effect of Doppler broadening will have to be considered now. Moreover, in an actual experiment, the optical source used for excitation will always have a finite bandwidth, a monochromatic or a broad-band source being a limiting case. Thus a complete study of the problem must take into account the effects of fluctuations in the exciting source which give rise to its finite bandwidth.

In this chapter, we entail a detailed study of the fluorescence emitted by atoms/molecules in cells when excited

by modulated lasers of arbitrary bandwidth and intensity and under arbitrary relaxations. Atomic relaxations are described by the parameters T_1 and T_2 which govern the decay of the energy (inversion) and dipole moment of the atom respectively. The results corresponding to radiative decay only (as in atomic beam experiments) are obtained as a special case of the general theory. In general, the modulated fluorescence is shown to be critically dependent on the nature of atomic relaxations, as well as the bandwidth of the exciting source and its frequency of modulation. The results for monochromatic[6] and broad-band[1-5] excitation are contained as limiting cases. The position, width and weight of the resonances in the amplitude of modulation are explicitly discussed for the case of intense, resonant excitation. For weak fields, the velocity averaged modulated fluorescence for monochromatic excitation exhibits the same behaviour as for broad-band excitation, so long as the Doppler width is much larger than the natural width, laser detuning and its frequency of modulation. For intense fields, numerical computations reveal a shift in the resonance at the Rabi frequency for large Doppler widths.

Section 3.1 : Dynamical Equations :

We now consider the modulated laser beam to be nearly resonant with a particular atomic transition. We take these

two levels to be those with energy eigenstates $|1\rangle$ and $|2\rangle$ having the energies $\frac{1}{2}\omega$ and $-\frac{1}{2}\omega$ respectively. From (2.1.6) we have the dipole moment operator of the atom as

$$\hat{\vec{d}} = \vec{d}_{12} |1\rangle\langle 2| + \vec{d}_{21} |2\rangle\langle 1|$$

$$\equiv \vec{d}_{12} (S^+ + S^-) \qquad (3.1.1)$$

where we have assumed that with a proper choice of phases the dipole matrix element can be made real. In (3.1.1), $S^{\pm}$ are the operators defined by

$$S^+ = |1\rangle\langle 2| \equiv A_{12} \ , \ S^- = |2\rangle\langle 1| \equiv A_{21} \qquad (3.1.2)$$

We also introduce the operator S^z defined by

$$S^z = \tfrac{1}{2} (|1\rangle\langle 1| - |2\rangle\langle 2|) \equiv \tfrac{1}{2} (A_{11} - A_{22}) \qquad (3.1.3)$$

and we have the normalization condition

$$|1\rangle\langle 1| + |2\rangle\langle 2| = 1 \equiv A_{11} + A_{22} \qquad (3.1.4)$$

Since we are considering excitation by a monochromatic laser beam whose amplitude is weakly modulated in time, we have the following expression from (2.1.7) for the electric field

$$\vec{E}(t) = \tfrac{1}{2} \vec{\mathcal{E}}_o [(1+a \cos (\nu t+\phi)] \, e^{-i\Omega t} + c.c. \qquad (3.1.5)$$

where $\vec{\mathcal{E}}_o$ is a deterministic quantity independent of time. In a frame rotating with angular frequency Ω, the interaction Hamiltonian between the atom and the external field is given by

$$\tilde{H}_{ext}(t) = \alpha\,[1+a\cos(\nu t+\phi)]\,(\tilde{S}^{+}+\tilde{S}^{-}) \qquad (3.1.6)$$

where

$$\tilde{S}^{\pm} = e^{\mp i\Omega t}\,S^{\pm} \quad , \quad \tilde{S}^{z} = S^{z} \qquad (3.1.7)$$

with

$$2\alpha = -\vec{d}_{12}\cdot\vec{\mathcal{E}}_o \qquad (3.1.8)$$

being the Rabi frequency characterising the coupling of the laser field with the $|1\rangle \leftrightarrow \langle 2|$ transition. In deriving (3.1.6), we have neglected terms oscillating as $e^{\pm 2i\Omega t}$ (RWA). For the two level atom under consideration, the excited state $|1\rangle$ decays radiatively to the ground state $|2\rangle$, so we may write the electric field of the spontaneously emitted fluorescence in the radiation zone with the help of equation (2.4.1) as

$$\vec{E}^{(+)}(\vec{r},t) = \vec{E}_o^{(+)}(\vec{r},t) - \frac{\omega^2}{c^2 r}\,[\hat{r}\times(\hat{r}\times\vec{d}_{12})]\,S^{-}(t-\tfrac{r}{c}) \qquad (3.1.9)$$

Hence the intensity of the fluorescence is given by

$$I_{scat} = \frac{\omega^4}{c^4 r^2} [\hat{r} \times (\hat{r} \times \vec{d}_{12})] . [\hat{r} \times (\hat{r} \times \vec{d}_{12})] \langle A_{11} (t-\tfrac{r}{c}) \rangle$$

$$\alpha \rho_{11} (t-\tfrac{r}{c})$$

$$= \tfrac{1}{2} + \langle S^z (t- \tfrac{r}{c}) \rangle \qquad (3.1.10)$$

Since the signals are detected at times much larger than the decay constants involved in the problem, the atomic-density matrix elements must be evaluated in the steady-state limit. The total intensity of the fluorescent light to first order in modulation is proportional to

$$I^{(1)}_{scat} \propto \langle S^z(\infty) \rangle^{(1)} \qquad (3.1.11)$$

In order to calculate $\langle S^z(\infty) \rangle^{(1)}$, we must solve the master equation for the atomic density matrix within the framework of a general relaxation theory. The master equation for a two-level atom undergoing arbitrary relaxation processes can be written in the form[79]

$$\frac{\partial \tilde{\rho}}{\partial t} = -i [\tilde{H}_A + \tilde{H}_{ext}(t), \tilde{\rho}] + \tfrac{1}{2} \gamma_{12} (2 S^+ \tilde{\rho} S^- - \tilde{\rho} S^- S^+ - S^- S^+ \tilde{\rho})$$

$$+ \tfrac{1}{2} \gamma_{21} (2 S^- \tilde{\rho} S^+ - \tilde{\rho} S^+ S^- - S^+ S^- \tilde{\rho}) + \Gamma^{ph}_{21} (2 S^z \tilde{\rho} S^z - \tilde{\rho} S^z S^z - S^z S^z \tilde{\rho})$$

$$(3.1.12)$$

where

$$\Gamma_{ij}^{ph} = \pi \sum_{E_R} \left| \langle E_R | v_{ii} - v_{jj} | E_R \rangle \right|^2 \rho_R(E_R) \qquad (3.1.13)$$

while E_R and $|E_R\rangle$ are the energy eigenvalues and eigenfunctions of H_R and v's in the above equation are functions of the reservoir operators only. Equations (3.1.6) and (3.1.12) lead to the following equations for the macroscopic mean values :

$$\langle \dot{\tilde{S}}^+(t) \rangle = \left(-\frac{1}{T_2} + i\Delta\right) \langle \tilde{S}^+(t) \rangle - 2i\alpha \left[1 + a\cos(\upsilon t + \phi)\right] \langle \tilde{S}^z(t) \rangle$$

$$\langle \dot{\tilde{S}}^z(t) \rangle = -\frac{1}{T_1} \left(\langle \tilde{S}^z(t) \rangle - \langle S^z \rangle_{eq} \right) - i\alpha \left[1 + a\cos(\upsilon t + \phi)\right] \left(\langle \tilde{S}^+(t) \rangle - c.c. \right) \qquad (3.1.14)$$

where

$$\Delta = \omega - \Omega \,, \quad \langle \tilde{S}^{\pm}(t) \rangle = e^{\mp i\Omega t} \langle S^{\pm}(t) \rangle \,; \quad \langle \tilde{S}^z(t) \rangle = \langle S^z(t) \rangle \qquad (3.1.15)$$

and where T_1 and T_2 are the longitudinal and transverse relaxation times defined by

$$\frac{1}{T_1} = \gamma_{12} + \gamma_{21} \,, \quad \frac{1}{T_2} = \left(\frac{1}{2}\gamma_{12} + \frac{1}{2}\gamma_{21} + \Gamma_{21}^{ph} \right) \qquad (3.1.16)$$

with $2\gamma_{fi}$ being the transition probability per unit time that the atom makes a transition from $|i\rangle \rightarrow |f\rangle$ due to relaxations.

Thus T_1 and T_2 govern the decay of the energy and dipole moment of the atom respectively and in general $T_1 \neq T_2$.[84] In the strong collision model[85] of atomic relaxation, $T_1 = T_2 \equiv T$ where T^{-1} is the mean collision rate, so that a collision causes decay of energy too whenever it damps the dipole oscillations. $\langle S^z \rangle_{eq}$ is the equilibrium value to which the inversion relaxes to in the absence of the external field. In the case of radiative relaxation, T_1^{-1} is the spontaneous emission rate, and $T_2 = 2T_1$, with $\langle S^z \rangle_{eq} = -\frac{1}{2}$ since the atom decays to the ground state when no fields are present.

Section 3.2 : Analytical Expressions for Modulated Fluorescence

(i) Weak Field : For weak fields ($\frac{\alpha}{\gamma} \ll 1$), we may expand the expectation values in powers of α and obtain the various order perturbation results. We will be interested in calculating the intensity to first order in the laser intensity i.e. to second order in the Rabi frequency α . The modulation will as usual be considered weak ($a \ll 1$). Indicating the order of perturbation with respect to the laser field (modulation) by subscript (superscript), we have from (3.1.14) the equation for $\langle \tilde{S}^z(t) \rangle$ to second order in α :

$$\langle \dot{\tilde{S}}^z(t) \rangle_{(2)} = -\frac{1}{T_1} \langle \tilde{S}^z(t) \rangle_{(2)} - i\alpha[1+a\cos(\upsilon t+\phi)] \times \left(\langle \tilde{S}^+(t) \rangle_{(1)} - \langle \tilde{S}^-(t) \rangle_{(1)} \right) \qquad (3.2.1)$$

which on integration yields

$$\langle \tilde{S}^z(t) \rangle_{(2)} = -i\alpha \int_0^t dt' \; e^{-\frac{1}{T_1}(t-t')} \langle \tilde{S}^+(t') \rangle_{(1)}$$

$$-\frac{a i\alpha}{2} e^{i\phi} \int_0^t dt' \; e^{-\frac{1}{T_1}(t-t')} \cdot e^{i\nu t'} \langle \tilde{S}^+(t') \rangle_{(1)}$$

$$-\frac{a i\alpha}{2} e^{-i\phi} \int_0^t dt' \; e^{-\frac{1}{T_1}(t-t')} \cdot e^{-i\nu t'} \langle \tilde{S}^+(t') \rangle_{(1)} + \text{c.c.}$$

(3.2.2)

We still have to calculate $\langle \tilde{S}^+(t) \rangle_{(1)}$, the equation for which is obtained from (3.1.14) as

$$\langle \dot{\tilde{S}}^+(t) \rangle_{(1)} = \left(-\frac{1}{T_2} + i\Delta\right) \langle \tilde{S}^+(t) \rangle_{(1)} - 2i\alpha \, [1 + a \cos(\nu t + \phi)] \times \langle \tilde{S}^z(t) \rangle_{(o)}$$

(3.2.3)

On integration we obtain

$$\langle \tilde{S}^+(t') \rangle_{(1)} = -i\alpha \langle S^z \rangle_{eq} \left\{ 2 \int_0^{t'} dt'' \; e^{\left(-\frac{1}{T_2} + i\Delta\right)(t'-t'')} \right.$$

$$+ a e^{i\phi} \int_0^{t'} dt'' \; e^{\left(-\frac{1}{T_2} + i\Delta\right)(t'-t'')} e^{i\nu t''} + a e^{-i\phi} \int_0^{t'} dt'' \; e^{\left(-\frac{1}{T_2} + i\Delta\right)(t'-t'')} e^{-i\nu t''} \Big\}$$

(3.2.4)

where we have used the result

$$\langle \tilde{S}^{z}(t) \rangle_{(o)} = \langle S^{z} \rangle_{eq} \qquad (3.2.5)$$

Substituting (3.2.4) in (3.2.2) and retaining terms only linear in a, we obtain

$$\langle S^{z}(t) \rangle_{(2)}^{(1)} = -a\alpha^{2} \langle S^{z} \rangle_{eq} \, e^{i\phi} \Big\{ \int_{o}^{t} dt' \, e^{-\frac{1}{T_1}(t-t')} \int_{o}^{t'} dt'' \, e^{(-\frac{1}{T_2}+i\Delta)(t'-t'')} \, e^{i\nu t''}$$

$$+ \int_{o}^{t} dt' \, e^{-\frac{1}{T_1}(t-t')} \int_{o}^{t'} dt'' \, e^{(-\frac{1}{T_2}+i\Delta+i\nu)(t'-t'')} \, e^{i\nu t''} \Big\}$$

$$-a\alpha^{2} \langle S^{z} \rangle_{eq} \, e^{-i\phi} \Big\{ \int_{o}^{t} dt' \, e^{-\frac{1}{T_1}(t-t')} \int_{o}^{t'} dt'' \, e^{(-\frac{1}{T_2}+i\Delta)(t'-t'')} \, e^{-i\nu t''}$$

$$+ \int_{o}^{t} dt' \, e^{-\frac{1}{T_1}(t-t')} \int_{o}^{t'} dt'' \, e^{(-\frac{1}{T_2}+i\Delta-i\nu)(t'-t'')} \, e^{-i\nu t''} \Big\} + c.c$$

(3.2.6)

n taking the Laplace transform of above equation and using quations (2.3.8) and (2.3.10), we obtain

$$\eta^{(+)} = \eta^{(-)*} = -a\alpha^{2} \langle S \rangle_{eq}^{z} \frac{e^{i\phi}}{\frac{1}{T_1}+i\nu} \left[\frac{2/T_2}{\frac{1}{T_2^2}+\Delta^2} + \frac{1}{\frac{1}{T_2}+i\nu-i\Delta} + \frac{1}{\frac{1}{T_2}+i\nu+i\Delta} \right]$$

(3.2.7)

Hence we find from equation (3.1.11) that the intensity of the modulated fluorescence in the weak field limit is proportional to

$$I^{(1)}_{scat} \propto \langle \tilde{S}^z(\infty) \rangle^{(1)}_{(2)} = \eta^{(+)} e^{i\nu t} + \eta^{(-)} e^{-i\nu t}$$

$$= -2a\alpha^2 \langle S^z \rangle_{eq} A \cos(\nu t + \phi + \Phi) \qquad (3.2.8)$$

where

$$A = (X_R^2 + X_I^2)^{\frac{1}{2}}$$

$$\tan\Phi = \frac{X_I}{X_R} \qquad (3.2.9)$$

with

$$X = \frac{1}{\frac{1}{T_1} + i\nu} \left[\frac{2/T_2}{\frac{1}{T_2^2} + \Delta^2} + \frac{1}{\frac{1}{T_2} + i\nu - i\Delta} + \frac{1}{\frac{1}{T_2} + i\nu + i\Delta} \right] \qquad (3.2.10)$$

When the laser field is exactly resonant with the atomic transition ($\Delta = 0$) then the phaseshift is given by

$$\tan\Phi = -\nu \frac{\frac{1}{T_2}\left(\frac{1}{T_2} + \frac{2}{T_2}\right) + \nu^2}{\frac{2}{T_1 T_2^2} + \nu^2\left(\frac{1}{T_1} - \frac{1}{T_2}\right)} \qquad (3.2.11)$$

For strong collisions ($T_1 = T_2 \equiv T$), the above expression reduces to

$$\tan\Phi \simeq -\tfrac{1}{2}\nu T (3 + \nu^2 T^2) \qquad (3.2.12)$$

so that for small frequencies of modulation ($\nu T \ll 1$), the plot of $\tan\Phi$ against ν is linear with a slope $-\frac{3T}{2}$, but if $\frac{1}{T_1} \gg \frac{1}{T_2}$ ($\nu T_1 \ll 1$), then the slope is $-\frac{1}{2} T_2$. For purely radiative damping ($T_2 = 2T_1 = \frac{1}{\gamma}$), equation (3.2.12) reduces to

$$\tan\Phi = -\frac{\nu}{\gamma} = -2\nu\tau \qquad (3.2.13)$$

where τ is the lifetime of the excited state, and the curve of $\tan\Phi$ against ν is linear for all frequencies of modulation, a result identical to the phaseshift obtained by Armstrong and Feneiulle.[6] Thus the phaseshift is seen to be critically dependent on the relaxation parameters occuring in the problem, as well as the modulation frequency. From the structure of (3.2.10), we expect the resonance in the amplitude of modulation at $\nu = 0$ to be a superposition of two resonances of width $\frac{1}{T_1}$ and $\frac{1}{T_2}$ when there is no detuning ($\Delta = 0$). With the presence of detunings, the peak at $\nu = 0$ shifts to a higher frequency, since we now expect the resonances at $\nu = \pm\Delta$ to contribute to A. Figures (1) and (2) show the effect of relaxation parameters on the phaseshift and amplitude of modulation respectively, which are studied as functions of modulation frequency. The strong collision ($T_2 = T_1$) or radiative relaxation ($T_2 = 2T$) models give similar behaviour for the phaseshift ; however if $T_1 > T_2$, then an altogether different behaviour is

obtained. Thus experimental measurements of atomic lifetimes by the phase-shift method using weak sources will have to be interpreted by taking into account the nature of atomic relaxation. Figure (2) reveals that the maxima at $\upsilon = 0$ is sharper for the strong collision than radiative relaxation case. For finite detunings, the shift of the maxima to a higher value of modulation frequency is different for the various relaxation mechanisms. Clearly $\Delta \gg \gamma$ for the resonances at $\upsilon = 0$ and $\upsilon = \Delta$ to be clearly resolved.

(ii) <u>Intense Field</u> : We now calculate the intensity of the modulated fluorescence to first order in modulation, the coupling with the intense laser beam being treated to all orders. To be able to do that, we write the coupled equations (3.1.14) in the matrix form introduced in (2.2.9)

$$\dot{\tilde{\psi}}(t) = B\,\tilde{\psi}(t) + I + a\cos(\upsilon t + \phi)\, M\,\tilde{\psi}(t)$$

where $\tilde{\psi}(t)$, I, B and M are the following matrices

$$\tilde{\psi}(t) = \begin{bmatrix} \langle \tilde{S}^{+}(t) \rangle \\ \langle \tilde{S}^{-}(t) \rangle \\ \langle \tilde{S}^{z}(t) \rangle \end{bmatrix} \qquad I = \begin{bmatrix} 0 \\ 0 \\ \dfrac{\langle S^{z} \rangle_{eq}}{T_1} \end{bmatrix} \qquad (3.2.14)$$

$$B = \begin{bmatrix} -\frac{1}{T_2} + i\Delta & 0 & -2i\alpha \\ 0 & -\frac{1}{T_2} - i\Delta & 2i\alpha \\ -i\alpha & i\alpha & -\frac{1}{T_1} \end{bmatrix}, \quad \begin{matrix} M_{ij} = B_{ij} \\ M_{ii} = 0 \end{matrix} \qquad (3.2.15)$$

By following exactly the procedure outlined in section 3 of chapter II for solving equations of the type given by (3.2.15), we obtain the intensit of the scattered light as

$$I^{(1)}_{scat} \propto \langle \tilde{S}^{z}(\omega) \rangle^{(1)}$$

$$= -a\alpha^2 \langle S^z \rangle_{eq} e^{i(\nu t + \phi)} \sum_{k,l=1}^{3} (i\nu - B)^{-1}_{3k} M_{kl} \tilde{\psi}^{(o)}_{l}(\omega) + c.c.$$

$$\equiv -a\alpha^2 \langle S^z \rangle_{eq} A \cos(\nu t + \phi + \Phi) \qquad (3.2.16)$$

where

$$A = 2 (Y_R^2 + Y_I^2)^{\frac{1}{2}}$$

$$\tan \Phi = Y_I / Y_R \qquad (3.2.17)$$

with

$$Y \equiv \frac{1}{(T_1/T_2)[4\alpha^2 + \frac{1}{T_1}(\frac{1}{T_2} + \Delta^2 T_2)]} \cdot \frac{(i\nu + \frac{1}{T_2})(\frac{1}{T_2^2} + \Delta^2) + \frac{1}{T_2}[(i\nu + \frac{1}{T_2})^2 + \Delta^2]}{(i\nu + \frac{1}{T_1})[(i\nu + \frac{1}{T_2})^2 + \Delta^2] + 4\alpha^2 (i\nu + \frac{1}{T_2})}$$

(3.2.18)

To obtain an idea of the position, width and height of the resonances in the amplitude of modulation, we must examine the following expression from (3.2.18)

$$P(z) = (\frac{1}{T_1} + z)[(\frac{1}{T_2} + z)^2 + \Delta^2] + 4\alpha^2 (\frac{1}{T_2} + z) \quad , \quad z \equiv i\nu \qquad (3.2.19)$$

the roots of which are the eigenvalues of the problem. In the most general case there are no simple factorisations so that a cubic equation will have to be solved. However, we shall study the special case of intense, resonant laser fields in detail. Equation (3.2.19) now becomes

$$P(z) = \left(\frac{1}{T_2} + z\right)\left[z^2 + z\left(\frac{1}{T_1} + \frac{1}{T_2}\right) + 4\alpha^2 + \frac{1}{T_1 T_2}\right] \qquad (3.2.20)$$

The roots are

$$z = \begin{cases} -\frac{1}{T_2} \\ -\frac{1}{2}\left(\frac{1}{T_1} + \frac{1}{T_2}\right) \pm \frac{1}{2} i \sqrt{16\,\alpha^2 - \left(\frac{1}{T_1} - \frac{1}{T_2}\right)^2} \end{cases} \qquad (3.2.21)$$

For intense fields ($\alpha \gg \frac{1}{T_1}, \frac{1}{T_2}$), the roots have the simple form

$$z_1 = -\frac{1}{T_2}, \quad z_2 = -\frac{1}{2}\left(\frac{1}{T_1} + \frac{1}{T_2}\right) + 2i\alpha, \quad z_3 = -\frac{1}{2}\left(\frac{1}{T_1} + \frac{1}{T_2}\right) - 2i\alpha \qquad (3.2.22)$$

which shows that for intense, resonant fields, there are three peaks at $\nu = 0,\ 2\alpha,\ -2\alpha$ with widths $\frac{1}{T_2}, \frac{1}{2}\left(\frac{1}{T_1} + \frac{1}{T_2}\right)$ respectively provided the weight of the peak is not zero. The following analysis shows that the central peak will not be present. If we write (3.2.18) in the form

$$Y = K\,\frac{\frac{1}{T_2}\left(z + \frac{1}{T_2}\right)\left(z + \frac{2}{T_2}\right)}{(z - z_1)(z - z_2)(z - z_3)} \qquad (3.2.23)$$

where

$$K = \frac{1}{\frac{T_1}{T_2}\left(4\alpha^2 + \frac{1}{T_1 T_2}\right)} \qquad (3.2.24)$$

then we have by the method of partial fractions

$$Y = K\left(\frac{A_1}{z - z_1} + \frac{A_2}{z - z_2} + \frac{A_3}{z - z_3}\right) \qquad (3.2.25)$$

Using the values of z_1, z_2 and z_3 from equation (3.2.22), we find that

$$A_1 = 0, \; A_2 = \frac{-i}{8\alpha T_2}\left(\frac{3}{T_2} - \frac{1}{T_1} + 4i\alpha\right), \; A_3 = A_2^* \qquad (3.2.26)$$

which shows that the central peak is absent for intense, resonant fields. The behaviour of the phaseshift for intense fields is shown in Figure (3). As is evident from the figure, the phaseshift is rather sensitive to the type of relaxation the atom is undergoing in this case. Figure (4) is a plot of the amplitude of modulation as a function of modulation frequency for the intense field case. The position of the dynamic Stark component is $\pm [(2\alpha)^2 + \Delta^2]^{\frac{1}{2}}$ irrespective of the magnitude of the relaxation parameters, the width of the peak being $\frac{1}{2}\left(\frac{1}{T_1} + \frac{1}{T_2}\right)$ as calculated, while the resonance at $\nu = 0$ is vanishingly small.

Section 3.3 : Effect of Temporal Fluctuations in the Laser Beam :

In this section, we study the influence of laser fluctuations which as emphasized in the introduction play a crucial role in the determination of lifetimes using phaseshifts [cf.Eqs. (1.1), (1.3)].

(i) Weak Field : We assume that the strength of the applied laser field is much less than the saturation field. In this case a perturbative solution in powers of Rabi frequency can be obtained. Such approximate results involve, to lowest order in field strengths, only the second order correlation functions of the field and hence are model independent. We now represent the applied field (3.1.5) in the form

$$\vec{E}(t) = \frac{1}{2}\,\vec{\mathcal{E}}_0(t)\,[1+a\cos(\nu t+\phi)]\,e^{-i\Omega t} + c.c. \qquad (3.3.3.)$$

where $\vec{\mathcal{E}}_0(t)$ is the complex fluctuating amplitude of the field. We assume that $\vec{\mathcal{E}}_0(t)$ is an analytic signal i.e.

$$\langle \vec{\mathcal{E}}_0(t) \rangle = 0, \langle \vec{\mathcal{E}}_0(t)\,\vec{\mathcal{E}}_0^{*}(t') \rangle = |\mathcal{E}_0|^2\, e^{-\gamma_c |t-t'|}$$

$$\langle \vec{\mathcal{E}}_0(t)\ \vec{\mathcal{E}}_0(t') \rangle = 0 \qquad (3.3.4)$$

where γ_c is the bandwidth of the laser beam, and we also assumed the stationarity of the field.[80] The angular brackets in equations (3.3.4) denote the ensemble averaging over the random

distribution of $\vec{\mathcal{E}}_o(t)$. The damped Bloch equation (3.1.14) now have the form

$$\langle \dot{\tilde{S}}^+(t)\rangle = (-\tfrac{1}{T_2}+i\Delta)\langle \tilde{S}^+(t)\rangle - 2i\alpha(t)\,[1+a\cos(\nu t+\phi)]\langle \tilde{S}^z(t)\rangle$$

$$\langle \dot{\tilde{S}}^z(t)\rangle = -\tfrac{1}{T_1}(\langle \tilde{S}^z(t)\rangle - \langle S^z_{eq}\rangle) + \{-i\alpha(t)\,[1+a\cos(\nu t+\phi)]\times$$

$$\langle \tilde{S}^+(t)\rangle + c.c.\} \qquad (3.3.5)$$

where

$$\alpha(t) = -\tfrac{1}{2}\,\vec{d}_{12}\cdot\vec{\mathcal{E}}_o(t) \qquad (3.3.6)$$

The Laplace transform method can not be used in the present case to solve (3.3.5) due to the random time dependence of $\alpha(t)$. Since we are interested in calculating the intensity of the modulated fluorescence to second order in applied field, we use the iteration method directly to solve (3.3.5). Proceeding exactly as in section 2 of this chapter for weak fields (the only difference being that the Rabi frequency is now time dependent), we obtain

$$\langle \tilde{S}^z(t)\rangle_{(2)} = -2\,\langle S^z\rangle_{eq}\int_o^t dt'\, e^{-\frac{1}{T_1}(t-t')}\,\alpha(t')\times$$

$$\int_o^{t'} dt''\; e^{(\frac{1}{T_2}+i\Delta)(t'-t'')}\,\alpha^*(t'')\,[1+a\cos(\nu t'+\phi)][1+a\cos(\nu t''+\phi)]+c.c.$$

(3.3.7)

Hence, we have to first order in modulation

$$\langle\langle \tilde{S}^z(t) \rangle\rangle_{(2)} = -2a\alpha^2 \langle S^z \rangle_{eq} \int_0^t dt' \, e^{-\frac{1}{T_1}(t-t')} \int_0^{t'} dt'' e^{(-\frac{1}{T_2}-\gamma_c+i\Delta)(t'-t'')} [\cos(\nu t'+\phi)+ \cos(\nu t''+\phi)] + c.c. \quad (3.3.8)$$

where the double bracket $\langle\langle \rangle\rangle$ now denotes the ensemble averaging over the random distribution of $\alpha(t)$, and we have made use of the relations (3.3.4). On taking the Laplace transform of the above equation and using the relations (2.3.8) and (2.3.10), we have

$$\eta^{(+)} = \eta^{(-)*} = \frac{2a\alpha^2 \langle S^z \rangle_{eq} e^{i\phi}}{(\frac{1}{T_1}+i\nu)} \left[\frac{\frac{1}{T_2}+\gamma_c}{(\frac{1}{T_2}+\gamma_c)^2+\Delta^2} + \frac{(\frac{1}{T_2}+\gamma_c+i\nu)}{(\frac{1}{T_2}+\gamma_c+i\nu)^2+\Delta^2} \right] \quad (3.3.9)$$

Thus, similar to (3.2.8), we may write the intensity of the modulated fluorescence averaged over the random distribution of the laser amplitude as

$$\langle I_{scat} \rangle^{(1)} = -2a\,\alpha^2 \langle S^z \rangle_{eq} A \cos(\nu t + \phi + \Phi) \quad (3.3.10)$$

where

$$A = (X_R^2 + X_I^2)^{\frac{1}{2}}$$

$$\tan\Phi = \frac{X_I}{X_R} \quad (3.3.11)$$

with

$$X = \frac{2}{\frac{1}{T_1}+i\nu}\left[\frac{\frac{1}{T_2}+\gamma_c}{(\frac{1}{T_2}+\gamma_c)^2+\Delta^2} + \frac{\frac{1}{T_2}+\gamma_c+i\nu}{(\frac{1}{T_2}+\gamma_c+i\nu)^2+\Delta^2}\right] \qquad (3.3.12)$$

The result (3.3.12) for radiative relaxation is in agreement with that of McClean and Swain[8] in the limit of weak field. We now study explicitly the case of radiative relaxation when the laser beam is exactly in resonance with the atomic transition. Equation (3.3.12) now has the form

$$X = \frac{2}{2\gamma+i\nu}\left(\frac{1}{\gamma+\gamma_c} + \frac{1}{\gamma+\gamma_c+i\nu}\right) \qquad (3.3.13)$$

so that the phaseshift in this case is given by

$$\tan\Phi = -\nu\,\frac{\nu^2+4\gamma^2+2\gamma_c^2+6\gamma\gamma_c}{\gamma(\nu^2+4\gamma^2)+\gamma_c(8\gamma^2+4\gamma_c-\nu^2)} \qquad (3.3.14)$$

We now study the following limiting cases in the laser bandwidth

(a) Broad-Band Source :

If $\gamma_c \longrightarrow \infty$ such that $\frac{\alpha^2}{\gamma_c}$ is a constant equal to say β, then we obtain after some simplifications

$$\langle I_{scat}\rangle^{(1)} = \frac{4a\beta\tau}{\sqrt{(\nu^2\tau^2+1)}}\cos(\nu t+\phi+\Phi) \qquad (3.3.15)$$

where

$$\tan \Phi = -\nu\tau \qquad (3.3.16)$$

Thus, the amplitude of modulation varies as $(\nu^2\tau^2+1)^{-\frac{1}{2}}$ while the phaseshift is given by (3.3.16), which are also the results of calculations for excitation with a broad-band, weak source that is weakly modulated.[1-5]

(b) Monochromatic Source :

$$\langle I_{scat}\rangle^{(1)} = \frac{16a\,\alpha^2\tau^2}{\sqrt{(4\nu^2\tau^2+1)}} \cos\,(\nu t + \phi + \Phi) \qquad (3.3.17)$$

where

$$\tan \Phi = -2\nu\tau \qquad (3.3.18)$$

Comparing equations (3.3.17) and (3.3.18) with the corresponding ones for the broad-band case, we find that for monochromatic lasers, the amplitude of modulation varies as $(\nu^2+\gamma^2)^{-\frac{1}{2}}$ instead of $(\nu^2+4\gamma^2)^{-\frac{1}{2}}$ as in the broad-band case, while the phaseshift differs by a factor two - a result that has also been obtained by Armstrong and Fenneuille[6]

(c) Intermediate - Bandwidth Source :

If the bandwidth of the exciting source is of the same order as the width of the excited state, i.e. $\gamma_c \sim \gamma$, then we find

$$\langle I_{scat}\rangle^{(1)} = \frac{2a\alpha^2\tau}{(4\gamma^2+\nu^2)^2}\,[\nu^2(\nu^2+12\gamma^2)^2+256\gamma^6]^{\frac{1}{2}} \cos(\nu t+\phi+\Phi) \qquad (3.3.19)$$

where

$$\tan \Phi = -\frac{3}{2} \nu \tau \left(1 + \frac{\nu^2 \tau^2}{3} \right) \qquad (3.3.20)$$

so that for small modulation frequencies ($\nu \tau \ll 1$), the plot of $\tan \Phi$ against ν is still linear, but with a slope $-\frac{3}{2} \tau$.

Hence it is clear that if the lifetime measurements are to be interpreted on the basis of the relationship between the phaseshift and modulation frequency, then one must take into account the bandwidth of the exciting source. Figure (5) shows the effect of the bandwidth of the laser on the phaseshift of the emitted light. For small modulation frequencies, the plot of $\tan \Phi$ against ν is linear for both large and small bandwidths, with slopes τ and 2τ, respectively, in accordance with equations (3.3.16) and (3.3.18). For a source with negligible bandwidth, the plot of $\tan \Phi$ vs ν remains linear even at high modulation frequencies. However, for a broadband source, we have from equation (3.3.14)

$$\tan \Phi \simeq -\nu \frac{\nu^2 + 2\gamma_c^2}{4\gamma\gamma_c^2 - \gamma_c \nu^2} \qquad (3.3.21)$$

so that the curve changes sign when $\nu^2 > 4\gamma\gamma_c$. Figure (6) displays the amplitude of modulation as a function of the modulation frequency for various values of the bandwidth. For a given value of the lifetime of the excited state, the peak

observed at $\nu = 0$ is broader for a larger bandwidth source, as expected from equations (3.3.16), (3.3.18) and (3.3.20).

(ii) Intense Fields : In the case of intense laser beams, the information given by (3.3.4) is not sufficient and one has to prescribe the arbitrary order correlation functions of the laser. The laser electric field is a classical random variable, but we now assume it to be very well stabilized in amplitude (an assumption that is well justified for a laser above threshold) while the phase is a fluctuating variable. We then write

$$\vec{E}(t) = \frac{1}{2} \vec{\mathcal{E}}_o \left[1 + a \cos (\nu t + \phi)\right] e^{-i\Omega t - i\theta(t)} + c.c. \quad (3.3.22)$$

where $\theta(t)$ is the random phase of the laser field. For the fluctuations of the field, we adopt the phase diffusion model,[87] as for this model, the correlation functions of the electric field are known to all orders. For this model, the amplitude is a deterministic variable independent of time, as in expression (3.3.22), while the phase undergoes diffusion.

$$\dot{\theta}(t) = \mu(t) \qquad \theta(o) = \theta_o \quad (3.3.23)$$

where θ_o is uniformly distributed between 0 and 2π, and $\mu(t)$ is a δ-correlated Gaussian random process with

$$\langle \mu(t) \rangle = 0 \ , \ \langle \mu(t_1)\, \mu(t_2) \rangle = 2\gamma_c\, \delta(t_1 - t_2) \quad (3.3.24)$$

where the angular brackets denote the ensemble average with respect to the distribution of the random process $\mu(t)$ and γ_c^{-1} is the correlation time for the laser phase fluctuations. The optical Bloch equations (3.1.14) now have the form

$$\langle \dot{\tilde{S}}^{+}(t) \rangle = (-\frac{1}{T_2} + i\Delta) \langle \tilde{S}^{+}(t) \rangle - 2i\alpha[1 + a\cos(\nu t + \phi)] e^{i\theta(t)} \langle \tilde{S}^{z}(t) \rangle$$

$$\langle \dot{\tilde{S}}^{z}(t) \rangle = -\frac{1}{T_1} \left(\langle \tilde{S}^{z}(t) \rangle - \langle S^{z} \rangle_{eq} \right) - i\alpha[1 + a\cos(\nu t + \phi)] \times$$

$$\left(e^{-i\theta(t)} \langle \tilde{S}^{+}(t) \rangle - \text{c.c.} \right) \qquad (3.3.25)$$

Equations (3.3.25) involve the phase $\theta(t)$ in a nonlinear fashion. By a redefinition of the variables, they can be cast into a linearized set of equations. On introducing the variables

$$\chi_1 = \chi_2^{*} = e^{-i\theta(t)} \langle \tilde{S}^{+}(t) \rangle , \quad \chi_3 = \langle \tilde{S}^{z}(t) \rangle , \quad \chi_4 = 1 \qquad (3.3.26)$$

We find the equations for χ as

$$\dot{\chi} = [B_o + a\cos(\nu t + \phi) M_o + i\mu(t) B_1] \chi \qquad (3.3.27)$$

where B_o, M_o and B_1 are the following matrices

$$B_o = \begin{bmatrix} -\frac{1}{T_2}+i\Delta & 0 & -2i\alpha & 0 \\ 0 & -\frac{1}{T_2}-i\Delta & 2i\alpha & 0 \\ -i\alpha & i\alpha & -\frac{1}{T_1} & \frac{\langle S^z\rangle_{eq}}{T_1} \\ 0 & 0 & 0 & 0 \end{bmatrix} , \quad (B_1)_{ij}=\delta_{ij}(\delta_{i2}-\delta_{i1})$$

$$(M_o)_{13} = 2(M_o)_{31} = -2i\alpha = -(M_o)_{23} = -2(M_o)_{32} \qquad (3.3.28)$$

and we have made use of (3.3.23) in deriving equation (3.3.27). This equation has the form of the standard equation of the multiplicative stochastic processes discussed in detail by Agarwal.[9-10] It follows that the average of χ over the distribution of $\theta(t)$ satisfies the equation

$$\dot{\langle\chi\rangle} = \begin{bmatrix} -\frac{1}{T_2}-\gamma_c+i\Delta & 0 & -2i\alpha & 0 \\ 0 & -\frac{1}{T_2}-\gamma_c-i\Delta & 2i\alpha & 0 \\ -i\alpha & i\alpha & -\frac{1}{T_1} & \frac{\langle S^z\rangle_{eq}}{T_1} \\ 0 & 0 & 0 & 0 \end{bmatrix} \langle\chi\rangle + a\cos(\upsilon t+\phi) M_o \langle\chi\rangle$$

(3.3.29)

where the angular brackets $\langle\ \rangle$ denote the ensemble average with respect to the distribution of the random process. Writing equations (3.3.29) in the form of the familiar Bloch equations, we have

$$\frac{\partial}{\partial t}\langle\langle \tilde{S}^{+} e^{-i\theta(t)} \rangle\rangle = (-\frac{1}{T_2} - \gamma_c + i\Delta)\ \langle\langle \tilde{S}^{+} e^{-i\theta(t)} \rangle\rangle$$

$$-2i\alpha\ [1+a\cos(\upsilon t+\phi)]\ \langle\langle \tilde{S}^{z} \rangle\rangle$$

$$\frac{\partial}{\partial t}\langle\langle \tilde{S}^{z} \rangle\rangle = -\frac{1}{T_1}\langle\langle \tilde{S}^{z} - S^{z}_{eq} \rangle\rangle - i\alpha\ [1+a\cos(\upsilon t+\phi)]\langle\langle \tilde{S}^{+} e^{-i\theta(t)} \rangle\rangle$$

$$+ i\alpha\ [1+a\cos(\upsilon t+\phi)]\ \langle\langle \tilde{S}^{-} e^{i\theta(t)} \rangle\rangle \qquad (3.3.30)$$

The above equations have the same structure as (3.1.14) but with $\frac{1}{T_2} \longrightarrow \frac{1}{T_2} + \gamma_c$. In view of this, the results (3.2.16) for the modulated fluorescence are valid in the presence of laser temporal fluctuations provided we make the replacement

$$\frac{1}{T_2} \longrightarrow \frac{1}{T_2} + \gamma_c$$

Section 3.4 : Velocity-averaged modulated fluorescence :

In all the previous sections, we have ignored the effects due to the finite velocity of the atom by assuming that the atom was at rest at the origin. To take into account the effect of Doppler broadening, we assume that the atom is moving with a uniform velocity $\vec{V}$ and that its position at some instant of time 't' is $\vec{r}$ ($= \vec{v}t$). The electric field (3.1.5) of the modulated

laser beam at the position of the atom is

$$\vec{E}(r,t) = \frac{1}{2}\vec{\mathcal{E}}_o[1+a\cos(\nu t+\phi)]\, e^{-i(\Omega+\vec{K}_L.\vec{V})t} + c.c. \qquad (3.4.1)$$

where $\vec{K}_L$ is the laser wave vector ($\kappa_L = \Omega/c$). The Bloch equations (3.1.14) are modified to

$$\langle\dot{\tilde{S}}^+(t)\rangle = \left(-\frac{1}{T_2}+i\tilde{\Delta}\right)\langle\tilde{S}^+(t)\rangle - 2i\alpha\,[1+a\cos(\nu t+\phi)]\,\langle\tilde{S}^z(t)\rangle$$

$$\langle\dot{\tilde{S}}^z(t)\rangle = -\frac{1}{T_1}\left(\langle\tilde{S}^z(t)\rangle - \langle S^z\rangle_{eq}\right) - i\alpha\,[1+a\cos(\nu t+\phi)]\left(\langle\tilde{S}^+(t)\rangle - c.c.\right) \qquad (3.4.2)$$

where

$$\tilde{\Delta} = \omega - \Omega - \vec{K}_L.\vec{V} \qquad (3.4.3)$$

and tilde now denotes a quantity in a frame rotating with angular frequency $(\Omega + \vec{K}_L.\vec{V})$ i.e.

$$\langle\tilde{S}^{\pm}(t)\rangle = \langle S^{\pm}(t)\rangle\, e^{\mp i(\Omega+\vec{K}_L.\vec{V})t} \qquad (3.4.4)$$

Equations (3.4.2) have the same structure as (3.1.14), except that we now have $\tilde{\Delta}$ instead of the laser detuning Δ. In view of this, all the previous results for the modulated fluorescence are still valid provided we make the replacement

$$\tilde{\Delta} = \Delta - \vec{K}_L.\vec{V} \qquad (3.4.5)$$

We still have to average the intensity of the modulated fluorescence, $I^{(1)}_{scat}$, over the velocity distribution. We assume that the atoms obey a Maxwellian velocity distribution so that the velocity averaged intensity, $\bar{I}^{(1)}_{scat}$, is given by

$$\bar{I}^{(1)}_{scat} = \frac{1}{\sqrt{\pi}\, v_m} \int_{-\infty}^{\infty} dv \quad e^{-v^2/v_m^2} \quad I^{(1)}_{scat} \tag{3.4.6}$$

where v is the component of the atomic velocity along the direction of the laser wave vector $\vec{K}_L$, and v_m is the most probable velocity equal to $v_m = (2K_BT/M)^{\frac{1}{2}}$ at temperature T. After integration over the velocity distribution, we will be able to study the modulated fluorescence line shape itself. This integration may be performed exactly with the use of Plasma Dispersion Function[88] which is frequently used in gas laser theories to yield explicit formulae for the line shape. As the expressions obtained are very cumbersome, we will not present them but will give the equivalent results obtained by numerical integration. We first study the limiting case of infinite Doppler width for weak fields (since analytical expressions can be obtained in this case) and then discuss the results from the numerical study. In view of (3.4.5), the weak field result (3.2.10) may be written as

$$X = \frac{1}{\frac{1}{T_1} + i\nu} \left[\frac{1}{\frac{1}{T_2} + i\tilde{\Delta}} + \frac{1}{\frac{1}{T_2} - i\tilde{\Delta}} + \frac{1}{\frac{1}{T_2} + i\nu + i\tilde{\Delta}} + \frac{1}{\frac{1}{T_2} + i\nu - i\tilde{\Delta}} \right] \tag{3.4.7}$$

On averaging over the velocity distribution, we obtain

$$\overline{\chi} = \frac{-i}{\sqrt{\pi} D\left(\frac{1}{T_1} + i\nu\right)} \int_{-\infty}^{\infty} dy\, e^{-y^2} \left[\frac{1}{y - \left(\frac{\Delta}{D} + \frac{i}{T_2 D}\right)} + \frac{1}{y - \left(\frac{-\Delta}{D} + \frac{i}{T_2 D}\right)} + \right.$$

$$\left. \frac{1}{y - \left(\frac{\Delta}{D} - \frac{\nu}{D} + \frac{i}{T_2 D}\right)} + \frac{1}{y - \left(\frac{-\Delta}{D} - \frac{\nu}{D} + \frac{i}{T_2 D}\right)} \right] \qquad (3.4.8)$$

where

$$y = v / v_m \quad , \quad D = K_L v_m \equiv \text{Doppler width} \qquad (3.4.9)$$

Equation (3.4.8) may be rewritten as

$$\overline{\chi} = \frac{-i}{\sqrt{\pi} D\left(\frac{1}{T_1} + i\nu\right)} \left[Z\left(\frac{\Delta}{D} + \frac{i}{T_2 D}\right) + Z\left(-\frac{\Delta}{D} + \frac{i}{T_2 D}\right) + \right.$$

$$\left. Z\left(\frac{\Delta - \nu}{D} + \frac{i}{T_2 D}\right) + Z\left(\frac{-\Delta - \nu}{D} + \frac{i}{T_2 D}\right) \right] \qquad (3.4.10)$$

where $Z(\mu)$ is the Plasma Dispersion Function defined by

$$Z(\mu) = \int_{-\infty}^{\infty} dy \, \frac{e^{-y^2}}{y - \mu} \quad , \quad \text{Im}\, \mu > 0 \qquad (3.4.11)$$

If $-\mu = \xi + i\eta$, then in the limit of small values of η, we have the following asymptotic expansion for $Z(\mu)$ to first order in η :

$$Z(\mu) \cong -2e^{-\xi^2}\int_0^{\xi} e^{y^2}dy + 2\sqrt{\pi}\eta\xi\, e^{-\xi^2} + i\left(-2\eta + \sqrt{\pi}e^{-\xi^2} + 4\eta\xi\, e^{-\xi^2}\int_0^{\xi} e^{y^2}dy\right)$$

$$= -2e^{-\xi^2}\int_0^{\xi} e^{y^2}dy + i\sqrt{\pi}\, e^{-\xi^2} \qquad (\eta \to 0) \qquad (3.4.12)$$

If in addition we have that $\xi \ll 1$, then equation (3.4.12) simply reduces to

$$Z(\mu) = i\sqrt{\pi}\, e^{-\xi^2} - 2\xi e^{-\xi^2} \qquad (3.4.13)$$

For the Plasma Dispersion Functions that occur in (3.4.10), we find that $\eta = \frac{1/T_2}{D}$, $\xi = \pm \frac{\Delta}{D}$ or $\frac{-\nu \pm \Delta}{D}$. In the Doppler limit, the homogeneous widths are much smaller than the Doppler widths i.e. $\frac{1}{T_2} \ll D$ $\left(\eta \ll 1\right)$. If in addition we further assume that the Dopper width is much larger than the laser detuning and modulation frequency i.e. $\nu, \Delta \ll D$ $\left(\xi \ll 1\right)$, then on using (3.4.13) in (3.4.10), we obtain the following results for $\overline{X}$

$$\overline{X} = \frac{-i}{\sqrt{\pi}D\left(\frac{1}{T_1} + i\nu\right)}\left\{ 2i\sqrt{\pi}\, e^{-\frac{\Delta^2}{D^2}} + e^{-\left(\frac{\Delta-\nu}{D}\right)^2}\left[i\sqrt{\pi} + 2\,\frac{\nu-\Delta}{D}\right] + e^{-\left(\frac{\Delta+\nu}{D}\right)^2}\left[i\sqrt{\pi} + 2\,\frac{\nu+\Delta}{D}\right]\right\} \qquad (3.4.14)$$

In the limit of infinite Doppler width, this reduces to

$$\overline{X} = \frac{4}{D\left(\frac{1}{T_1} + i\nu\right)}$$

so that the velocity averaged intensity is given by

$$\bar{I}^{(1)}_{scat} = -2a\alpha^2 \langle S^z \rangle_{eq} \frac{4}{D(\frac{1}{T_1^2} + \nu^2)^{\frac{1}{2}}} \cos(\nu t + \phi + \Phi) \quad (3.4.15)$$

where

$$\tan \Phi = -\nu T_1$$

$$A = \frac{4}{D(\frac{1}{T_1^2} + \nu^2)^{\frac{1}{2}}} \quad (3.4.16)$$

Comparing the equation with (3.3.17), we find that the width of the resonance in A at $\nu = 0$ is determined now by $1/T_1$ rather than $1/T_2$ as before. For radiative relaxation, the above results have the structure

$$\bar{I}^{(1)}_{scat} = \frac{4a\alpha^2\tau}{D} \frac{1}{(1+\nu^2\tau^2)^{\frac{1}{2}}} \cos(\nu t + \phi + \Phi) \quad (3.4.17)$$

where

$$\tan \Phi = -\nu\tau$$

Comparing equations (3.4.16) and (3.4.18) with (3.3.15) and (3.3.16), we find that for weak fields and infinite Doppler widths, the velocity-averaged amplitude of modulation and the phaseshift for a monochromatic source have the same structure as in the case of excitation by a weak, broad-band source.

Figure (7) is a normalized plot of the velocity averaged amplitude of modulation for various values of Doppler widths. The numerical plots reveal that when D is small (0.1γ to γ), the Doppler - broadened resonance is not significantly different from those corresponding to $\vec{v} = 0$, but for large values ($D \sim 10\gamma$), the peaks are much broader and displaced slightly for strong fields.

CHAPTER - IV

MODULATED HANLE SIGNALS IN PARTIALLY COHERENT FIELDS

The fluorescence emitted by atoms in the presence of a magnetic field[25-33] has been used to determine the Zeeman and hyperfine structures for the ground and excited state. These studies have also yielded radiative relaxation rates, since the fluorescence exhibits resonant behaviour as the static magnetic field is varied, the width of the resonance curve being determined by the natural lifetime of the atoms in the excited state. An important and useful variation of the usual Hanle effect consists in the excitation of atoms by light whose intensity is periodically modulated in time (the degeneracy of the energy levels having been lifted by a static magnetic field as before).[22,23] The intensity of the fluorescent light is itself modulated at the same frequency as the exciting light, while the amplitude of modulation exhibits resonant structures when the frequency of modulation is equal to the Zeeman splitting between the levels. The width of these resonances was once again determined by the natural lifetime of atoms in the excited state when the source used for preparing the atoms in a coherent superposition of the Zeeman sublevels was a weak, broad band lamp.

With the advent of high power, tunable dye lasers, it would be interesting to study the Hanle effect excited by a modulated laser beam of arbitrary intensity and bandwidth. A detailed study of this problem is carried out in this chapter. The shape as well as the linewidth of the resonances observed in the modulated fluorescence are shown to be critically dependent on the bandwidth of the exciting source and its frequency of modulation. The results of a previous study of the problem[22,23] using a weak, incoherent broad-band pump are obtained as a limiting case of our general theory, while excitation by a truly monochromatic laser leads to entirely new results. For intense fields, the amplitude of modulation exhibits resonances corresponding to the dynamic Stark splitting[7] of the various energy levels of the composite system consisting of the atom and the intense, coherent laser field.

Section 4.1 : Dynamical Equations :

We consider here the typical atomic beam Hanle-type experiment[25-33] shown in Figure (8) An atomic beam travels along the Z axis and a constant magnetic field H is applied in the same direction. The atoms are irradiated by a fluctuating laser beam (whose intensity is time modulated) propagating along Y-direction and having a linear polarization $\hat{X}$ parallel

to X-axis. We can detect the fluorescence emitted along the Z-axis with a linear polarization $\hat{X}$ (= L(x)) or the fluorescence emitted along the X-axis with a linear polarization Y (= L (y)). For simplicity, we restrict our study of the hanle effect to transitions between levels with angular momentum J=0 and J=1. The static magnetic field H splits the latter into three Zeeman sublevels with $m_J=0, \pm 1$. However, the plane polarized laser beam for our geometry couples the non-degenerate ground state $|J=0, m_J=0\rangle$ to the $m_J = \pm 1$ sublevels of the excited state (J=1) only, the laser being near resonance with the atomic transition $|J=0, m_J=0\rangle \longleftrightarrow |J=1, m_J=0\rangle$, which is forbidden. Thus we can neglect the existence of $m_J=0$ excited sublevel in further consideration and the relevant atomic levels are shown in Figure (9), where the labels 1, 2 and 3 refer respectively to the $|J=1, m_J=1\rangle$, $|J=1, m_J=-1\rangle$ and $|J=0, m_J=0\rangle$ levels, ' S ' is the Zeeman splitting or Larmor frequency of the excited state and 'δ' is the detuning of the laser beam of mean frequency Ω from the atomic transition $|J=0, m_J=0\rangle \leftrightarrow |J=1, m_J=0\rangle$.

The energy of the atomic states $|1\rangle$, $|2\rangle$ and $|3\rangle$ are ω_{13} $(=\Omega + S-\delta)$, $\omega_{23}(=\Omega - S-\delta)$ and zero respectively.

In order to take into account the temporal fluctuations present in the laser, we treat the laser electric field $\vec{E}(t)$ as a classical random variable. For a laser well above threshold, $\vec{E}(t)$ can be written as the following by making use of equations (2.1.7) and (3.3.22),

$$\vec{E}(t) = \frac{1}{2} \vec{\mathcal{E}}_0 [1+a \cos (\nu t + \phi)] \, e^{-i\Omega t - i\theta(t)} + c.c. \qquad (4.1.1)$$

where $\theta(t)$ is the random phase of the laser field undergoing diffusion as discussed in section (3.3), and the other symbols have the usual meaning defined in section (2.1). From (2.1.6), we find that the atomic dipole moment operator for this case may be written as

$$\hat{\vec{d}} = \vec{d}_{13} A_{13} + \vec{d}_{23} A_{23} + H.c. \qquad (4.1.2)$$

This leads to the following form for the interaction Hamiltonian between the atoms and the external field :

$$H_{ext}(t) = \alpha_1(t)[1+a \cos (\nu t+\phi)] \, (A_{13}+A_{31}) \, e^{-i\Omega t}$$

$$+ \alpha_2(t) \, [1+a \cos(\nu t+\phi)] \, e^{-i\Omega t} \, (A_{23}+A_{32}) + H.c. \qquad (4.1.3)$$

where

$$2\alpha_1(t) = -\vec{d}_{13} \cdot \vec{\mathcal{E}}_0 \, e^{-i\theta(t)}$$

$$2\alpha_2(t) = -\vec{d}_{23} \cdot \vec{\mathcal{E}}_0 \, e^{-i\theta(t)} \qquad (4.1.4)$$

substituting (4.1.3) in (2.2.7), we obtain the following equations for the evolution of the density matrix elements

$$\dot{\tilde{\rho}}_{11} = -2\gamma_1 \tilde{\rho}_{11} + \{-i\alpha_1(t)\,[1+a\cos(\upsilon t+\phi)]\,\tilde{\rho}_{31} + c.c.\}$$

$$\dot{\tilde{\rho}}_{12} = -(\gamma_1+\gamma_2+2is)\,\tilde{\rho}_{12} - i\alpha_1(t)\,[1+a\cos(\upsilon t+\phi)]\,\tilde{\rho}_{32} + i\alpha_2^*(t)\,[1+a\cos(\upsilon t+\phi)]\,\tilde{\rho}_{13}$$

$$\dot{\tilde{\rho}}_{13} = -[\gamma_1+i(s-\delta)]\,\tilde{\rho}_{13} + i\alpha_1(t)\,[1+a\cos(\upsilon t+\phi)]\,(-\tilde{\rho}_{33}+\tilde{\rho}_{11}) + i\alpha_2(t)\,[1+a\cos(\upsilon t+\phi)]\,\tilde{\rho}_{12}$$

$$\dot{\tilde{\rho}}_{22} = -2\gamma_2 \tilde{\rho}_{22} + \{-i\alpha_2(t)\,[1+a\cos(\upsilon t+\phi)]\,\tilde{\rho}_{32} + c.c.\}$$

$$\dot{\tilde{\rho}}_{23} = -[\gamma_2-i(s+\delta)]\,\tilde{\rho}_{23} + i\alpha_2(t)\,[1+a\cos(\upsilon t+\phi)]\,(\tilde{\rho}_{22}-\tilde{\rho}_{33}) + i\alpha_1(t)\,[1+a\cos(\upsilon t+\phi)]\,\tilde{\rho}_{21}$$

$$\dot{\tilde{\rho}}_{33} = 2\gamma_1 \tilde{\rho}_{11} + 2\gamma_2 \tilde{\rho}_{22} + \{i\alpha_1(t)\,[1+a\cos(\upsilon t+\phi)]\,\tilde{\rho}_{31} + i\alpha_2(t)\,[1+a\cos(\upsilon t+\phi)]\,\tilde{\rho}_{32} + cc.\} \qquad (4.1.5)$$

where the oscillations at optical frequencies have been removed by transforming to the slowly varying quantities

$$\tilde{\rho}_{ii} = \rho_{ii} \qquad \tilde{\rho}_{13} = \rho_{13}\, e^{i\Omega t}$$
$$\tilde{\rho}_{12} = \rho_{12} \qquad \tilde{\rho}_{23} = \rho_{23}\, e^{i\Omega t} \qquad (4.1.6)$$

and neglecting terms with $e^{\pm 2i\Omega t}$ (RWA). The parameter $2\gamma_1$ ($2\gamma_2$) represents the transition probability per unit time for spontaneously emitting a photon in the transition $|1\rangle \rightarrow |3\rangle$ ($|2\rangle \longrightarrow |3\rangle$). Equations (4.1.5) involve the phase $\theta(t)$ in a nonlinear fashion. By a redefinition of the variables, the equations may be written as a linearized set of equations. On introducing the variables

$$\sigma_1 = \tilde{\rho}_{11}\,,\ \sigma_2 = \tilde{\rho}_{12}\,,\ \sigma_3 = \tilde{\rho}_{13}\, e^{i\theta(t)}\,,\ \sigma_4 = \sigma_2^*$$
$$\sigma_5 = \tilde{\rho}_{22}\,,\ \sigma_6 = \tilde{\rho}_{23} e^{i\theta(t)}\,,\ \sigma_7 = \sigma_3^*\,,\ \sigma_8 = \sigma_6^* \qquad (4.1.7)$$

and the normalisation condition $\tilde{\rho}_{11} + \tilde{\rho}_{22} + \tilde{\rho}_{33} = 1$, the coupled equations (4.1.5) may be written in matrix form as

$$\dot{\sigma}(t) = [B - i\mu(t)C]\,\sigma(t) + I + a\cos(\nu t + \phi)\,[M\sigma(t) + I] \qquad (4.1.8)$$

where $\sigma(t)$ and I are the following column matrices of order 8 :

$$\sigma(t) = \begin{bmatrix} \sigma_1(t) \\ \sigma_2(t) \\ \sigma_3(t) \\ \sigma_4(t) \\ \sigma_5(t) \\ \sigma_6(t) \\ \sigma_7(t) \\ \sigma_8(t) \end{bmatrix}, \qquad I = \begin{bmatrix} 0 \\ 0 \\ -i\alpha_1 \\ 0 \\ 0 \\ -i\alpha_2 \\ i\alpha_1^* \\ i\alpha_2^* \end{bmatrix} \tag{4.1.9}$$

while B, C and M are the following 8x8 matrices :

$$\begin{bmatrix}
-2\gamma_1 & 0 & i\alpha_1^* & 0 & 0 & 0 & -i\alpha_1 & 0 \\
0 & -\gamma_1-\gamma_2-2is & i\alpha_2^* & 0 & 0 & 0 & 0 & -i\alpha_1 \\
2i\alpha_1 & i\alpha_2 & -\gamma_1-i(s-\delta) & 0 & i\alpha_1 & 0 & 0 & 0 \\
0 & 0 & 0 & -\gamma_1-\gamma_2+2is & 0 & i\alpha_1^* & -i\alpha_2 & 0 \\
0 & 0 & 0 & 0 & -2\gamma_2 & i\alpha_2^* & 0 & -i\alpha_2 \\
i\alpha_2 & 0 & 0 & i\alpha_1 & 2i\alpha_2 & -\gamma_2+i(s+\delta) & 0 & 0 \\
-2i\alpha_1^* & 0 & 0 & i\alpha_2^* & -i\alpha_1^* & 0 & -\gamma_1+i(s-\delta) & 0 \\
-i\alpha_2^* & -i\alpha_1^* & 0 & 0 & -2i\alpha_2^* & 0 & 0 & -\gamma-i(s+\delta)
\end{bmatrix}$$

$$C_{33} = C_{66} = -C_{77} = -C_{88} = -1$$

$$M_{ii} = 0 \quad , \; M_{ij} = B_{ij} \tag{4.1.10}$$

the remaining elements of B being zero. Equation (4.1.8) has the form of the standard equation of the multiplicative stochastic processes discussed in detail in references [9,10], from which it follows that the average of σ over the distribution of θ satisfies the exact equation

$$\langle \dot{\sigma} \rangle = B \langle \sigma \rangle - \gamma_c \Lambda \langle \sigma \rangle + I + a \cos (\upsilon t + \phi) (M \langle \sigma \rangle + I) \tag{4.1.11}$$

where

$$\Lambda_{ii} = 1 \text{ for } i = 3, 6, 7, 8 \tag{4.1.12}$$

the remaining elements of Λ being zero. Thus, the diagonal elements of the density matrix are unaffected by the phase fluctuations present in the laser beam, while the effect on the off-diagonal elements is to alter the decay rates as

$$\begin{aligned} \gamma_1 &\longrightarrow \gamma_1 + \gamma_c \\ \gamma_2 &\longrightarrow \gamma_2 + \gamma_c \end{aligned} \tag{4.1.13}$$

This is an example of the substitution rule which has been emphasized in the context of optical resonances in partially coherent fields by several workers [9-11]. As outlined in

section 3 of chapter II, we obtain the solutions of (4.1.11) within the framework of perturbation theory since the modulation is always assumed to be weak i.e. $a \ll 1$. The interaction with the laser beam will be treated exactly. From (2.2.19), we have the solutions at steady states to first order in the modulation rates

$$\langle \sigma(\infty) \rangle^{(1)} = \frac{1}{2} a \left[(i\nu - B - \gamma_c \Lambda)^{-1} e^{i(\nu t + \phi)} + (-i\nu - B - \gamma_c \Lambda)^{-1} e^{-i(\nu t + \phi)} \right.$$

$$\left. \times (M \langle \sigma(\infty) \rangle^{(o)} + I) \right. \qquad (4.1.14)$$

We are interested in the shape of the detection signals L(x) and L(y) obtained when one measures the total intensity of the modulated fluorescence along Z and X directions, with linear polarizations $\hat{X}$ and $\hat{Y}$ respectively. From (2.3.1), we have the positive frequency part of the spontaneously-emitted radiation field as

$$\vec{E}^{(+)}(\vec{r},t) = \vec{E}_o^{(+)}(\vec{r},t) - \frac{\omega_{13}^2}{c^2 r} [\hat{r} \times (\hat{r} \times \vec{d}_{13})] A_{31}(t - \frac{r}{c})$$

$$- \frac{\omega_{23}^2}{c^2 r} [\hat{r} \times (\hat{r} \times \vec{d}_{23})] A_{32}(t - \frac{r}{c}) \qquad (4.1.15)$$

By a proper choice of the phase of the wave function, the dipole matrix elements between a Zeeman sublevel $| J=1, m_J = \pm 1 \rangle$ and the ground state $| J=0, m_J=0 \rangle$ will have the structure

$$\vec{d}_{13} = R\,(\hat{X}-i\hat{Y}) \quad , \quad \vec{d}_{23} = R\,(\hat{X}+i\hat{Y}) \tag{4.1.16}$$

R being the radial matrix element. We are interested in the shape of the detection signals L(x) and L(y) obtained when one measures the total intensity of the modulated fluorescence along Z and X directions, with linear polarizations $\hat{X}$ and $\hat{Y}$ respectively. Substituting (4.1.16) in (4.1.15), we find that the positive frequency part of the radiation field in the Z(x) direction with polarization $\hat{X}(\hat{Y})$ is given by

$$E_{\hat{x}}^{(+)}(z,t) = E_{ox}^{(+)}(z,t) + \frac{\omega_{13}^2 R}{c^2 z} A_{31}\left(t - \frac{z}{c}\right) + \frac{\omega_{23}^2 R}{c^2 z} A_{32}\left(t - \frac{z}{c}\right)$$

$$E_{\hat{y}}^{(+)}(x,t) = E_{oy}^{(+)}(x,t) - \frac{i\omega_{13}^2 R}{c^2 X} A_{31}\left(t - \frac{x}{c}\right) + \frac{i\omega_{23}^2 R}{c^2 x} A_{32}\left(t - \frac{x}{c}\right) \tag{4.1.17}$$

The corresponding intensity of the fluorescent light with $\hat{X}(\hat{Y})$ polarization is

$$I_{\text{seat}}(\hat{X}) = \frac{\omega_{13}^4 R^2}{c^4 z^2}\left\langle A_{11}\left(t - \frac{z}{c}\right)\right\rangle + \frac{\omega_{23}^4 R^2}{c^4 z^2}\left\langle A_{22}\left(t - \frac{z}{c}\right)\right\rangle + \frac{\omega_{13}^2 \omega_{23}^2 R^2}{c^4 z^2}$$

$$\times\left[\left\langle A_{12}\left(t - \frac{z}{c}\right)\right\rangle + \left\langle A_{21}\left(t - \frac{z}{c}\right)\right\rangle\right]$$

$$I_{\text{seat}}(\hat{Y}) = \frac{\omega_{13}^4 R^2}{c^4 x^2}\left\langle A_{11}\left(t - \frac{x}{c}\right)\right\rangle + \frac{\omega_{23}^4 R^2}{c^4 x^2}\left\langle A_{22}\left(t - \frac{x}{c}\right)\right\rangle - \frac{\omega_{13}^2 \omega_{23}^2 R^2}{c^4 x^2}$$

$$\times\left[\left\langle A_{12}\left(t - \frac{x}{c}\right)\right\rangle + \left\langle A_{21}\left(t - \frac{x}{c}\right)\right\rangle\right] \tag{4.1.18}$$

Now it is obvious from figure (9) that

$$\omega_{23} = \omega_{13}\left(1-\frac{2s}{\omega_{13}}\right) \qquad (4.1.19)$$

In our study, the maximum value assigned to the Zeeman splitting is 10γ, where γ is typically of the order of a few tens of MHz. Hence, $\frac{s}{\omega_{13}} \sim 10^{-6}$, which is a negligibly - small quantity, so that $\omega_{23} \sim \omega_{13}$. In view of this, the above signals at steady state may be written as ($I_{scat} \equiv L$)

$$L(x) \propto \rho_{11} + \rho_{22} + 2\mathrm{Re}\ \rho_{12}$$

$$L(y) \propto \rho_{11} + \rho_{22} - 2\mathrm{Re}\ \rho_{12} \qquad (4.1.20)$$

Using equations (4.1.7), we may write the signals averaged over the laser phase fluctuations as

$$L(x) \propto \langle\sigma_1\rangle + \langle\sigma_5\rangle + 2\ \mathrm{Real}\ \langle\sigma_2\rangle$$

$$L(y) \propto \langle\sigma_1\rangle + \langle\sigma_5\rangle - 2\ \mathrm{Real}\ \langle\sigma_2\rangle \qquad (4.1.21)$$

Thus the signals are proportional to the populations in the Zeeman sublevels $|1\rangle$, $|2\rangle$ and to the Zeeman coherence between these two levels. Since these occur as the first, fifth

and second elements of the column vector $\langle \sigma \rangle$ defined by (4.1.9), we have from (4.1.14)

$$\langle \rho_{11}(\infty) \rangle^{(1)} = \tfrac{1}{2} a \Big\{ e^{i(\nu t+\phi)} \sum_{i,j=1}^{8} (i\nu - B - \gamma_c \Lambda)^{-1}_{1i} \times [M_{ij} \langle \sigma_j(\infty) \rangle^{(o)} + I_i] + c.c. \Big\}$$

$$\langle \rho_{22}(\infty) \rangle^{(1)} = \tfrac{1}{2} a \Big\{ e^{i(\nu t+\phi)} \sum_{i,j=1}^{8} (i\nu - B - \gamma_c \Lambda)^{-1}_{5i} \times [M_{ij} \langle \sigma_j(\infty) \rangle^{(o)} + I_i] + c.c. \Big\}$$

$$\langle \rho_{12}(\infty) \rangle^{(1)} = \tfrac{1}{2} a \Big\{ e^{(i(\nu t+\phi)} \sum_{i,j=1}^{8} (i\nu - B - \gamma_c \Lambda)^{-1}_{2i} \times [M_{ij} \langle \sigma_j(\infty) \rangle^{(o)} + I_i]$$

$$+ e^{-i(\nu t+\phi)} \sum_{i,j=1}^{8} (-i\nu - B - \gamma_c \Lambda)^{-1}_{2i} [M_{ij} \langle \sigma_j(\infty) \rangle^{(o)} + I_i] \qquad (4.1.22)$$

For general values of the parameters occuring in the problem, the solutions (4.1.22) require the inversion of 8x8 matrices

which is cumbersome. We will obtain analytical expressions for the intensity of the modulated fluorescence in steady states for weak laser fields in the next section. The numerical studies for the intense field case will be presented in section 3.

Section 4.2 : Weak Fields :

In this section, we consider the optical source used to prepare the atom in a coherent superposition of the Zeeman sublevels as a weak, partially coherent one. From (4.1.14), we have the steady-state solutions to first order in modulation as

$$\langle\sigma(\infty)\rangle^{(1)} = \frac{1}{2}\, a\Big\{\Big[(i\nu - B_o)^{-1}\, e^{i(\nu t+\phi)} + (-i\nu - B_o)^{-1}\, e^{-i(\nu t+\phi)}\Big] \times \left(M\langle\sigma(\infty)\rangle^{(o)} + I\right)\Big\} \qquad (4.2.1)$$

where

$$B_o = B - \gamma_c \Lambda \qquad (4.2.2)$$

Till now, no approximation has been made regarding the atom-laser coupling and solutions (4.2.1) are valid to all orders in the Rabi frequency. We now assume the exciting source to be weak i.e. $\frac{\alpha}{\gamma} \ll 1$, and study the signals which are linear in the laser intensity i.e. to second order in α. We write the matrix B_o as the sum of the following two matrices

$$B_o = M + N \tag{4.2.3}$$

where

$$N_{ii} = (B_o)_{ii} \tag{4.2.4}$$

all the other elements of N being zero. The matrix M has been defined in the previous section by (4.1.10). It is obvious that the elements of the diagonal matrix N involve the various decay constants, Zeeman splitting and laser detuning only, while the off-diagonal matrix M has only the Rabi frequency as its elements. Since we have assumed that the Rabi frequency is much smaller than the decay constants etc., it follows that $M_{ij} \ll N_{ij}$. Hence we may expand the matrix inverse occuring in the solutions (4.2.1) to first order in the elements of M (i.e. to first order in the Rabi frequency)

$$(\pm i\nu - B_o)^{-1} = (\pm i\nu - M - N)^{-1}$$

$$\approx (\pm i\nu - N)^{-1} + (\pm i\nu - N)^{-1} M (\pm i\nu - N)^{-1} \tag{4.2.5}$$

Substituting (4.2.5) in (4.2.1), we obtain

$$\langle \sigma(\infty) \rangle^{(1)} \cong \tfrac{1}{2}a[(i\nu - N)^{-1} + (i\nu - N)^{-1} M (i\nu - N)^{-1}] (M \langle \sigma(\infty) \rangle^{(o)} + I)$$

$$+ \tfrac{1}{2}a [(-i\nu - N)^{-1} + (-i\nu - N)^{-1} M (-i\nu - N)^{-1}] (M \langle \sigma(\infty) \rangle^{(o)} + I)$$

(4.2.6)

Finding the inverse of the 8x8 matrices occuring in the above expression analytically presents no problem since these are diagonal matrices and hence

$$(\pm i\nu - N)^{-1}_{ij} = (\pm i\nu - N_{ii})^{-1} \delta_{ij} \qquad (4.2.7)$$

The steady state solutions occuring in (4.2.6) in the absence of modulation viz. $\langle\sigma(\infty)\rangle^{(o)}$ may be expanded in a power series with the Rabi frequency as the perturbation parameter. Indicating the order of expansion with respect to α as a subscript, we have

$$\langle\sigma(\infty)\rangle^{(o)} = \langle\sigma(\infty)\rangle^{(o)}_{(o)} + \langle\sigma(\infty)\rangle^{(o)}_{(1)} + \langle\sigma(\infty)\rangle^{(o)}_{(2)} + \cdots (4.2.8)$$

By substituting (4.2.8) in (4.2.6) and remembering that the matrix elements of M and I are linear in the Rabi frequency, we have

$$\langle\sigma(\infty)\rangle^{(1)}_{(2)} = \frac{a}{2} e^{i\nu t} \{ (i\nu - N)^{-1} M \langle\sigma(\infty)\rangle^{(o)}_{(1)} + (i\nu - N)^{-1} M (i\nu - N)^{-1} \times [M \langle\sigma(\infty)\rangle^{(o)}_{(o)} + I] \}$$

$$+ \frac{a}{2} e^{-i\nu t} \{ (-i\nu - N)^{-1} M \langle\sigma(\infty)\rangle^{(o)}_{(1)} + (-i\nu - N)^{-1} M (-i\nu - N)^{-1} \times [M \langle\sigma(\infty)\rangle^{(o)}_{(o)} + I] \} \qquad (4.2.9)$$

Now the coupled equations in the absence of modulation may be obtained from (4.1.11) by putting a = o.

We have

$$\langle \dot{\sigma}_1 \rangle = -2\gamma \langle \sigma_1 \rangle + i\alpha (\langle \sigma_3 \rangle - \langle \sigma_7 \rangle)$$

$$\langle \dot{\sigma}_2 \rangle = -(2\gamma+2is) \langle \sigma_2 \rangle + i\alpha (\langle \sigma_3 \rangle - \langle \sigma_8 \rangle)$$

$$\langle \dot{\sigma}_3 \rangle = -[\gamma+\gamma_c+i(s-\delta)] \langle \sigma_3 \rangle + i\alpha(2 \langle \sigma_1 \rangle + \langle \sigma_2 \rangle + \langle \sigma_5 \rangle - 1)$$

$$\langle \dot{\sigma}_5 \rangle = -2\gamma \langle \sigma_5 \rangle + i\alpha (\langle \sigma_6 \rangle - \langle \sigma_8 \rangle)$$

$$\langle \dot{\sigma}_6 \rangle = -[\gamma+\gamma_c-i(s+\delta)] \langle \sigma_6 \rangle + i\alpha(\langle \sigma_7 \rangle + 2 \langle \sigma_5 \rangle + \langle \sigma_4 \rangle - 1)$$

(4.2.10)

From (4.2.10), it at once follows that

$$\langle \sigma_i(\infty) \rangle_{(o)} = 0 \quad \text{for all } i \tag{4.2.11}$$

Using this result in (4.2.9), we obtain

$$\langle \sigma(\infty) \rangle^{(1)}_{(2)} = \frac{a}{2} e^{i\nu t} \{ (i\nu - N)^{-1} M \langle \sigma(\infty) \rangle^{(o)}_{(1)} + (i\nu - N)^{-1} M (i\nu - N)^{-1} I \}$$

$$+ \frac{a}{2} e^{-i\nu t} \{ (-i\nu - N)^{-1} M \langle \sigma(\infty) \rangle^{(o)}_{(1)} + (-i\nu - N)^{-1} M (-i\nu - N)^{-1} I \}$$

(4.2.12)

We still have to obtain the first-order steady-statc solutions in the absence of modulation. These are easily obtained from (4.2.10) as

$$\langle \sigma_3(\infty) \rangle_{(1)} = \frac{-i\alpha}{\gamma+\gamma_c+i(s-\delta)} \qquad \langle \sigma_7(\infty) \rangle_{(1)} = \langle \sigma_3(\infty) \rangle^*_{(1)}$$

$$\langle \sigma_6(\infty) \rangle_{(1)} = \frac{-i\alpha}{\gamma+\gamma_c-i(s+\delta)} \qquad \langle \sigma_8(\infty) \rangle_{(1)} = \langle \sigma_6(\infty) \rangle^*_{(1)}$$

$$\langle \sigma_1(\infty) \rangle_{(1)} = \langle \sigma_2(\infty) \rangle_{(1)} = \langle \sigma_5(\infty) \rangle_{(1)} = 0 \qquad (4.2.13)$$

From (4.1.7), we know that $\langle \tilde{\rho}_{11} \rangle$, $\langle \tilde{\rho}_{22} \rangle$ and $\langle \tilde{\rho}_{12} \rangle$ occur as the first, fifth and second element of $\langle \sigma(t) \rangle$ respectively. Hence the desired solutions are obtained as

$$\langle \tilde{\rho}_{11}(\infty) \rangle^{(1)}_2 = \frac{a}{2} e^{i\nu t} (i\nu - N)^{-1}_{11} \sum_{j=1}^{8} \{ M_{1j} \langle \sigma_j(\infty) \rangle^{(0)}_{(1)} + M_{1j} (i\nu - N)^{-1}_{jj} I_j \} + c.c.$$

$$= \frac{a\alpha^2}{2} \frac{e^{i\nu t}}{2\gamma+i\nu} \left[\frac{1}{\gamma+\gamma_c+i(s-\delta)} + \frac{1}{\gamma+\gamma_c-i(s-\delta)} + \frac{1}{\gamma+\gamma_c+i(\nu+s-\delta)} + \frac{1}{\gamma+\gamma_c+i(\nu-s+\delta)} \right]$$

$$\langle \tilde{\rho}_{22}(\infty)\rangle^{(1)}_{(2)} = \frac{a}{2}\, e^{i\nu t}\,(i\nu - N)^{-1}_{55} \sum_{j=1}^{8}\Big\{ M_{5j}\langle \sigma_j(\infty)\rangle^{(o)}_{(1)} + M_{5j}\,(i\nu - N)^{-1}_{jj}\; I_j \Big\} + c.c.$$

$$= \frac{a\alpha^2}{2}\;\frac{e^{i\nu t}}{2\gamma + i\nu}\Big\{ \frac{1}{\gamma+\gamma_c + i(s+\delta)} + \frac{1}{\gamma+\gamma_c - i(s+\delta)} + \frac{1}{\gamma+\gamma_c + i[\nu + (s+\delta)]}$$

$$+ \frac{1}{\gamma+\gamma_c + i[\nu - (s+\delta)]}\Big\} + c.c.$$

$$\langle \tilde{\rho}_{12}(\infty)\rangle^{(1)}_{(2)} = \frac{a}{2}\, e^{i\nu t}(i\nu - N)^{-1}_{22} \sum_{j=1}^{8}\Big\{ M_{2j}\langle \sigma_j(\infty)\rangle^{(o)}_{(1)} + M_{2j}(i\nu - N)^{-1}_{jj}\, I_j\Big\}$$

$$+ \frac{a}{2}\, e^{-i\nu t}(-i\nu - N)^{-1}_{22} \sum_{j=1}^{8}\Big\{ M_{2j}\langle \sigma_j(\infty)\rangle^{(o)}_{(1)} + M_{2j}(-i\nu - N)^{-1}_{jj}\, I_j\Big\}$$

$$= \frac{a\alpha^2}{4}\;\frac{e^{i\nu t}}{\gamma + i(s + \frac{\nu}{2})}\Big[\; \frac{1}{\gamma+\gamma_c + i(s+\delta)} + \frac{1}{\gamma+\gamma_c + i(s-\delta)}$$

$$+ \frac{1}{\gamma+\gamma_c + i[\nu + (s+\delta)]} + \frac{1}{\gamma+\gamma_c + i[\nu + (s-\delta)]}\Big]$$

$$+ \frac{a\alpha^2}{4}\;\frac{e^{-i\nu t}}{\gamma + i(s - \frac{\nu}{2})}\Big[\; \frac{1}{\gamma+\gamma_c + i(s+\delta)} + \frac{1}{\gamma+\gamma_c + i(s-\delta)}$$

$$+ \frac{1}{\gamma+\gamma_c - i[\nu - (s+\delta)]} + \frac{1}{\gamma+\gamma_c - i[\nu - (s-\delta)]}\Big] \qquad (4.2.14)$$

The fluorescent light will thus be modulated at the same frequency as the exciting light, while the amplitude of modulation will exhibit resonances. The modulated fluorescence may be observed with the phase sensitive detector, which enables the cosine and sine component of the fluorescence to be studied separately. The resonances in the amplitude of the sine or cosine component may be studied by varying the magnetic field (hence the Larmor frequency or Zeeman splitting of the excited state) for a fixed frequency of modulation, or by changing the modulation frequency of the exciting source for a fixed strength of the static magnetic field. We first study the conventional magnetic field scan of the modulated Hanle signals.

[I] Magnetic Field Scan of Signals :

Further simplification of (4.2.14) for the case of resonant excitation (δ=o) yields

$$L\begin{pmatrix} x \\ y \end{pmatrix} = 2a\,\alpha^2 \cos\nu t \Big[\Big(\frac{4\gamma}{4\gamma^2+\nu^2} \mp \frac{4\gamma_c}{4\gamma_c^2+\nu^2} \Big) \mathcal{L}_o + \Big(\frac{2\gamma}{4\gamma^2+\nu^2} \mp \frac{2\gamma_c}{4\gamma_c^2+\nu^2} \Big)(\mathcal{L}_1+\mathcal{L}_2)$$

$$- \Big(\frac{\nu}{4\gamma^2+\nu^2} \mp \frac{\nu}{4\gamma_c^2+\nu^2} \Big)(\mathcal{S}_1-\mathcal{S}_2) \pm \frac{4\gamma_c}{4\gamma_c^2+\nu^2}(\mathcal{L}_3+\mathcal{L}_4) \Big]$$

$$+ 2a\alpha^2 \sin\nu t \left[\left(\frac{2\nu}{4\gamma^2+\nu^2} \pm \frac{2\nu}{4\gamma_c^2+\nu^2} \right) \mathcal{L}_0 + \left(\frac{\nu}{4\gamma^2+\nu^2} \mp \frac{\nu}{4\gamma_c^2+\nu^2} \right) (\mathcal{L}_1 + \mathcal{L}_2) \right.$$

$$\left. + \left(\frac{2\gamma}{4\gamma^2+\nu^2} \mp \frac{2\gamma_c}{4\gamma_c^2+\nu^2} \right) (\mathcal{D}_1 - \mathcal{D}_2) \pm \frac{4\gamma_c}{4\gamma_c^2+\nu^2} (\mathcal{D}_3 - \mathcal{D}_4) \right] \tag{4.2.15}$$

where

$$\mathcal{L}_0 = \frac{\gamma+\gamma_c}{(\gamma+\gamma_c)^2+ s^2} \qquad \mathcal{D}_0 = \frac{s}{(\gamma+\gamma_c)^2 + s^2}$$

$$\mathcal{L}_1 = \frac{\gamma+\gamma_c}{(\gamma+\gamma_c)^2+(s+\nu)^2} \qquad \mathcal{D}_1 = \frac{s+\nu}{(\gamma+\gamma_c)^2+ (s+\nu)^2}$$

$$\mathcal{L}_2 = \frac{\gamma+\gamma_c}{(\gamma+\gamma_c)^2+(s-\nu)^2} \qquad \mathcal{D}_2 = \frac{s-\nu}{(\gamma+\gamma_c)^2+(s-\nu)^2}$$

$$\mathcal{L}_3 = \frac{\gamma}{\gamma^2+(s+\nu/2)^2} \qquad \mathcal{D}_3 = \frac{s+\nu/2}{\gamma^2+(s+\nu/2)^2}$$

$$\mathcal{L}_4 = \frac{\gamma}{\gamma^2+(s-\nu/2)^2} \qquad \mathcal{D}_4 = \frac{s-\nu/2}{\gamma^2+ (s-\nu/2)^2} \tag{4.2.16}$$

Thus, there are resonances in the amplitude of the sine and cosine component of the modulated fluorescence at field values ($s = gH$) $H = 0, \pm \frac{\nu}{2g}, \pm \frac{\nu}{g}$ where g is the gyromagnetic ratio.. The width of the resonances at field value $0, \pm \frac{\nu}{g}$ is affected by the bandwidth of the fluctuating laser

beam, the laser bandwidth (γ_c) simply adding to half the radiative bandwidth (γ) to give the width ($\gamma+\gamma_c$) of these resonances. However, the resonances obtained when the modulation frequency is equal to the separation between the Zeeman sublevels ($\upsilon = \pm 2s$) have a width γ associated with the spontaneous decay of the excited state only. As may be seen from (4.2.15), the weight factor of each resonant structure depends critically on the bandwidth of the exciting source, besides its frequency of modulation. It should be borne in mind that one expects additional resonances in higher orders of perturbation theory i.e. terms of order higher than a. The resonances in the modulated signals could be understood as follows - The resonances at $s = \pm\upsilon$ essentially arise due to the absorption of photon with frequency $\Omega \pm \upsilon$. Note that the intensity of emission has contributions from the populations ρ_{11} and ρ_{22} of the excited states and the population of the excited states is given by the absorption line shape, which is known to have a width ($\gamma+\gamma_c$) in the limit of weak fields, [cf. the damping term in the $\langle\sigma_3\rangle$ equation (4.1.11)]. Therefore, the modulated signals at $s = \pm\upsilon$ have absorptive and dispersive type of contributions with width ($\gamma+\gamma_c$). The interference of the probability amplitudes

ıor the absorption of $\Omega+\upsilon$ and $\Omega-\upsilon$ photons produces the resonance at s = o with width $(\gamma+\gamma_c)$. The resonances at $s = \pm \upsilon/2$ arise due to the interference term $\mathrm{Re}\,\tilde{\rho}_{12}$ in the intensity of fluorescence, where $\mathrm{Re}\,\tilde{\rho}_{12}$ represents the optical coherence, between the two magnetic sublevels, induced by the exciting field. In the rotating frame $\mathrm{Re}\,\tilde{\rho}_{12}$ has the oscillation frequency 2s and the damping parameter associated with $\tilde{\rho}_{12}$ is 2γ. Therefore this leads to extra resonances at $s = \pm \upsilon/2$ with width γ. We now consider the following limiting cases explicitly.

(i) Broad Band Excitation :

If the optical source used to prepare the atoms in a coherent superposition of the Zeeman sublevel is a weak, incoherent broad-band lamp such as a discharge tube ($\gamma_c \to \infty$, such that a^2/γ_c remains a constant equal to say, β), then (4.2.15) reduces to

$$L\begin{pmatrix} x \\ y \end{pmatrix} = 4a\beta \cos\upsilon t \left\{ \frac{4\gamma}{4\gamma^2+\upsilon^2} \pm \frac{1}{2}\left[\frac{\gamma}{\gamma^2+(s+\upsilon/2)^2} + \frac{\gamma}{\gamma^2+(s-\upsilon/2)^2} \right] \right\}$$

$$+ 4a\beta \sin\upsilon t \left\{ \frac{2\upsilon}{4\gamma^2+\upsilon^2} \pm \frac{1}{2}\left[\frac{s+\upsilon/2}{\gamma^2+(s+\upsilon/2)^2} - \frac{s-\upsilon/2}{\gamma^2+(s-\upsilon/2)^2} \right] \right\}$$

(4.2.17)

which agrees with the results of Corney and Series[22] for signals detected perpendicular to H (which corresponds to the latter choice of sign in expression (4.2.17), the incident light being linearly polarized at right angles to the magnetic field direction. The result was later verified experimentally by Corney[23]. Thus, as the magnetic field is scanned around zero value, resonances appear in the amplitude of modulation for field values $H = \pm \frac{\nu}{2g}$, the shape of these resonances being Lorentzian (dispersion) for the cosine (sine) component of the fluorescence, the width being determined by the natural width of the excited state. In general for small frequencies of modulation, the resonances at $s = \pm \frac{\nu}{2}$ in the amplitude of the cosine or sine component overlap. Use of higher frequency of modulation of the exciting source ($\frac{\nu}{\gamma} = 10$, as in figure 10-13, results in well defined lorentzian (dispersion) shaped resonances at $s = \pm \frac{\nu}{2} = \pm 5$ for the cosine (sine) component of the fluorescence detected parallel or perpendicular to the magnetic field when $\frac{\gamma_c}{\gamma} \gg 1$ (see the dot - dash curves of figures 10-13, which correspond to $\gamma_c = 5\gamma$). The signals detected along the two directions are similar in shape but inverted about the horizontal axis (compare figure 10 with 12, figure 11 with 13), as may also be seen from (4.2.17), this result having been confirmed experimentally by Corney[23].

(ii) Monochromatic Excitation :

For a strictly monochromatic laser beam ($\gamma_c=0$), expression (5.2.15) simplifies to :

$$L\begin{pmatrix}x\\y\end{pmatrix}= 2a\hat{\alpha}^2\cos\upsilon t\left[\frac{4\gamma}{4\gamma^2+\upsilon^2}\mathcal{L}_0 + \frac{2\gamma}{4\gamma^2+\upsilon^2}(\mathcal{L}_1+\mathcal{L}_2) - \left(\frac{\upsilon}{4\gamma^2+\upsilon^2} \mp \frac{1}{\upsilon}\right)(\mathcal{D}_1-\mathcal{D}_2\right.$$

$$+ 2a\alpha^2 \sin\upsilon t\left[\left(\frac{2\upsilon}{4\gamma^2+\upsilon^2} \pm \frac{2}{\upsilon}\right)\mathcal{L}_0+\left(\frac{\upsilon}{4\gamma^2+\upsilon^2} \mp \frac{1}{\upsilon}\right)(\mathcal{L}_1+\mathcal{L}_2) + \frac{2\gamma}{4\gamma^2+\upsilon^2}(\mathcal{D}_1-\mathcal{D}_2)\right]$$

(4.2.18)

Hence, in contrast to the broad band excitation, we expect resonances in the magnetic field scan of the modulated fluorescence whenever $s=0, \pm\upsilon$, the resonance at $s = \pm\upsilon/2$ disappears. As we have shown earlier that this coherence arises from $\mathrm{Re}\,\tilde{\rho}_{12}$- which is a measure of optical coherence between two magnetic sublevels, and it can be shown to be directly proportional to the bandwidth of the exciting field. The resonance at $s=0$ is a Lorentzian, while the resonances at $s = \pm\upsilon$ have both a Lorentzian and dispersion part with different weight factors. When $\frac{\upsilon}{\gamma} = 10$ (fig. 10-13) the resonances are clearly resolved. Use of higher value of modulation frequency also has the added advantage of making the weight factors of the Lorentzian and dispersion shaped

parts of the resonances at $s = \pm \upsilon$ very different from each other, so that either one of them is predominantly present at $s = \pm \upsilon$, thus making the interpretation of results easy. The signals detected along and perpendicular to the magnetic field are no longer similar in shape for a particular component of the modulated fluorescence. For instance, the resonance at $s = \upsilon$ in the amplitude of the cosine component is a Lorentzian for L(x) (solid curve of figure 10) and is predominantly dispersion shaped for L(y) (solid curve of figure 12), as may also be verified from (4.2.18).

(iii) Intermediate Case :

For an optical source with bandwidth of the same order of magnitude as the radiative width of the excited state ($\gamma_c \sim \gamma$), the expressions (4.2.15) for the signals simplify to :

$$L(x) \propto \cos \upsilon t \ \frac{4\gamma}{4\gamma^2+\upsilon^2}(\mathcal{L}_3+\mathcal{L}_4) + \sin \upsilon t \left[\frac{4\upsilon}{4\gamma^2+\upsilon^2}\mathcal{L}_0 + \frac{4\gamma}{4\gamma^2+\upsilon^2}(\mathcal{S}_3-\mathcal{S}_4)\right]$$

$$L(y) \propto \cos \upsilon t \left\{\frac{8\gamma}{4\gamma^2+\upsilon^2}\left[\mathcal{L}_0+\tfrac{1}{2}(\mathcal{L}_1+\mathcal{L}_2)-\tfrac{1}{2}(\mathcal{L}_3+\mathcal{L}_4\right] - \frac{2\upsilon}{4\gamma^2+\upsilon^2}(\mathcal{S}_1-\mathcal{S}_2)\right\}$$

$$+ \sin \upsilon t \left\{\frac{2\upsilon}{4\gamma^2+\upsilon^2}(\mathcal{L}_1+\mathcal{L}_2) + \frac{4\gamma}{4\gamma^2+\upsilon^2}(\mathcal{S}_1-\mathcal{S}_2-\mathcal{S}_3+\mathcal{S}_4)\right\}$$

(4.2.19)

Thus, for signals detected along the magnetic field direction the amplitude of the cosine component has a Lorentzian shaped resonance at $s = \pm \frac{\upsilon}{2}$ (broken curve of figure 10), while for the sine component, there are resonances at $s = 0, \pm \frac{\upsilon}{2}$, the weight of the Lorentzian peak at $s = 0$ (broken curve of figure 11) being much greater than the dispersion shaped resonance at $s = \pm \frac{\upsilon}{2}$ for large frequencies of modulation. Magnetic field scan of fluorescence detected perpendicular to H reveal resonant structure corresponding to $s = 0, \pm \frac{\upsilon}{2}$, $\pm \upsilon$ with considerable overlap even for $\frac{\upsilon}{\gamma} = 10$, as may be seen from the broken curves of figures 12 and 13 .

[II] Modulation Frequency Scan of Signals :

We have seen that when atoms whose excited states have Zeeman structures are excited by light whose intensity is modulated in time, then the fluorescent light is itself modulated at the same frequency, while the amplitude of modulation exhibits resonances. These resonances may also be studied by tuning the modulation frequency of the exciting source for a fixed strength of the static magnetic field. Simplification of Eqs. (4.2.14) for the case of resonant excitation yields :

$$L\begin{pmatrix} x \\ y \end{pmatrix} = 2a\alpha^2 \cos \upsilon t \left\{ \left[\frac{\gamma+\gamma_c}{(\gamma+\gamma_c)^2+s^2} - \frac{\gamma-\gamma_c}{(\gamma-\gamma_c)^2+s^2} \right] \left[2\mathcal{L}'_0 \pm (\mathcal{L}'_3 + \mathcal{L}'_4) \right] \right.$$

$$\mp \left[\frac{s}{(\gamma+\gamma_c)^2+s^2} - \frac{s}{(\gamma-\gamma_c)^2+s^2}\right](\mathcal{B}_3'-\mathcal{B}_4') + \frac{\gamma-\gamma_c}{(\gamma-\gamma_c)^2+s^2}\left[\mathcal{L}_1'+\mathcal{L}_2' \pm (\mathcal{L}_1'+\mathcal{L}_2')\right.$$

$$\left. + \frac{s}{(\gamma-\gamma_c)^2+s^2}\left[\mathcal{B}_1'-\mathcal{B}_2' \mp (\mathcal{B}_1'-\mathcal{B}_2')\right]\right\}$$

$$\begin{matrix}+ \\ - \end{matrix} 2a\alpha^2 \sin\upsilon t\left\{\left[\frac{\gamma+\gamma_c}{(\gamma+\gamma_c)^2+s^2} - \frac{\gamma-\gamma_c}{(\gamma-\gamma_c)^2+s^2}\right]\left[2\mathcal{B}_0' \pm (\mathcal{B}_3'+\mathcal{B}_4')\right]\right.$$

$$\pm \left[\frac{s}{(\gamma+\gamma_c)^2+s^2} - \frac{s}{(\gamma-\gamma_c)^2+s^2}\right](\mathcal{L}_3'-\mathcal{L}_4') + \frac{\gamma-\gamma_c}{(\gamma-\gamma_c)^2+s^2}\left[\mathcal{B}_1'+\mathcal{B}_2' \pm (\mathcal{B}_1'+\mathcal{B}_2')\right.$$

$$\left. - \frac{s}{(\gamma-\gamma_c)^2+s^2}\left[\mathcal{L}_1'-\mathcal{L}_2' \mp (\mathcal{L}_1'-\mathcal{L}_2')\right]\right\} \qquad (4.2.20)$$

where

$$\mathcal{L}_0' = \frac{2\gamma}{4\gamma^2+\upsilon^2} \qquad \mathcal{B}_0' = \frac{\upsilon}{4\gamma^2+\upsilon^2}$$

$$\mathcal{L}_1' = \mathcal{L}_1 \qquad \mathcal{B}_1' = \mathcal{B}_1$$

$$\mathcal{L}_2' = \mathcal{L}_2 \qquad \mathcal{B}_2' = -\mathcal{B}_2$$

$$\mathcal{L}_3' = \mathcal{L}_3/2 \qquad \mathcal{B}_3' = \mathcal{B}_3/2$$

$$\mathcal{L}_4' = \mathcal{L}_4/2 \qquad \mathcal{B}_4' = \mathcal{B}_4/2$$

(4.2.21)

Hence, there are resonances in the amplitude of modulation of the fluorescent light whenever the frequency of modulation is

zero or equal to the Larmor precessional frequency or twice of it i.e. at $\nu = 0, \pm s, \pm 2s$. The bandwidth of the fluctuating laser beam effects the width of the resonance at $\nu = \pm s$ only, the remaining resonances having a width determined by the life-time of the excited state only. The shape of the detected signals is critically dependent on the relative magnitude of the Larmor frequency and the decay constants. For small strengths of the magnetic field so that the separation between the Zeeman sublevels is of the same order as the natural width ($\frac{s}{\gamma} \sim 1$), there is considerable overlap of the resonances. At higher magnetic field strengths, the resonances are clearly resolved and the effect of source bandwidth more pronounced, as will be shown by the study of the following limiting cases.

(i) Broad Band Excitation :

The signals (5.2.20) reduce to

$$L(x) \propto \cos\nu t\ (2\mathcal{L}'_0+\mathcal{L}'_3+\mathcal{L}'_4) + \sin\nu t\ (2\mathcal{B}'_0+\mathcal{B}'_3+\mathcal{B}'_4)$$

$$L(y) \propto \cos\nu t\ (2\mathcal{L}'_0-\mathcal{L}'_3-\mathcal{L}'_4) + \sin\nu t\ (2\mathcal{B}'_0-\mathcal{B}'_3-\mathcal{B}'_4) \qquad (4.2.22)$$

Thus when the optical source used to excite the atoms is a weak, incoherent broad-band lamp, the modulated fluorescence exhibits

resonances at $\upsilon = 0, \pm 2s$ of width 2γ. These resonances are Lorentzian (dispersion) shaped for the amplitude of the cosine (sine) component of the modulated fluorescence and are shown as the dot - dash curves of figs.14-17 corresponding to $\gamma_c = 5\gamma$, when the magnetic field is held constant such that $\frac{s}{\gamma} = 5$.

(ii) Monochromatic Excitation :

The detection signals are

$$L(x) \propto \cos \upsilon t \, (\mathcal{L}'_1 + \mathcal{L}'_2) + \sin \upsilon t \, (\mathcal{D}'_1 + \mathcal{D}'_2)$$

$$L(y) \propto \cos \upsilon t \, (\mathcal{D}'_1 - \mathcal{D}'_2) + \sin \upsilon t \, (\mathcal{L}'_2 - \mathcal{L}'_1) \qquad (4.2.23)$$

In contrast to (i), resonances are obtained at $\upsilon = \pm s$ of width (γ), the two detection signals being entirely different for a particular component of the fluorescence, as may be seen from the solid curves of figs. 14-17 corresponding to $\gamma_c = 0$.

(iii) Intermediate Case :

For an optical source of bandwidth of the same order as the width of the excited state $(\gamma_c \sim \gamma)$, the detection signals are no longer so simple

$$L(x) \propto \cos \upsilon t \left\{ \frac{2\gamma}{4\gamma^2 + s^2} (2\mathcal{L}'_0 + \mathcal{L}'_3 + \mathcal{L}'_4) + \frac{4\gamma^2}{s(4\gamma^2 + s^2)} (\mathcal{D}'_3 - \mathcal{D}'_4) \right\}$$

$$+ \sin \upsilon t \left\{ \frac{2\gamma}{4\gamma^2 + s^2} (2\mathcal{D}'_0 + \mathcal{D}'_3 + \mathcal{D}'_4) - \frac{4\gamma^2}{s(4\gamma^2 + s^2)} (\mathcal{L}'_3 - \mathcal{L}'_4) \right\}$$

$$L(y) \propto \cos\nu t \left\{ \frac{2\gamma}{4\gamma^2+s^2}(2\mathcal{L}'_0 - \mathcal{L}'_3 - \mathcal{L}'_4) - \frac{4\gamma^2}{s(4\gamma^2+s^2)}(\mathcal{D}'_3 - \mathcal{D}'_4) + \frac{2}{s}(\mathcal{D}'_1 - \mathcal{D}'_2) \right\}$$

$$+ \sin\nu t \left\{ \frac{2\gamma}{4\gamma^2+s^2}(2\mathcal{D}'_0 - \mathcal{D}'_3 - \mathcal{D}'_4) + \frac{4\gamma^2}{s(4\gamma^2+s^2)}(\mathcal{L}'_3 - \mathcal{L}'_4) - \frac{2}{s}(\mathcal{L}'_1 - \mathcal{L}'_2) \right\}$$

(4.2.24)

Thus for detection along magnetic field direction there are resonances at $\nu = 0, \pm 2s$. The resonance at $\nu = 0$ is Lorentzian (dispersion) in shape for the cosine (sine) component of the fluorescence, while the resonances at $\nu = \pm 2s$ have both a Lorentzian and dispersion part, with different weight factors such that either one predominates when $s \gg \gamma$ (see broken curves of Figs. 14 and 15). The modulated fluorescence detected perpendicular to the magnetic field direction exhibits resonances at $\nu = 0, \pm s, \pm 2s$ so that for the case studied ($\frac{s}{\gamma} = 5$) there is considerable overlap of these resonances (see broken curves of Figs. 16 and 17).

Section 4.3 : Intense Fields :

It is no longer possible to carry out the above procedure for intense laser beams of arbitrary bandwidth, so that the solutions (4.1.14) to first order in 'a' (the depth of modulation

of the exciting laser is always assumed to be weak) are valid to all orders in the atom-laser coupling. In order to obtain analytical expressions for the signals, we would have to invert 8x8 matrices and find their products, which though possible in principle, is extremely cumbersome and would yield complicated expressions which lend no insight into the problem. Hence we study the modulated Hanle signals in intense laser beams numerically. For intense laser fields ($\alpha >> \gamma, \gamma_c, s, \delta$), one expects the resonances in the amplitude of modulation to yield the energy spectra of the composite system consisting of the atom and the coherent laser field i.e. resonances are expected in the modulated fluorescence corresponding to the dynamical stark splitting of the various energy levels. An idea of the position of the resonant components may be obtained by finding the eigenvalues of the resonant part of the Hamiltonian (without the damping term), which now requires dealing with 3x3 matrix. The resonant Hamiltonian may be written as

$$H = \begin{bmatrix} s-\delta & 0 & \alpha \\ 0 & -s-\delta & \alpha \\ \alpha & -\alpha & 0 \end{bmatrix} \qquad (4.3.1)$$

and the secular equation $|H - \lambda \hat{1}| = 0$ yields the following cubic equation for the eigenvalues

$$\lambda(\lambda + s + \delta)(\lambda - s + \delta) - 2\alpha^2(\lambda + \delta) = 0 \qquad (4.3.2)$$

For arbitrary values of the detuning there are no simple factorisations of the cubic equation and the roots have a complicated form. In the special case of the saturing laser field being resonant with the atomic transition $|J=0, m_J=0\rangle \leftrightarrow |J=1, m_J=0\rangle$ ($\delta=0$) the eigenvalues have the simple form

$$\lambda = 0, \pm \alpha_o \qquad \alpha_o^2 \equiv s^2 + 2\alpha^2 \qquad (4.3.3)$$

In the magnetic field scan of the signals we expect the corresponding resonances to show up at Larmor frequency equal to $s = \pm\sqrt{\frac{\upsilon^2}{4} - 2\alpha^2}, \pm\sqrt{\upsilon^2 - 2\alpha^2}$. If $\upsilon/\alpha \ll 1$, the modulated signals are similar in shape to the weak field case so that the modulation frequency must be fixed at a much higher value than the Rabi frequency for these peaks to be well resolved. When the modulated fluorescence is scanned as a function of the modulation frequency, resonances are expected at $0, \pm\alpha_o, \pm 2\alpha_o$ provided of course their weight is not zero. The resonances at $\upsilon = 0, \pm\alpha_o, \pm 2\alpha_o$ are the analog of the resonances at $\upsilon = 0, \pm s, \pm 2s$ in the weak fields. The weight factors of these lines can be calculated using secular approximation as will be done in the next chapter and yields the results in agreement with numerical computations which are shown in Figs. 18-21.

The signal detected perpendicular to magnetic field direction exhibits well defined resonant structures at $\upsilon = \pm \alpha_o, \pm 2\alpha_o$, while the fluorescence detected along the magnetic field exhibits sharp resonances at $\upsilon = \pm 2\alpha_o$ only. These resonances are lorentzian (dispersion) in shape for the sine (cosine) component of the modulated fluorescence in both the directions. The effect of laser bandwidth is to alter the peak height as well as the width of the resonances, resulting in considerable broadening for large γ_c.

CHAPTER - V

TWO PHOTON STEPWISE EXCITATION IN MODULATED FIELDS

In the previous chapter, we obtained the important result that an atom excited by a near-resonant, modulated laser exhibits resonances in the amplitude of the modulated fluorescence due to dynamical Stark effect.[7] In view of this result, it would be interesting to study the modulated fluorescence when two atomic transitions are saturated successively by two modulated laser beams having different frequencies. The first beam induces the transition to an intermediate level and the second to the final level. Two-photon stepwise excitations are specially useful when one wishes to study states that are not connected to the ground state by an electric dipole transition.[57-61] For excitation with modulated laser beams, the total intensity of the emitted radiation is expected to show resonant behaviour in the modulation frequency corresponding to the Stark split energy levels of the multilevel system. In this chapter, we study two-photon, stepwise excitations in modulated fields. The basic problem is formulated and the equations for the evolution of the density matrix elements are obtained using the master equation techniques outlined in section 2.2. Using the general results of chapter II, we obtain the intensity of the modulated fluorescence to first order in the interaction with the modulated fields, the coupling with

the intense laser fields being treated to all orders. It is shown that the resonances in the amplitude of modulation yield the energy spectra of the composite system consisting of the atom and the coherent laser fields. For instance, we show that the amplitude corresponding to the steady-state population of the highest excited state exhibits resonances at the modulation frequency values 0, $\pm \alpha_0$, $\pm 2\alpha_0$ due to dynamical stark splitting of the various energy levels (α_0 is related to the Rabi frequencies α_1 and α_2 characterising the coupling of the atom with the two fields, by $\alpha_0^2 = \alpha_1^2 + \alpha_2^2$). In the presence of detunings of the applied fields, numerical results reveal additional structures. This should be contrasted with the zeroth order fluorescence (unmodulated) which does <u>not</u> exhibit dynamic Stark splittings even when both the lasers are very intense[59].

Section 5.1 - Dynamical Equations :

Since we envisage the experiment to be performed with atomic beams, the only excitation mechanism is the external field, and the only damping present is natural radiative decay. We consider a three-level atom with unequidistant, non-degenerate energy levels labelled 1, 2 and 3 in figure The energy eigenstates corresponding to these levels are $|1\rangle$, $|2\rangle$ and $|3\rangle$, with energies $\omega_{13}(=\omega_{12}+\omega_{23})$, ω_{23} and zero respectively. The atomic system is driven simultaneously by two intense laser beams which saturate two

atomic transitions with a common intermediate level. Level $|1\rangle$ can decay to level $|2\rangle$, and level $|2\rangle$ to $|3\rangle$ at the rate of $2\gamma_{21}$ $(\equiv 2\gamma_1)$ and $2\gamma_{32}$ $(\equiv 2\gamma_2)$ respectively, where $2\gamma_{fi}$ is the transition probability per unit time that the system makes a transition from state $|i\rangle \rightarrow |f\rangle$ as a result of spontaneous emission. The transition from $|1\rangle$ to $|3\rangle$ is forbidden, say, due to parity considerations. In accordance with the notation developed in chapter II, the electric fields of the modulated sources at the position of the atom (chosen as the origin) may be written as

$$\vec{E}_i(t) = \tfrac{1}{2}\vec{\mathcal{E}}_i[1+a_i \cos(\nu_i t + \phi_i)]\, e^{-i\Omega_i t} + \text{c.c.} \qquad (5.1.1)$$

$$i = 1,2$$

The subscript i=1 [2] serves to distinguish the laser field saturating the $|1\rangle \longleftrightarrow |2\rangle$ [$|2\rangle \longleftrightarrow |3\rangle$] transition and the symbols have the meaning defined in (2.1.7). From (2.1.6), we find that the atomic dipole moment operator for this case may be written as

$$\hat{\vec{d}} = \vec{d}_{12} A_{12} + \vec{d}_{23} A_{23} + \text{H.c.} \qquad (5.1.2)$$

This leads to the following form for the interaction Hamiltonian between the atom and the external fields :

$$H_{ext}(t) = \alpha_1[1+a_1 \cos(\nu_1 t+\phi_1)]e^{-i\Omega_1 t}\,(A_{12} + A_{21})$$

$$+\alpha_2[1+a_2 \cos(\nu_2 t+\phi_2)]e^{-i\Omega_2 t}\,(A_{23} + A_{32}) + \text{c.c.} \qquad (5.1.3)$$

$$2\alpha_1 = -\vec{d}_{12}\cdot\vec{\mathcal{E}}_1 \quad ; \quad 2\alpha_2 = -\vec{d}_{12}\cdot\vec{\mathcal{E}}_2 \qquad (5.1.4)$$

are the Rabi frequencies characterising the coupling of the laser fields $\vec{E}_1(t)$ and $\vec{E}_2(t)$ with the $|1\rangle \leftrightarrow |2\rangle$ and $|2\rangle \leftrightarrow |3\rangle$ transitions respectively. Substituting (5.1.3) in (2.2.7), we obtain the equations for the evolution of the density matrix elements as

$$\dot{\tilde{\rho}}_{11} = -2\gamma_1\tilde{\rho}_{11} + \{ -i\alpha_1 [1+a_1 \cos(\upsilon_1 t+\phi_1)]\tilde{\rho}_{21} + c.c.\}$$

$$\dot{\tilde{\rho}}_{12} = -(\gamma_1+\gamma_2+i\Delta_1)\tilde{\rho}_{12} + i\alpha_1 [1+a_1 \cos(\upsilon_1 t+\phi_1)](\tilde{\rho}_{11}-\tilde{\rho}_{22}) + i\alpha_2^* [1+a_2 \cos(\upsilon_2 t+\phi_2)]\tilde{\rho}_{13}$$

$$\dot{\tilde{\rho}}_{13} = -[\gamma_1+i(\Delta_1+\Delta_2)]\tilde{\rho}_{13} - i\alpha_1 [1+a_1 \cos(\upsilon_1 t+\phi_1)]\tilde{\rho}_{23} + i\alpha_2 [1+a_2 \cos(\upsilon_2 t+\phi_2)]\tilde{\rho}_{12}$$

$$\dot{\tilde{\rho}}_{22} = 2\gamma_1\tilde{\rho}_{11} - 2\gamma_2\tilde{\rho}_{22} + \{ i\alpha_1 [1+a_1 \cos(\upsilon_1 t+\phi_1)]\tilde{\rho}_{21} - i\alpha_2 [1+a_2 \cos(\upsilon_2 t+\phi_2)]\tilde{\rho}_{32} + c.c.\}$$

$$\dot{\tilde{\rho}}_{23} = -(\gamma_2+i\Delta_2)\tilde{\rho}_{23} - i\alpha_1^* [1+a_1 \cos(\upsilon_1 t+\phi_1)]\tilde{\rho}_{13} + i\alpha_2 [1+a_2 \cos(\upsilon_2 t+\phi_2)](\tilde{\rho}_{22}-\tilde{\rho}_{33})$$

$$\dot{\tilde{\rho}}_{33} = 2\gamma_2\tilde{\rho}_{22} + \{ i\alpha_2 [1+a_2 \cos(\upsilon_2 t+\phi_2)]\tilde{\rho}_{32} + c.c.\}$$

(5.1.5)

where the oscillations at optical frequencies have been removed by transforming to the slowly varying quantities :

$$\tilde{\rho}_{ii} = \rho_{ii} \qquad \tilde{\rho}_{12} = \rho_{12}\, e^{-i\Omega_1 t}$$

$$\tilde{\rho}_{23} = \rho_{23}\, e^{i\Omega_2 t} \qquad \tilde{\rho}_{13} = \rho_{13}\, e^{i(\Omega_1+\Omega_2)t} \tag{5.1.6}$$

and neglecting terms with $e^{\pm 2i\Omega_1 t}$, $e^{\pm 2i\Omega_2 t}$, $e^{\pm 2i(\Omega_1+\Omega_2)t}$ (RWA). The detunings between the laser and atomic frequencies are given by the parameters

$$\Delta_1 = \omega_{12} - \Omega_1 \qquad \Delta_2 = \omega_{23} - \Omega_2 \tag{5.1.7}$$

Equations (5.1.5) form a closed set of coupled, linear differential equations and in matrix notation, they may be written in the following form (compare from equation (2.2.10)):

$$\dot{\tilde{\psi}} = B\tilde{\psi} + I + a_1\cos(\nu_1 t+\phi_1)M_1\tilde{\psi} + a_2\cos(\nu_2 t+\phi_2)(M_2\tilde{\psi}+J_2) \tag{5.1.8}$$

where $\tilde{\psi}$, I and J_2 are the following column matrices :

$$\tilde{\psi} = \begin{bmatrix} \tilde{\rho}_{11} \\ \tilde{\rho}_{12} \\ \tilde{\rho}_{13} \\ \tilde{\rho}_{21} \\ \tilde{\rho}_{22} \\ \tilde{\rho}_{23} \\ \tilde{\rho}_{31} \\ \tilde{\rho}_{32} \end{bmatrix}, \quad I_6 = (J_2)_6 = -i\alpha_2 = I_8^* = (J_2)_8^* \tag{5.1.9}$$

M_1 and M_2 are square matrices of order 8x8 having the structure

$$\begin{bmatrix}
-2\gamma_1 & i\alpha_1^* & 0 & -i\alpha_1 & 0 & 0 & 0 & 0 \\
i\alpha_1 & -\gamma_1-\gamma_2-i\Delta_1 & i\alpha_2^* & 0 & -i\alpha_1 & 0 & 0 & 0 \\
0 & i\alpha_2 & -\gamma_1-i\Delta_1-i\Delta_2 & 0 & 0 & -i\alpha_1 & 0 & 0 \\
-i\alpha_1^* & 0 & 0 & -\gamma_1-\gamma_2+i\Delta_1 & i\alpha_1^* & 0 & -i\alpha_2 & 0 \\
2\gamma_1 & -i\alpha_1^* & 0 & i\alpha_1 & -2\gamma_2 & i\alpha_2^* & 0 & -i\alpha_1 \\
i\alpha_2 & 0 & -i\alpha_1^* & 0 & 2i\alpha_2 & -\gamma_2-i\Delta_2 & 0 & 0 \\
0 & 0 & 0 & -i\alpha_2^* & 0 & 0 & -\gamma_1+i\Delta_1+i\Delta_2 & i\alpha_1 \\
-i\alpha_2^* & 0 & 0 & 0 & -2i\alpha_2^* & 0 & i\alpha_1 & -\gamma_2+i
\end{bmatrix}$$

(5.1.10)

with

$$(M_1)_{14} = (M_1)_{25} = (M_1)_{36} = -i\alpha_1 = -(M_1)_{21} = -(M_1)_{54} = -(M_1)_{87}$$

$$(M_1)_{12} = (M_1)_{45} = (M_1)_{78} = i\alpha_1^* = -(M_1)_{41} = -(M_1)_{52} = -(M_1)_{63}$$

(5.1.11)

and

$$(M_2)_{47} = (M_2)_{58} = -i\alpha_2 = -(M_2)_{32} = -(M_2)_{61} = -\frac{1}{2}(M_2)_{65}$$

$$(M_2)_{23} = (M_2)_{56} = i\alpha_2^* = -(M_2)_{74} = -(M_2)_{81} = -\frac{1}{2}(M_2)_{85} \ .$$

(5.1.12)

We have eliminated $\tilde{\rho}_{33}$ from equations (5.1.5) by using the normalisation condition $\tilde{\rho}_{11} + \tilde{\rho}_{22} + \tilde{\rho}_{33} = 1$ in order to avoid the difficulty that would arise in computing the steady-state solutions by Laplace transform techniques were the matrix B to possess a zero eigenvalue. As outlined in section 3 of chapter II we obtain the solutions of (5.1.8) within the framework of perturbation theory since the modulation is always assumed to be weak i.e. $a_1, a_2 \ll 1$. The interaction with the two different laser beams will be treated exactly. From (2.3.11), we have the solutions at steady state to first order in the modulation ratios :

$$\tilde{\psi}^{(1)}_{(\infty)} = \frac{1}{2}\sum_{j=1}^{2} a_j \Big\{ \Big[(i\upsilon_j - B)^{-1} e^{i(\upsilon_j t + \phi_j)} + (-i\upsilon_j - B)^{-1} e^{-i(\upsilon_j t + \phi_j)} \Big] (M_j \tilde{\psi}^{(o)}_{(\infty)} + J_j) \Big\}$$

(5.1.13)

with

$$J_1 = 0 \qquad (5.1.14)$$

We are interested in obtaining the total intensity of the fluorescent radiation. From (2.3.1), we have the positive frequency part of the radiation field as

$$\vec{E}^{(+)}(\vec{r},t)=\vec{E}_o^{(+)}(\vec{r},t) - \frac{\omega_{12}^2}{c^2 r}[\hat{r} \times(\hat{r} \times \vec{d}_{12})]A_{21}(t-\frac{r}{c})- \frac{\omega_{23}^2}{c^2 r}[\hat{r}\times(\hat{r}\times\vec{d}_{23})]A_{32}($$

(5.1.15)

which leads to the following expression for the intensity of the spontaneously emitted radiation

$$I_{scat} = \frac{\omega_{12}^4}{c^4 r^2}[\hat{r} \times (\hat{r} \times \vec{d}_{12}^{*})] \cdot [\hat{r} \times (\hat{r} \times \vec{d}_{12}] \langle A_{11}(t-\tfrac{r}{c})\rangle \cdot$$

$$+\frac{\omega_{23}^4}{c^4 r^2}[\hat{r} \times (\hat{r} \times \vec{d}_{23}^{*})]\cdot[\hat{r} \times (\hat{r} \times \vec{d}_{23})] \langle A_{22}(t-\tfrac{r}{c})\rangle \quad (5.1.16)$$

so that apart from unessential constants, the total intensity of the emitted radiation is given by the behaviour of the populations of the upper and intermediate states. Since these occur as the first and fifth elements of the column vector $\tilde{\psi}$ defined in (5.1.9), we have from (5.1.13)

$$\tilde{\rho}_{11}^{(1)}(\infty) = \tfrac{1}{2} a_1 \Big\{ e^{i(\nu_1 t+\phi_1)} \sum_{i,j=1}^{8} (i\nu_1-B)^{-1}_{1i}(M_1)_{ij}\tilde{\psi}_j^{(o)}(\infty)+ c.c.\Big\}$$

$$+\tfrac{1}{2}a_2\Big\{ e^{i(\nu_2 t+\phi_2)} \sum_{i,j=1}^{8} (i\nu_2-B)^{-1}_{1i}(M_2)_{ij}\tilde{\psi}_j^{(o)}(\infty)+(J_2)_i]+ c.c.\Big\}$$

$$\equiv a_1 A_1 \cos(\nu_1 t+\phi_1+\Phi_1) + a_2 A_2 \cos(\nu_2 t+\phi_2+\Phi_2)$$

(5.1.17)

Similarly

$$\tilde{\rho}^{(1)}_{22(\infty)} = \tfrac{1}{2} a_1 \Big\{ e^{i(\nu_1 t+\phi_1)} \sum_{i,j=1}^{8} (i\nu_1-B)^{-1}_{5i}(M_1)_{ij} \tilde{\psi}^{(o)}_j(\infty) + c.c. \Big\}$$

$$+ \tfrac{1}{2} a_2 \Big\{ e^{i(\nu_2 t+\phi_2)} \sum_{ij=1} (i\nu_2-B)^{-1}_{5i}[(M_2)_{ij} \tilde{\psi}^{(o)}_j(\infty)+(J_2)_i] + c.c. \Big\}$$

$$\equiv a_1 B_1 \cos(\nu_1 t+\phi_1+\Theta_1) + a_2 B_2 \cos(\nu_2 t+\phi_2+\Theta_2) \qquad (5.1.18)$$

where we have used the following notation for the phaseshifts ($\equiv \Phi, \Theta$). The amplitudes of modulation ($\equiv$ A,B) will be shown to exhibit resonant structures in the next two sections and in general the behaviour is critically dependent on the six parameters occuring in the problem, namely, the two decay rates γ_1, γ_2, the Rabi frequencies α_1 , α_2 corresponding to the two saturating laser beams and the detunings Δ_1 , Δ_2 between the atomic and laser frequencies. For general values of these six parameters, the solutions (5.1.17) and (5.1.18) require the inversion of 8x8 matrices which is cumbersome. However, analytical forms of the steady-state solutions for the intense fields at resonance with the respective atomic transitions will be obtained in the next section, while numerical studies for general values of the parameters occuring in the problem will be presented in section 5.3.

Section-5.2 - Analytical results for the Modulated Fluorescence in Intense, Resonant Fields :

If both the exciting laser fields are strong, i.e. $|\alpha_1|$, $|\alpha_2| \gg \gamma_1, \gamma_2$, then instead of solving nine coupled equations for the density matrix elements to obtain the relevant information, one can greatly simplify the calculations by solving the master equation in a representation in which the resonant part of the Hamiltonian (without the damping terms) is diagonal.[89] This will require diagonalisation of a 3 x 3 matrix (instead of a 9 x 9 one), and will directly yield the positions, widths and weights of the resonant components of the modulated fluorescence in principle.

The master equation, with the radiative decay term, may be written as,

$$\frac{\partial \tilde{\rho}}{\partial t} = -i[H, \tilde{\rho}] + \mathcal{L}\tilde{\rho} \qquad (5.2.1)$$

Let S be the time-independent matrix which diagonalises H, i.e.

$$S^{\dagger} H S = \Lambda \qquad (5.2.2)$$

where

$$H = \begin{bmatrix} \Delta_1+\Delta_2 & \alpha_1 & 0 \\ \alpha_1^* & \Delta_2 & \alpha_2 \\ 0 & \alpha_2^* & 0 \end{bmatrix} \qquad (5.2.3)$$

with

$$\Lambda_{ij} = \lambda_i \delta_{ij} \qquad i = 1,2,3 \tag{5.2.4}$$

λ_i being the eigenvalues of the resonant Hamiltonian H determined from the cubic equation :

$$\lambda[(\lambda-\Delta_2)(\lambda-\Delta_1-\Delta_2)-|\alpha_1|^2-|\alpha_2|^2]+|\alpha_2|^2(\Delta_1+\Delta_2) = 0 \ . \tag{5.2.5}$$

For arbitrary values of the detunings there are no simple factorisations of the cubic equation and λ_i have a complicated form. In the special case of both the saturating laser fields being resonant with the atomic transitions ($\Delta_1 = 0 = \Delta_2$) the eigenvalues have the simple form

$$\lambda = 0, \pm \alpha_o \qquad \alpha_o \equiv (|\alpha_1|^2 + |\alpha_2|^2)^{\frac{1}{2}} \tag{5.2.6}$$

so that the matrix S is

$$S = \begin{pmatrix} +\alpha_2|\alpha_1|/(\alpha_o\alpha_1^*) & -\alpha_1|\alpha_2|/(\sqrt{2}\alpha_o\alpha_2^*) & -\alpha_1|\alpha_2|/(\sqrt{2}\alpha_o\alpha_2^*) \\ 0 & -|\alpha_2|/(\sqrt{2}\alpha_2^*) & |\alpha_2|/(\sqrt{2}\alpha_2^*) \\ -|\alpha_1|/\alpha_o & -|\alpha_2|/(\sqrt{2}\alpha_o) & -|\alpha_2|/(\sqrt{2}\alpha_o) \end{pmatrix} \tag{5.2.7}$$

The master equation (5.2.1) in this representation has the form

$$\frac{\partial \bar{\rho}}{\partial t} = -i[\Lambda, \bar{\rho}] + S^\dagger\{\mathcal{L}(S\bar{\rho}S^\dagger)\}S \tag{5.2.8}$$

where

$$S^{\dagger}\tilde{\rho}\,S = \bar{\rho} \tag{5.2.9}$$

The radiative decay term leads to the coupling of the various matrix elements of $\bar{\rho}$ to each other. Since $|\alpha_1|, |\alpha_2| \gg \gamma_1, \gamma_2$ we can neglect any coupling of the diagonal elements of $\bar{\rho}$ with the off-diagonal elements, and also the coupling between the off-diagonal elements (valid only to the lowest order in $\gamma/|\alpha|$). The secular approximation leads to the following equations for the density matrix elements :

$$\begin{aligned}
\dot{\bar{\rho}}_{12} &= (-\beta_1 + i\alpha_o)\,\bar{\rho}_{12} \\
\dot{\bar{\rho}}_{13} &= (-\beta_1 - i\alpha_o)\,\bar{\rho}_{13} \\
\dot{\bar{\rho}}_{23} &= (-\beta_2 - 2i\alpha_o)\,\bar{\rho}_{23} \\
\dot{\bar{\rho}}_{11} &= -2\gamma_1(\alpha_2^2/\alpha_o^2)\,\bar{\rho}_{11} + \gamma_2(\alpha_1^2/\alpha_o^2)(\bar{\rho}_{22} + \bar{\rho}_{33}) \\
\dot{\bar{\rho}}_{22} + \dot{\bar{\rho}}_{33} &= 2\gamma_1(\alpha_2^2/\alpha_o^2)\,\bar{\rho}_{11} - \gamma_2(\alpha_1^2/\alpha_o^2)(\bar{\rho}_{22} + \bar{\rho}_{33}) \\
\dot{\bar{\rho}}_{22} - \dot{\bar{\rho}}_{33} &= -(\gamma_2 + \gamma_1\alpha_1^2/\alpha_o^2)(\bar{\rho}_{22} - \bar{\rho}_{33})
\end{aligned} \tag{5.2.10}$$

where

$$\beta_1 = \frac{1}{2\alpha_o^2}\left[\gamma_1(\alpha_1^2 + 2\alpha_2^2) + \gamma_2(\alpha_1^2 + \alpha_2^2)\right]$$

$$\beta_2 = \frac{1}{2\alpha_o^2}\left[3\gamma_1\alpha_1^2 + \gamma_2(2\alpha_1^2 + 3\alpha_2^2)\right] \tag{5.2.11}$$

and the Rabi frequencies α_1, α_2 have been assumed to be real by a proper choice of the relative phases of the energy levels. For large times, we may set the time derivatives in (5.2.10) equal to zero, so that in the steady-state limit, the solutions have the form

$$\overline{\rho}_{ij}(\infty) = \delta_{ij}\,[b\delta_{i3} + b\delta_{i2} + (1-2b)\delta_{i1}] \qquad (5.2.12)$$

where

$$b \equiv \frac{\gamma_1\alpha_2^2}{\gamma_2\alpha_1^2+2\gamma_1\alpha_2^2} \quad . \qquad (5.2.13)$$

In the presence of a weak modulating field, the master equation in the representation in which the resonant part of the Hamiltonian is diagonal (5.2.8) will have the form,

$$\frac{\partial\overline{\rho}}{\partial t} = -i[\Lambda, \overline{\rho}] - i\,[\overline{H}_{mod}, \overline{\rho}] + S^{\dagger}\{\mathcal{L}\,(S\overline{\rho}S^{\dagger})\}\,S \qquad (5.2.14)$$

where

$$\overline{H}_{mod} = S^{\dagger}H_{mod}S \qquad (5.2.15)$$

with

$$H_{mod} = \begin{bmatrix} 0 & -a_1\alpha_1\cos(\nu_1 t+\phi_1) & 0 \\ -a_1\alpha_1\cos(\nu_1 t+\phi_1) & 0 & -a_2\alpha_2\cos(\nu_2 t+\phi_2) \\ 0 & -a_2\alpha_2\cos(\nu_2 t+\phi_2) & 0 \end{bmatrix} \qquad (5.2.16)$$

The equations for the evolution of the density matrix elements are obtained from (5.2.14) to first order in the modulating fields:

$$\dot{\bar{\rho}}^{(1)}_{12} = (-\beta_1+i\alpha_o)\bar{\rho}^{(1)}_{12}-ix(\bar{\rho}^{(o)}_{11}-\bar{\rho}^{(o)}_{22}+\bar{\rho}^{(o)}_{32})+iy\,\bar{\rho}^{(o)}_{12}$$

$$\dot{\bar{\rho}}^{(1)}_{13} = (-\beta_1-i\alpha_o)\bar{\rho}^{(1)}_{13}+ix(\bar{\rho}^{(o)}_{11}-\bar{\rho}^{(o)}_{33}+\bar{\rho}^{(o)}_{23})-iy\bar{\rho}^{(o)}_{13}$$

$$\dot{\bar{\rho}}^{(1)}_{23} = (-\beta_2-2i\alpha_o)\,\bar{\rho}^{(1)}_{23} + ix(\bar{\rho}^{(o)}_{13} + \bar{\rho}^{(o)}_{21}) - 2iy\,\bar{\rho}^{(o)}_{23}$$

$$\dot{\bar{\rho}}^{(1)}_{11} =-2\gamma_1(\alpha_2^2/\alpha_o^2)\,\bar{\rho}^{(1)}_{11} + \gamma_2(\alpha_1^2/\alpha_o^2)(\bar{\rho}^{(1)}_{22}+\bar{\rho}^{(1)}_{33}-ix(\bar{\rho}^{(o)}_{12}-\bar{\rho}^{(o)}_{13} - c.c.)$$

$$\dot{\bar{\rho}}^{(1)}_{22}+\dot{\bar{\rho}}^{(1)}_{33} =2\gamma_1(\alpha_2^2/\alpha_o^2)\,\bar{\rho}^{(1)}_{11}-\gamma_2(\alpha_1^2/\alpha_o^2)(\bar{\rho}^{(1)}_{22}+\bar{\rho}^{(1)}_{33}) +ix(\bar{\rho}^{(o)}_{12}-\bar{\rho}^{(o)}_{13} - c.c.)$$

$$\dot{\bar{\rho}}^{(1)}_{22}-\dot{\bar{\rho}}^{(1)}_{33}= -(\gamma_2+\gamma_1\alpha_1^2/\alpha_o^2)\,(\bar{\rho}^{(1)}_{22}-\bar{\rho}^{(1)}_{33})+ix(\bar{\rho}^{(o)}_{12}+\bar{\rho}^{(o)}_{13}+c.c.)$$

(5.2.17)

Where

$$x = \sqrt{\tfrac{1}{2}}\alpha_1\alpha_2\alpha_o^{-1}[a_1\cos(\nu_1 t+\phi_1) - a_2\cos(\nu_2 t+\phi_2)],$$
$$y = \alpha_o^{-1}[a_1\alpha_1^2\cos(\nu_1 t+\phi_1)+a_2\alpha_2^2\cos(\nu_2 t+\phi_2)].$$

(5.2.18)

We are interested in obtaining the total intensity of the modulated fluorescence, which is proportional to the populations ρ_{11} and ρ_{22} in the original representation by equation (5.1.16). Using (5.2.9), we find that the populations in the original representation are related to the following matrix elements in the diagonal representation :

$$\tilde{\rho}_{11}^{(1)}(\omega) = \sum_{i,j=1}^{3} S_{1i}\, \bar{\rho}_{ij}^{(1)}(\omega)\, S_{j1}^{\dagger}$$

$$= \frac{\alpha_2^2}{\alpha_o^2}\, \bar{\rho}_{11}^{(1)}(\omega) + \frac{\alpha_1^2}{2\alpha_o^2}\,[\, \bar{\rho}_{22}^{(1)}(\omega) + \bar{\rho}_{33}^{(1)}(\omega) + (\bar{\rho}_{23}^{(1)}(\omega) + \text{c.c.})]$$

$$= \frac{\alpha_1\alpha_2}{\sqrt{2}\alpha_o^2}(\bar{\rho}_{12}^{(1)}(\omega) + \bar{\rho}_{13}^{(1)}(\omega) + \text{c.c.}). \qquad (5.2.19)$$

Similarly,

$$\tilde{\rho}_{22}^{(1)}(\omega) = \sum_{i,j=1}^{3} S_{2i}\, \bar{\rho}_{ij}^{(1)}(\omega) S_{j2}^{\dagger} = \frac{1}{2}[\, \bar{\rho}_{22}^{(1)}(\omega) - (\bar{\rho}_{23}^{(1)}(\omega) + \text{c.c.}) + \bar{\rho}_{33}(\omega)\,] \qquad (5.2.20)$$

The matrix elements $\bar{\rho}_{ij}^{(1)}(\omega)$ may be found by solving the equations (5.2.17). We obtain these solutions to first order in a_1, i.e. $a_2 = 0$ in (5.2.18). By an analysis similar to section 3 of chapter II, we may show that the solutions are of the form

$$\bar{\rho}_{ij}^{(1)}(\infty) = c_{ij}^{(+)} \exp[i\nu_1 t+\phi_1)] + c_{ij}^{(-)} \exp[-i(\nu_1 t+\phi_1)]$$

(5.2.21)

where the time-independent coefficients $c_{ij}^{(\pm)}$ are given by

$$c_{12}^{(\pm)} = (\pm i\nu_1+\beta_1-i\alpha_o)^{-1} \left[-ia_1 \frac{\alpha_1\alpha_2}{2\sqrt{2}\alpha_o} (\rho^{-(o)}_{11}(\infty) - \bar{\rho}^{(o)}_{22}(\infty) + \bar{\rho}^{(o)}_{32}(\infty)) + ia_1 \frac{\alpha_1^2}{2\alpha_o} \bar{\rho}^{(o)}_{12}(\infty)\right]$$

$$c_{13}^{(\pm)} = (\pm i\nu_1+\beta_1+i\alpha_o)^{-1} \left[ia_1 \frac{\alpha_1\alpha_2}{2\sqrt{2}\alpha_o} (\bar{\rho}^{(o)}_{11}(\infty) - \bar{\rho}^{(o)}_{33}(\infty) + \bar{\rho}^{(o)}_{23}(\infty)) - ia_1 \frac{\alpha_1^2}{2\alpha_o} \bar{\rho}^{(o)}_{13}(\infty)\right]$$

$$c_{23}^{(\pm)} = (\pm i\nu_1+\beta_2+2i\alpha_o)^{-1} \left[ia_1 \frac{\alpha_1\alpha_2}{2\sqrt{2}\alpha_o} (\bar{\rho}^{(o)}_{13}(\infty)+ \bar{\rho}^{(o)}_{21}(\infty)) - ia_1 \frac{\alpha_1^2}{\alpha_o} \bar{\rho}^{(o)}_{23}(\infty)\right]$$

$$c_{11}^{(\pm)} = \left(\pm i\nu_1+2\gamma_1 \frac{\alpha_2^2}{\alpha_o^2}\right)^{-1} \left[-ia_1 \frac{\alpha_1\alpha_2}{2\sqrt{2}\alpha_o} (\bar{\rho}^{(o)}_{12}(\infty)- \bar{\rho}^{(o)}_{13}(\infty)-c.c.\right.$$

$$c_{22}^{(\pm)} = c_{33}^{(\pm)} = \left(\pm i\nu_1+\gamma_2 \frac{\alpha_1^2}{\alpha_o^2}\right)^{-1} \left[ia_1 \frac{\alpha_1\alpha_2}{4\sqrt{2}\alpha_o} (\bar{\rho}^{(o)}_{12}(\infty)- \bar{\rho}^{(o)}_{13}(\infty)-c.c\right.$$

(5.2.22)

The solutions $\bar{\rho}^{(o)}_{ij}(\infty)$ in the absence of the modulating field, and to the lowest order in the secular approximation, are given by the equation (5.2.12), which shows that only the diagonal terms $\bar{\rho}^{(o)}_{ii}(\infty)$ are finite, all the off-diagonal elements being zero. Inserting these values, we obtain

$$C_{12}^{(\pm)} = (\pm i\nu_1 + \beta_1 - i\alpha_o)^{-1} i a_1 \frac{\alpha_1\alpha_2}{2\sqrt{2}\alpha_o}(3b-1)$$

$$C_{13}^{(\pm)} = (\pm i\nu_1 + \beta_1 + i\alpha_o)^{-1} i a_1 \frac{\alpha_1\alpha_2}{2\sqrt{2}\alpha_o}(1-3b)$$

$$C_{23}^{(\pm)} = C_{11}^{(\pm)} = C_{22}^{(\pm)} = C_{33}^{(\pm)} = 0 \qquad (5.2.23)$$

which shows that the resonant component in $\bar{\rho}_{11}^{(1)}(\infty)$ at the modulation frequency $\nu_1 = \pm\alpha_o$ is the most intense since it occurs to the lowest order in the secular approximation. In order to extract information about the other resonant component the off-diagonal matrix elements $\bar{\rho}_{ij}^{(o)}(\infty)$ which occur in the solutions (5.2.22) must be evaluated to first order in the secular approximation. Equations (5.2.10) in the absence of any secular approximation have the form:

$$\dot{\bar{\rho}}_{12} = (-\beta_1 + i\alpha_o)\bar{\rho}_{12} + \frac{\alpha_1\alpha_2}{\sqrt{2}\alpha_o^2}[\gamma_1\bar{\rho}_{11} + (\gamma_1+\gamma_2)\bar{\rho}_{22} + \gamma_2\bar{\rho}_{33} + (\gamma_1-\gamma_2)\bar{\rho}_{32} - \gamma_2\bar{\rho}_{23}] + \frac{1}{2}\left(\gamma_2 - \gamma_1\frac{\alpha_1^2}{\alpha_o^2}\right)\bar{\rho}_{13}$$

$$\dot{\bar{\rho}}_{13} = (-\beta_1 - i\alpha_o)\bar{\rho}_{13} + \frac{\alpha_1\alpha_2}{\sqrt{2}\alpha_o^2}[\gamma_1\bar{\rho}_{11} + \gamma_2\bar{\rho}_{22} + (\gamma_1+\gamma_2)\bar{\rho}_{33} + (\gamma_1-\gamma_2)\bar{\rho}_{23} - \gamma_2\bar{\rho}_{32}] + \frac{1}{2}\left(\gamma_2 - \gamma_1\frac{\alpha_1^2}{\alpha_o^2}\right)\bar{\rho}_{12}$$

$$\dot{\bar{\rho}}_{23}=(-\beta_2-2i\alpha_o)\,\bar{\rho}_{23}-\gamma_1-\frac{\alpha_2^2}{\alpha_o^2}\,\bar{\rho}_{11}+\frac{1}{\alpha_o^2}[-\gamma_1\alpha_1^2+\tfrac{1}{2}\gamma_2(\alpha_1^2+2\alpha_2^2)](\bar{\rho}_{22}+\bar{\rho}_{33}$$

$$-\frac{1}{2\alpha_o^2}\,(\gamma_1\alpha_1^2+\gamma_2\alpha_2^2)\bar{\rho}_{32}+\gamma_1\,\frac{\alpha_1\alpha_2}{\sqrt{2}\alpha_o^2}\,(\bar{\rho}_{12}+2\,\bar{\rho}_{21}+2\,\bar{\rho}_{13}+\bar{\rho}_{31}).$$

(5.2.24)

Using the zeroth-order secular approximation results from (5.2.12) we obtain the solution of (5.2.24) to first order in γ/α as

$$\bar{\rho}_{12}(\infty) = \frac{\alpha_1\alpha_2\Gamma_o}{\sqrt{2}\alpha_o^2(\beta_1-i\alpha_o)}$$

$$\bar{\rho}_{13}(\infty) = \bar{\rho}_{12}(\infty)$$

$$\bar{\rho}_{23}(\infty) = \frac{\alpha_1^2\Gamma_1+\alpha_2^2\Gamma_2}{\alpha_o^2(\beta_2+2i\alpha_o)} \qquad (5.2.25)$$

where

$$\Gamma_o \equiv \gamma_1(1-b) + 2\gamma_2 b$$

$$\Gamma_1 \equiv -2b\gamma_1+b\gamma_2$$

$$\Gamma_2 \equiv -\gamma_1(1-2b)+2b\gamma_2\ . \qquad (5.2.26)$$

Using the results (5.2.25) and (5.2.12) in (5.2.14), we may obtain $\bar{P}(\infty)$ to first order in modulation ratios. These show that for intense, resonant laser beams, the resonances in the modulation rates at $\nu = 0, \pm 2\alpha_o$, though finite, are weak compared to the corresponding ones at $\nu = \pm\alpha_o$ which occur to the zeroth order in secular approximation. Since these perturbati results are quite involved, we do not present them explicitly.

Section - 5.3 - Numerical results for the Modulated Fluorescence :

For general values of the parameters occuring in the problem, the solutions (5.1.17) and (5.1.18) require the inversion of several 8x8 matrices. This has been done numerically and we present the results for the modulation rate for a variety of situations.

[A] Modulation rates for the resonant case and for varying values of the field strength :

Both the applied field modes are exactly resonant with the respective atomic frequencies ($\Delta_1=0=\Delta_2$) and the decay constant has been taken to be the same for both the excited states ($\gamma_1=1=\gamma_2$). The laser exciting the $|2\rangle \longleftrightarrow |3\rangle$ transition is taken to be intense ($\alpha_2/\gamma_2=10$). We study the characteristics of the modulation rate for increasing values of the laser 1 intensity. Figure 23 shows the modulation rate A_1 (corresponding to linear response of $\tilde{\rho}_{11}(\infty)$ to modulation of laser 1) as a function of ν_1. The resonance at $\nu_1=\alpha_o(\simeq \alpha_2)$ is the predominant one (full curve), there being no peak at $\nu_1 = 2\alpha_o$. As the strength of the laser field 1 is increased ($\alpha_1/\gamma_1=5$, broken curve), the magnitude of the resonance increases , while when both the interaction energies are equal ($\alpha_1/\gamma_1=10=\alpha_2/\gamma_2$, chain curve) the resonances at $\nu_1=0$, α_o, $2\alpha_o$ are clearly resolvable, the peak at $\nu_1=\alpha_o$ being the

most intense. This also follows from our analytical results of 5.2. The expression (5.2.23) shows that the peak at α_0 occurs even in the zeroth order in the secular approximation, whereas the peaks at 0 and $2\alpha_0$ appear only in the first order. Figure 24 is a plot of A_2 (corresponding to linear response of $\tilde{\rho}_{11}(\omega)$ to modulation of laser 2 as a function of ν_2. Increasing the field strength of laser 1 just increases the magnitude of the broad dispersion-like resonances. The behaviour of the modulation rates B_1 and B_2 (corresponding to linear response of $\tilde{\rho}_{22}(\omega)$ to modulation of laser 1 and 2, respectively) is shown in figures 25 and 26, respectively. The former exhibits resonant behaviour for the modulation frequencies $\nu_1=0,2\alpha_0$ only at all field strengths of laser 1, while the latter exhibits a decrease in the magnitude of the dispersion-like resonances as α_1 is increased. It is evident that the resonances in A_1 and B_1 (corresponding to the modulation of laser 1) yield the energy spectra of the composite system consisting of the atom and the coherent laser fields. This should be contrasted with the zeroth-order fluorescence (unmodulated) which does not exhibit dynamic Stark splittings if both the lasers are very intense[5].

[B] Off-resonant modulation rates :

We now present the numerical results for the case when both the exciting laser fields are strong but are different

in frequencies from the respective resonant atomic frequencies. In this case also the analytical results can be obtained by following the procedure of 5.2. -- However the explicit results are extremely cumbersome and hence these are not presented here. The interaction energies for both the transitions are taken equal ($\alpha_1/\gamma_1=10=\alpha_2/\gamma_2$). If the strong laser fields are detuned from resonance by equal amounts ($\Delta_1=2=\Delta_2$), the behaviour of A_1 is not much different from that of the exact resonance case (figure 23, chain curve) except for a slight increase in the peak heights. However if the detuning of the laser 2 is further increased ($\Delta_2=5, \Delta_1=2$, figure 27, broken curve) then the peak at $\nu_1=\alpha_o$ splits into two, no such splitting being observed were the detuning of laser 1 to be increased only ($\Delta_1=5, \Delta_2=2$, full curve). When $\Delta_1=5=\Delta_2$ there is again no splitting, which clearly reveals that the additional resonant component in A_1 appears only when the two applied fields are detuned differently, such that $\Delta_2 > \Delta_1$. Figure 28 gives the behaviour of A_2 which shows that there is only a decrease in the magnitude of the resonances with increased detunings of the laser fields. A similar splitting is observed in B_1 (figure 29) for the reverse case ($\Delta_1 > \Delta_2$) at the modulation frequency $\nu_1=\alpha_o$; however B_2 does not show this effect (figure 30). On comparing figure 26 with figure 30, we see that the detuning leads to extra resonant structures in B_2 around $\nu_2 \sim \alpha_o$.

CHAPTER - VI

RESOLUTION BEYOND NATURAL LINEWIDTH VIA FLUORESCENCE IN MODULATED BEAM

One of the major problems of high resolution spectroscopy[28,49] is to achieve a resolution beyond natural linewidth. Recently several techniques[49-56] have been proposed involving transient response of atoms to pulsed excitation in order to obtain lines narrower than natural linewidth. For example, the time resolved fluorescence in the context of Hanle type of experiments, has the following intensity for an atom having a Zeeman splitting ω_o[49]

$$I(\omega_o,t) = [\ A + B \cos \omega_o t\ ]\ e^{-\Gamma t} \qquad (6.1)$$

where Γ^{-1} is the natural lifetime of the excited state, and the constant A and B are related to the pulse excitation parameters. In the usual quantum beat signal one observes $I(\omega_o,t)$ for fixed ω_o and varied times. In time-delayed level-crossing experiments, one fixes a time interval t_1-t_2 and studies the signal $I(\omega_o, t_1,t_2)$ integrated in this interval as a function of ω_o :

$$I(\omega_o, t_1 ,t_2) = \int_{t_1}^{t_2} (A+B \cos \omega_o t)\ e^{-\Gamma t}\ dt \qquad (6.2)$$

When $t_2 = \infty$, we obtain the simple result

$$I(\omega_o, t_1, t_2) = e^{-\Gamma t_1}\left(\frac{A}{\Gamma} + \frac{B\Gamma}{\Gamma^2+\omega_o^2}\cos\omega_o t_1 - \frac{B\omega_o}{\Gamma^2+\omega_o^2}\sin\omega_o t_1\right) \tag{6.3}$$

Studied as a function of ω_o, the delayed signal $I(\omega_o, t_1, \infty)$ exhibits a central peak around $\omega_o=0$ with a width which for $t_1 > 1/\Gamma$, is inversely proportional to t_1. The narrow width of the central peak allows one to separate two peaks unresolved in the natural linewidth. For zero time delay t_1, one obtains the well known level-crossing signal[25-33], which has a Lorentzian shaped curve around $\omega_o=0$ with a width Γ. However, there is a problem as oscillatory structures appear in the wings due to the detection having a step function sensitivity, and these have to be suppressed. It may be added that lines narrower than natural linewidth (limited by laser linewidth) have been obtained in several other experiments.[15-18]

In this chapter, we study a new method of achieving resolution beyond natural linewidth. This method does not use pulsed excitations but uses continuous wave sources that are weakly modulated in time at low frequencies. The experiment obviously is to be performed in the Doppler free situation - this could be done in an orthogonal geometry involving atomic

beams.[15-18] We consider the following experimental arrangement : a well collimated atomic beam is irradiated orthogonally by laser beam . The amplitude, and hence the intensity, of the laser beam is weakly modulated at a low frequency. The fluorescence modulated at twice this frequency is detected by a phase sensitive detector along a mutually perpendicular direction. The signal is scanned by tuning the laser frequency through the optical transition. We proceed by considering the incident field to be nearly resonant with the transition between the two levels denoted by $|1\rangle$ and $|2\rangle$ having the energies $\frac{1}{2}\omega$ and $-\frac{1}{2}\omega$ respectively. The electric field of the laser beam is described classically as in equation (2.1.7)

$$\vec{E}(t) = \frac{1}{2}\vec{\mathcal{E}}_o\,(1+a\,e^{i\theta}\sin\frac{\nu t}{2})\,e^{-i\Omega t} + c.c. \qquad (6.4)$$

where θ is the time-independent but random phase associated with modulation, and the other symbols have the usual meaning introduced in section (2.1). Note that equation (6.4) implies that the time-averaged intensity of the laser beam after averaging over the random distribution of θ is given by

$$I_{in}(t) \cong \frac{1}{2}\mathcal{E}_o^2\,(1 - \frac{a^2}{2}\cos\nu t) \qquad (6.5)$$

so that the intensity of the laser beam is modulated at ν .

As discussed earlier in section (3.1), the interaction of the atoms with the vacuum of the radiation fields leads to the following Bloch equations for the mean values :

$$\langle \dot{\tilde{S}}^{+}(t) \rangle = (-\gamma + i\Delta) \langle \tilde{S}^{+}(t) \rangle - 2i\alpha(t) \langle \tilde{S}^{z}(t) \rangle$$

$$\langle \dot{\tilde{S}}^{z}(t) \rangle = -2\gamma \left(\langle \tilde{S}^{z}(t) \rangle + \tfrac{1}{2} \right) + [-i\alpha(t) \langle \tilde{S}^{+}(t) \rangle + \text{c.c.}] \qquad (6.6)$$

where

$$\Delta = \omega - \Omega \qquad (6.7)$$

and where

$$\alpha(t) = -\tfrac{1}{2} \vec{d}_{12} \cdot \left[\vec{\mathcal{E}}_0 \left(1 + a\, e^{i\theta} \sin \tfrac{\nu}{2} t\right) \right]$$

$$\equiv \alpha \left(1 + a\, e^{i\theta} \sin \tfrac{\nu}{2} t\right) \qquad (6.8)$$

gives the coupling of the modulated laser field with the $|1\rangle \leftrightarrow |2\rangle$ transition in the dipole approximation. 2γ is equal to Einstein A coefficient and tilde denotes a quantity in a frame rotating with angular frequency Ω. In deriving (6.6), we have neglected terms oscillating as $e^{\pm 2i\Omega t}$. If the laser field is weak, then equation (6.6) may be solved within the framework of a perturbation theory with $\alpha(t)$ as the expansion parameter. We are interested in obtaining the total intensity of the modulated fluorescence. This has been shown to be proportional to (compare with equation (3.1.10))

$$I_s(t) = \langle S^+(t)\, S^-(t)\rangle = \frac{1}{2} + \langle S^z(t)\rangle \qquad (6.9)$$

By a simple perturbative analysis of equation (6.6), we get to second order in $\alpha(t)$ and in the long time limit

$$\langle S^z(t)\rangle^{(2)} = \int_0^t dt' e^{-2\gamma(t-t')}\alpha(t') \int_0^{t'} dt'' e^{(-\gamma+i\Delta)(t'-t'')}\alpha^*(t'')$$

$$+ \text{c.c.} \qquad (6.10)$$

Substituting the explicit form of $\alpha(t)$ from (6.8) in (6.10) and making use of equations (2.3.8) to (2.3.10), we obtain

$$\langle S^z(\infty)\rangle_{(2)} = \frac{\alpha^2/2}{\gamma^2+\Delta^2} + \frac{a^2\alpha^2}{8}\left[\frac{1}{\gamma^2+(\Delta+\frac{\nu}{2})^2} + \frac{1}{\gamma^2+(\Delta-\frac{\nu}{2})^2}\right]$$

$$-\frac{ia\alpha^2}{2}\,\frac{e^{i\frac{\nu}{2}t}}{2\gamma+i\frac{\nu}{2}}\left[e^{i\theta}\left(\frac{1}{\gamma-i\Delta}+\frac{1}{\gamma+i\Delta+i\frac{\nu}{2}}\right) + e^{-i\theta}\left(\frac{1}{\gamma+i\Delta}+\frac{1}{\gamma-i\Delta+i\frac{\nu}{2}}\right)\right]$$

$$-\frac{a^2\alpha^2}{4}\,\frac{e^{i\nu t}}{2\gamma+i\nu}\left(\frac{1}{\gamma+i\Delta+i\frac{\nu}{2}}+\frac{1}{\gamma-i\Delta+i\frac{\nu}{2}}\right) + \text{c.c.} \qquad (6.11)$$

On averaging over the random distribution of θ, the terms that are linear in the modulation ratio a vanish. If the modulation frequency is much smaller than the laser detuning

and the spontaneous emission rate i.e. $\nu \ll \Delta, \gamma$, then the above result and Eq. (6.8) lead to

$$I_{s,(2)} = \frac{\alpha^2}{\gamma^2+\Delta^2}\left(1+\frac{a^2}{2}\right) - \frac{a^2\alpha^2}{2(\gamma^2+\Delta^2)}\cos \nu t \qquad (6.12)$$

Thus the amplitude of cosine component of the modulated fluorescence oscillating at a frequency ν has a resonance centred at $\Omega = \omega$ with a FWHM equal to the natural width 2γ.

We now study the component of the modulated fluorescence oscillating at a frequency 2ν, this occuring to fourth order in modulation ratio a. It may be noted that an ingenious method based on modulated fluorescence, which is also fourth order in the modulationratio, has been used by Sorem and Schawlow[24] and other workers[90] to obtain Doppler free signals for samples in cell. Perturbative calculations of equations (6.6) yield

$$\langle S^z(t)\rangle_{(4)} = -2\int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_3} dt_4 \; e^{-\frac{1}{T_1}(t-t_1)}\alpha(t_1) \times$$
$$e^{(-\frac{1}{T_2}+i\Delta)(t_1-t_2)}\alpha^*(t_2)\, e^{-\frac{1}{T_1}(t_2-t_3)} \times$$
$$\left\{\alpha(t_3)e^{(-\frac{1}{T_2}+i\Delta)(t_3-t_4)}\alpha^*(t_4)+\alpha^*(t_3)\, e^{(-\frac{1}{T_2}-i\Delta)(t_3-t_4)}\alpha(t_4)\right\} + \text{c.c.} \qquad (6.13)$$

Substituting the explicit form of $\alpha(t)$ from equation (6.8) and making use of equations (2.3.8) to (2.3.10), we obtain

$$I_{s,(4)} = \eta^{(+)} e^{2i\nu t} + \eta^{(-)} e^{-2i\nu t} + \text{components oscillating at other multiples of } \frac{\nu}{2} \tag{6.14}$$

where

$$\eta^{(+)} = \eta^{(-)} = -\frac{a^4\alpha^4}{8}\left[\frac{1}{(2\gamma+2i\nu)(\gamma-i\Delta-\frac{3i\nu}{2})(2\gamma+i\nu)(\gamma-i\Delta+i\frac{\nu}{2})}\right.$$

$$+\frac{1}{(2\gamma+2i\nu)(\gamma+i\Delta+\frac{3i\nu}{2})(2\gamma+i\nu)(\gamma+i\Delta+\frac{i\nu}{2})}$$

$$+\frac{1}{(2\gamma+2i\nu)(\gamma-i\Delta+3\frac{i\nu}{2})(2\gamma+i\nu)(\gamma+i\Delta+\frac{i\nu}{2})}$$

$$\left.+\frac{1}{(2\gamma+2i\nu)(\gamma+i\Delta+\frac{3i\nu}{2})(2\gamma+i\nu)(\gamma-i\Delta+\frac{i\nu}{2})}\right] \tag{6.15}$$

For small modulation frequency ($\nu \ll \Delta, \gamma$), the fluorescence oscillating at 2ν is given by

$$I_{s,(4)} = \frac{-a^4\alpha^4}{4}\cos 2\nu t \ \frac{1}{(\gamma^2+\Delta^2)^2} \tag{6.16}$$

Thus the modulated fluorescence at 2Ω' has a rather simple structure and interpretation. The value of Δ at which the value is half of the peak-value occurs at

$$\Delta/\gamma = \pm (\sqrt{2}-1)^{\frac{1}{2}} = \pm .6434 \qquad (6.17)$$

so that the FWHM of the resonance centered at $\Omega = \omega$ is of the order of 1.3γ, which is considerably less than the natural linewidth of 2γ. Thus we have an effective and rather simple method for resolving closely-spaced spectral lines even when their frequency difference is less than the radiative linewidth. Thus CW methods can be used to study closely lying lines. It is hoped that the technique studied in this chapter would provide an alternative to the techniques used in time resolved experiments.

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FIGURE CAPTIONS

Fig. 1 Phase shift of the fluorescence as a function of the modulation frequency (in units of T_2^{-1}) for $\alpha T_2 = 0.1$, $\Delta = 0$. The values of the relaxation parameters are

(a) $\frac{1}{T_1} = \frac{10}{T_2}$ (b) $\frac{1}{T_1} = \frac{2}{T_2}$

(c) $\frac{1}{T_1} = \frac{1}{T_2}$ (d) $\frac{1}{T_1} = \frac{1}{10\ T_2}$

Fig. 2 Amplitude of modulation of the fluorescent light as a function of the modulation frequency. The values of the parameters are the same as in figure (1).

Fig. 3 Same as in figure 1, but with $\alpha T_2 = 10$.

Fig. 4 Same as in figure 2, but with $\alpha T_2 = 10$.

Fig. 5 Phase shift of the fluorescence as a function of the modulation frequency (in units of γ) for

(a) $\gamma_c = 10\gamma$ vertical scale for 1 division = 20.

(b) $\gamma_c = \gamma$ vertical scale for 1 division = 20.

(c) $\gamma_c = \frac{\gamma}{10}$ vertical scale for 1 division = 4.

Fig. 6 Amplitude of modulation of the fluorescent light as a function of the modulation frequency. The values of the parameters are the same as in figure 5.

Fig. 7 Normalised plot of the velocity-averaged amplitude of modulation for both weak ($\frac{\alpha}{\gamma} = 0.1$) and strong ($\frac{\alpha}{\gamma} = 10$) fields in the absence of detunings ($\Delta = 0$). The parameters are

(a) $D = \frac{\gamma}{10}$ (b) $D = \gamma$ (c) $D = 10\gamma$

Fig. 8 Schematic representation of experimental geometry.

Fig. 9 Schematic representation of the relevant energy levels.

Fig.10 Magnetic field scan of the amplitude of the cosine component of the modulated fluorescence detected parallel to H for $\frac{\nu}{\gamma} = 10$ and for the following values of the bandwidth parameter :

(a) ——— $\gamma_c = 0$

(b) -- -- $\gamma_c = \gamma$

(c) —·—·— $\gamma_c = 5\gamma$

Fig.11 Magnetic field scan of the amplitude of the Sine component of the modulated fluorescence detected parallel to H for the same values of the parameters as in Fig. 10.

Fig.12 Magnetic field scan of the amplitude of the cosine component of the modulated fluorescence detected perpendicular to H for the same values of the parameters as in Fig. 10.

Fig.13 Magnetic field scan of the amplitude of the Sine component of the modulated fluorescence detected perpendicular to H for the same values of the parameters as in Fig.10.

Fig.14 Modulation frequency scan of the amplitude of the cosine component of the modulated fluorescence detected parallel to H for $\frac{s}{\gamma} = 5$ and for the following values of the bandwidth parameter :

(a) ——— $\gamma_c = 0$

(b) - - - - $\gamma_c = \gamma$

(c) —•—•— $\gamma_c = 5\gamma$

Fig.15 Modulation frequency scan of the amplitude of the sine component of the modulated fluorescence detected parallel to H for the same values of the parameters as in Fig.14.

Fig.16 Modulation frequency scan of the amplitude of the cosine component of the modulated fluorescence detected perpendicular to H for the same values of the parameters as in Fig.14.

Fig.17 Modulation frequency scan of the amplitude of the cosine component of the modulated fluorescence detected perpendicular to H for the same values of the parameters as in Fig.14.

Fig.18 Same as in figure 14, but for $\frac{\alpha}{\gamma} = 10$.

Fig.19 Same as in figure 15, but for $\frac{\alpha}{\gamma} = 10$.

Fig.20 Same as in figure 16, but for $\frac{\alpha}{\gamma} = 10$.

Fig.21 Same as in figure 17, but for $\frac{\alpha}{\gamma} = 10$.

Fig.22 Schematic diagram of the energy levels of the system showing the various interactions and relaxations.

Fig.23 Amplitude of modulation A_1 corresponding to the linear response of $\rho_{11}(\infty)$ to modulation of laser field 1 as a function of the modulation frequency ν_1 for $\alpha_2 = 10$, $\gamma_1 = 1 = \gamma_2$, $\Delta_1 = 0 = \Delta_2$ and for various values of the laser field strength

(a) ——— $\alpha_1 = 1$

(b) - - - - $\alpha_1 = 5$

(c) —·—· $\alpha_1 = 10$

Fig.24 Amplitude of modulation A_2 corresponding to linear response of $\rho_{11}(\infty)$ to modulation of laser field 2 as a function of ν_2. The values of the parameters are the same as in figure 23.

Fig.25 Amplitude of modulation B_1 corresponding to linear response of $\rho_{22}(\infty)$ to modulation of laser field 1 as a function of ν_1 for the same values of the parameters as in figure 23.

Fig.26 Amplitude of modulation B_2 corresponding to linear response of $\rho_{22}(\infty)$ to modulation of laser field 2 as a function of ν_2 for the same values of the parameters as in figure 23.

Fig.27 Same as in figure 23, but with $\alpha_1 = 10 = \alpha_2$ and the following detunings (a) $\Delta_1 = 5$, $\Delta_2 = 2$ (——) (b) $\Delta_1 = 2$, $\Delta_2 = 5$ (- - - -).

Fig.28 Same as in figure 24, but with $\alpha_1 = 10 = \alpha_2$ and laser detunings (a) $\Delta_1 = 5$, $\Delta_2 = 2$(——) (b) $\Delta_1 = 2$, $\Delta_2 = 5$(- - - -)

Fig.29 Same as in figure 25, but with $\alpha_1 = 10 = \alpha_2$ and laser detunings (a) $\Delta_1 = 5$, $\Delta_2 = 2$ (——) (b) $\Delta_1 = 2$, $\Delta_2 = 5$(- - - -).

Fig.30 Same as in figure 26, but with $\alpha_1 = 10 = \alpha_2$ and laser detunings (a) $\Delta_1 = 5$, $\Delta_2 = 2$(——) (b) $\Delta_1 = 2$, $\Delta_2 = 5$ (- - - -).

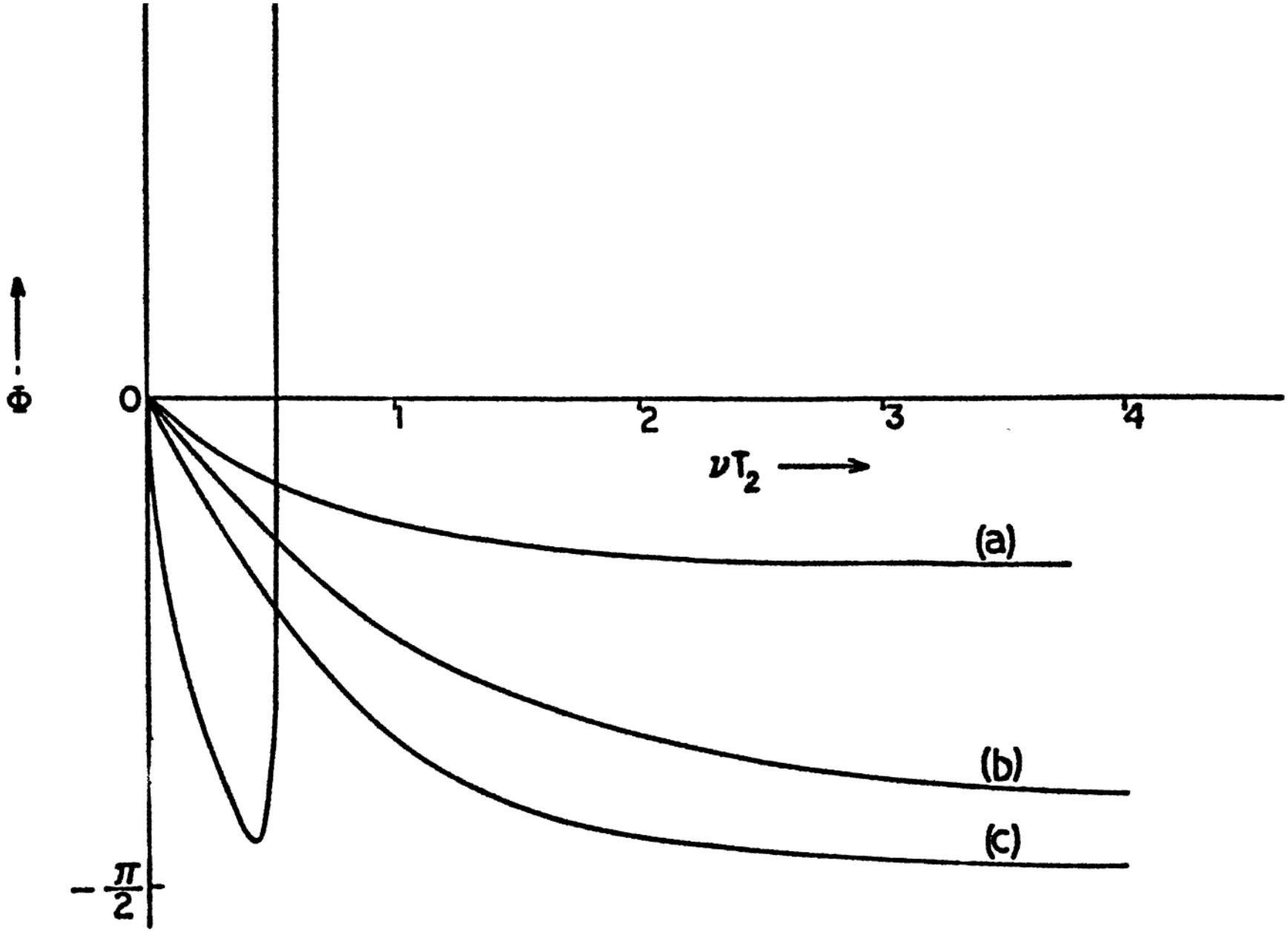


FIG. 1

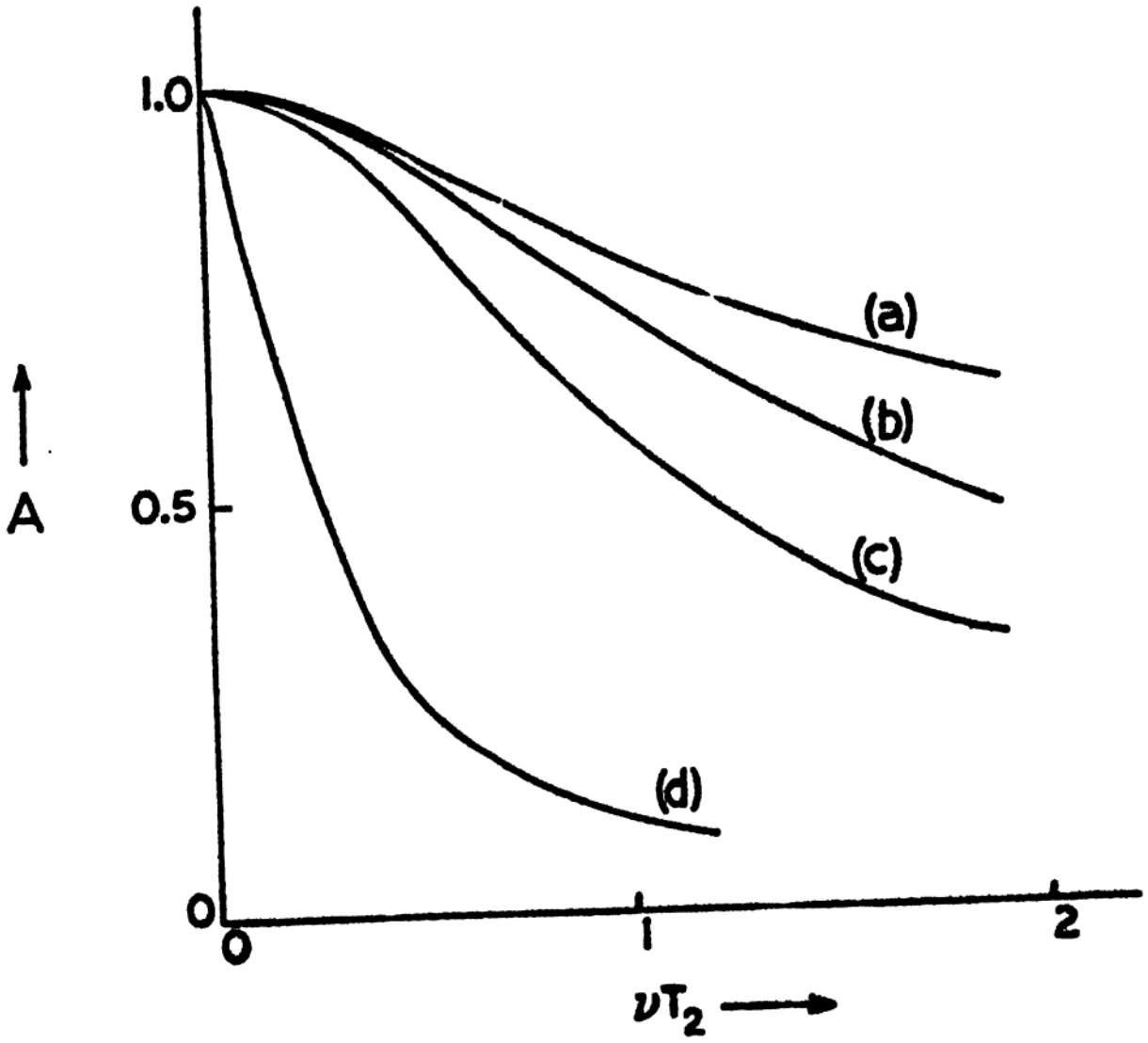
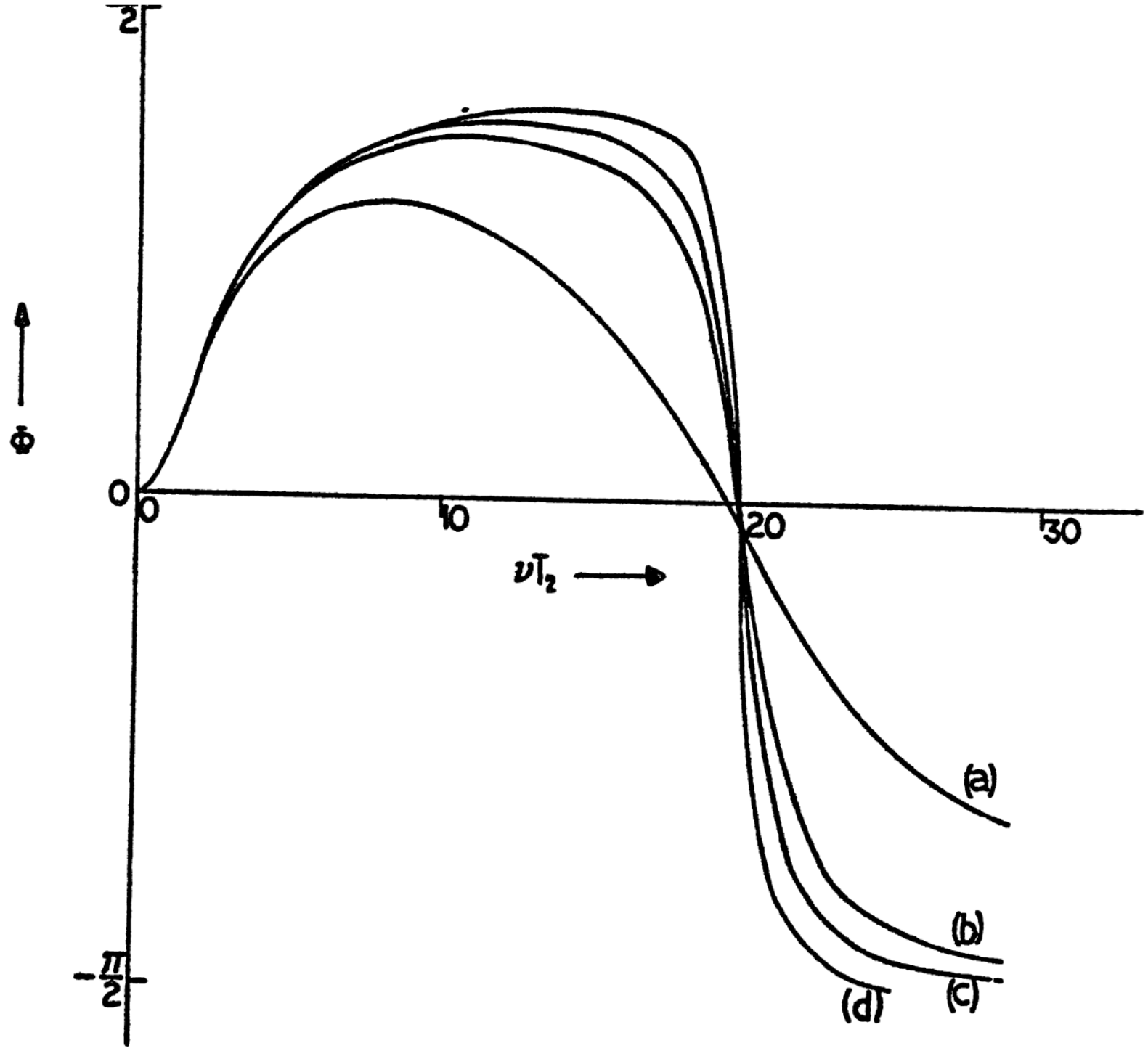


FIG. 2



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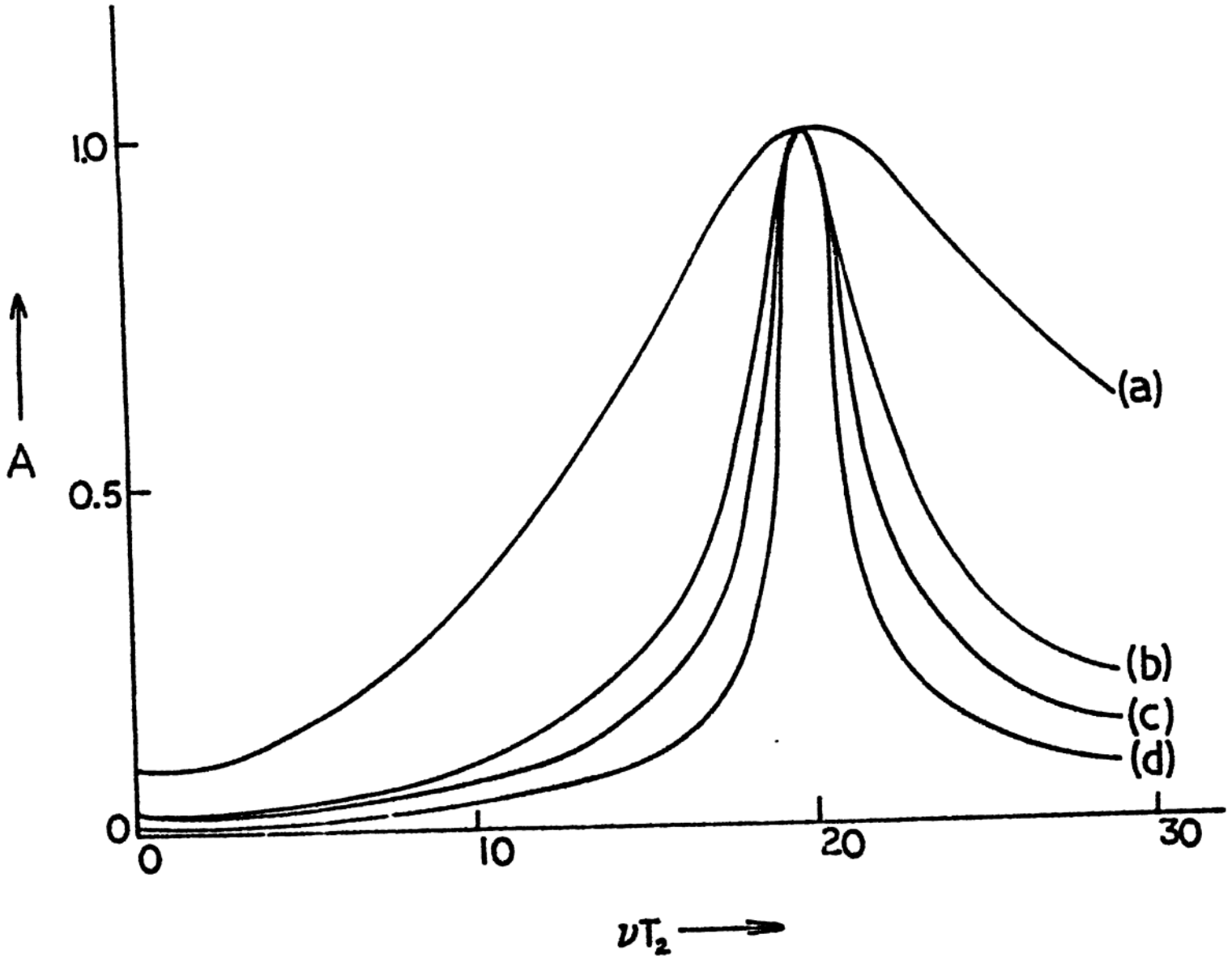


FIG. 4

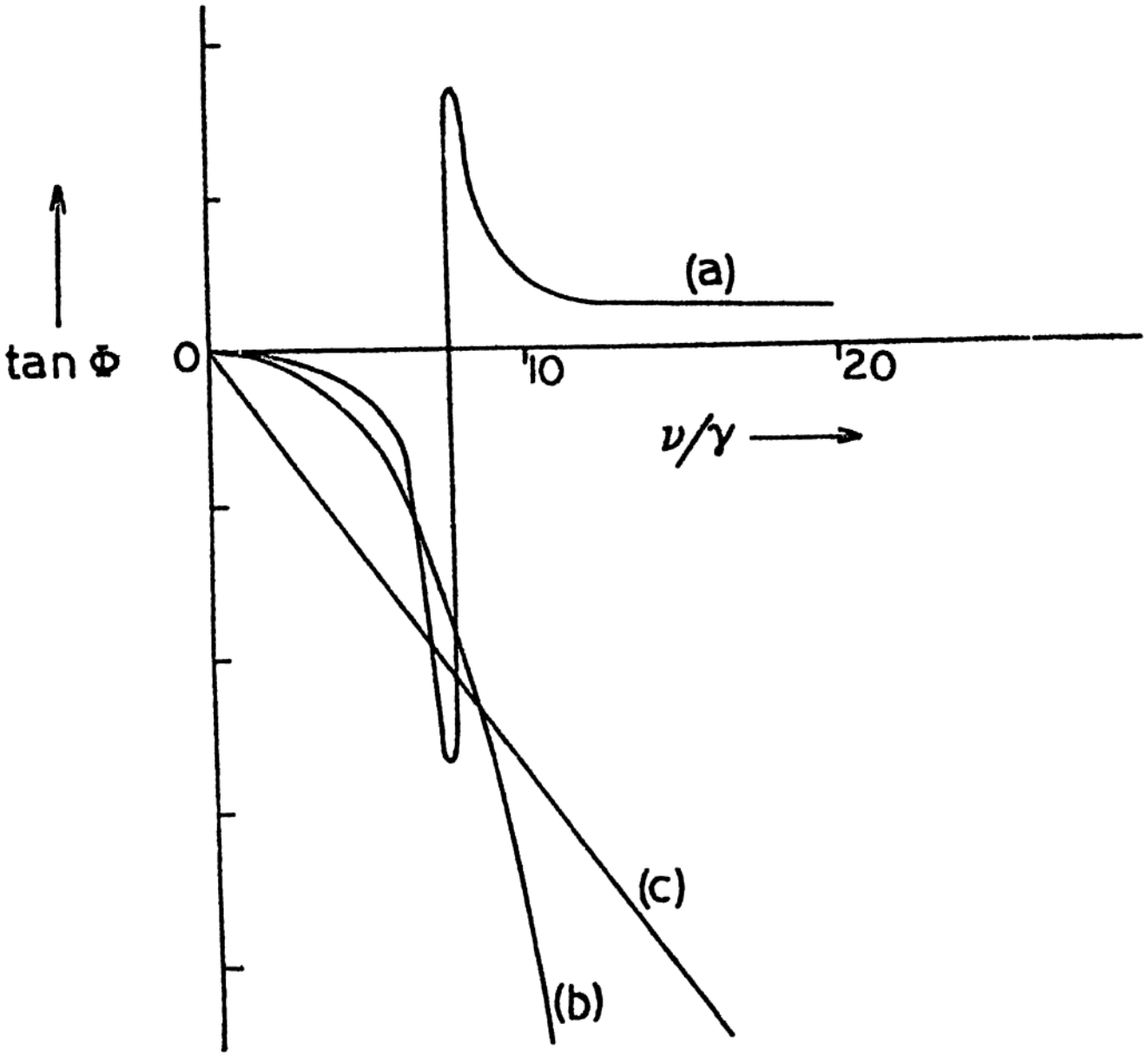
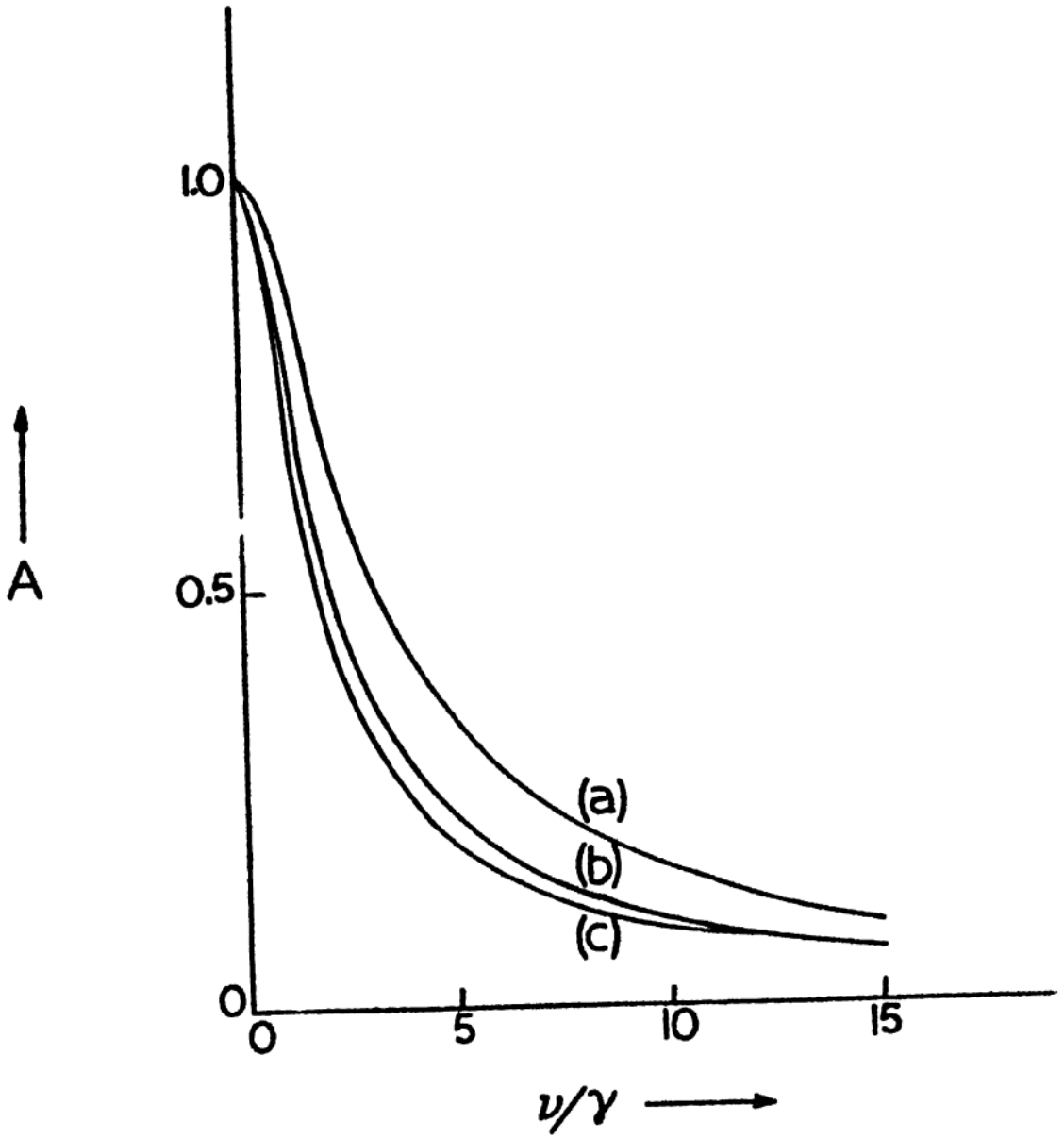


FIG. 5



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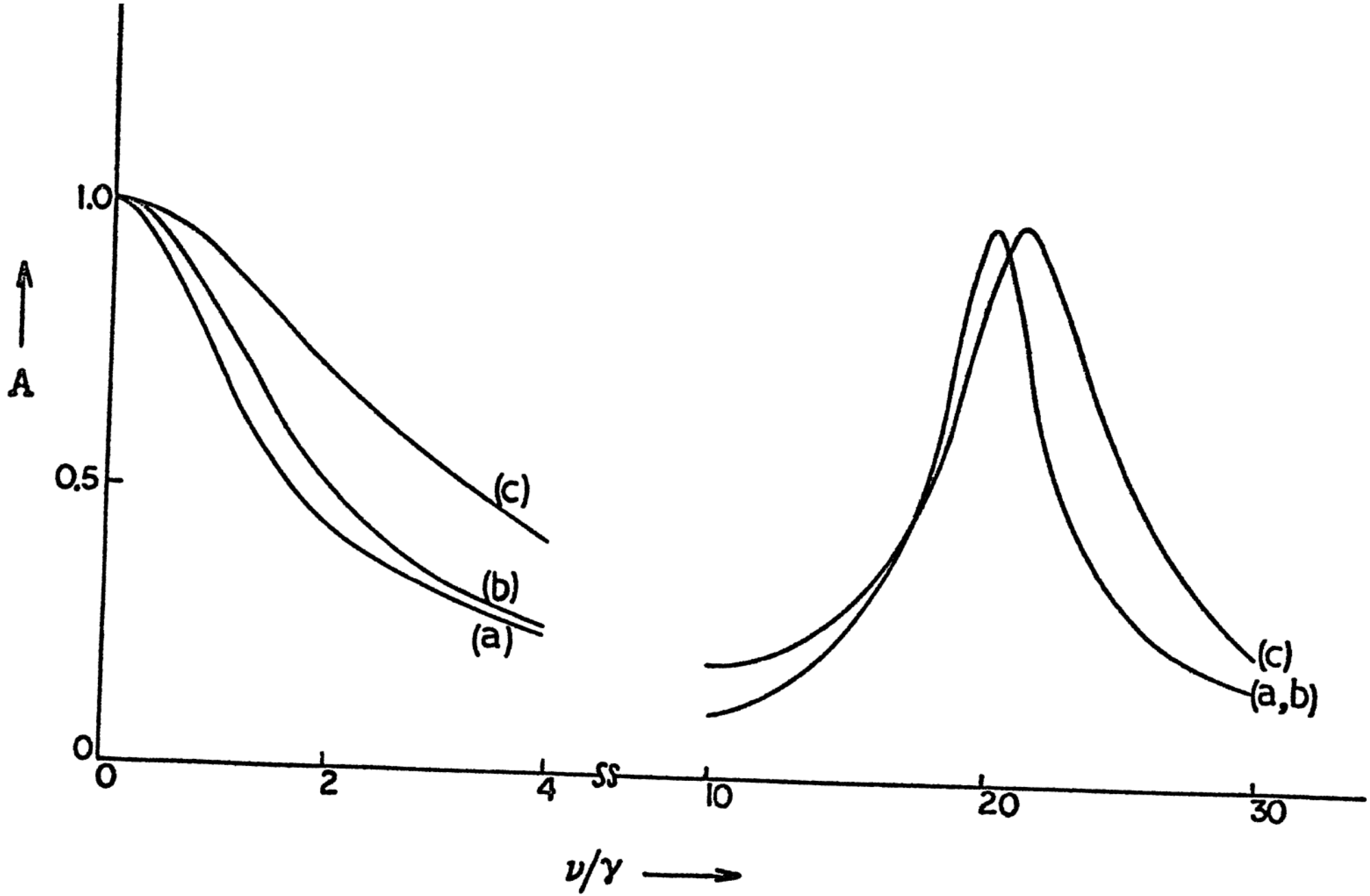


FIG. 7

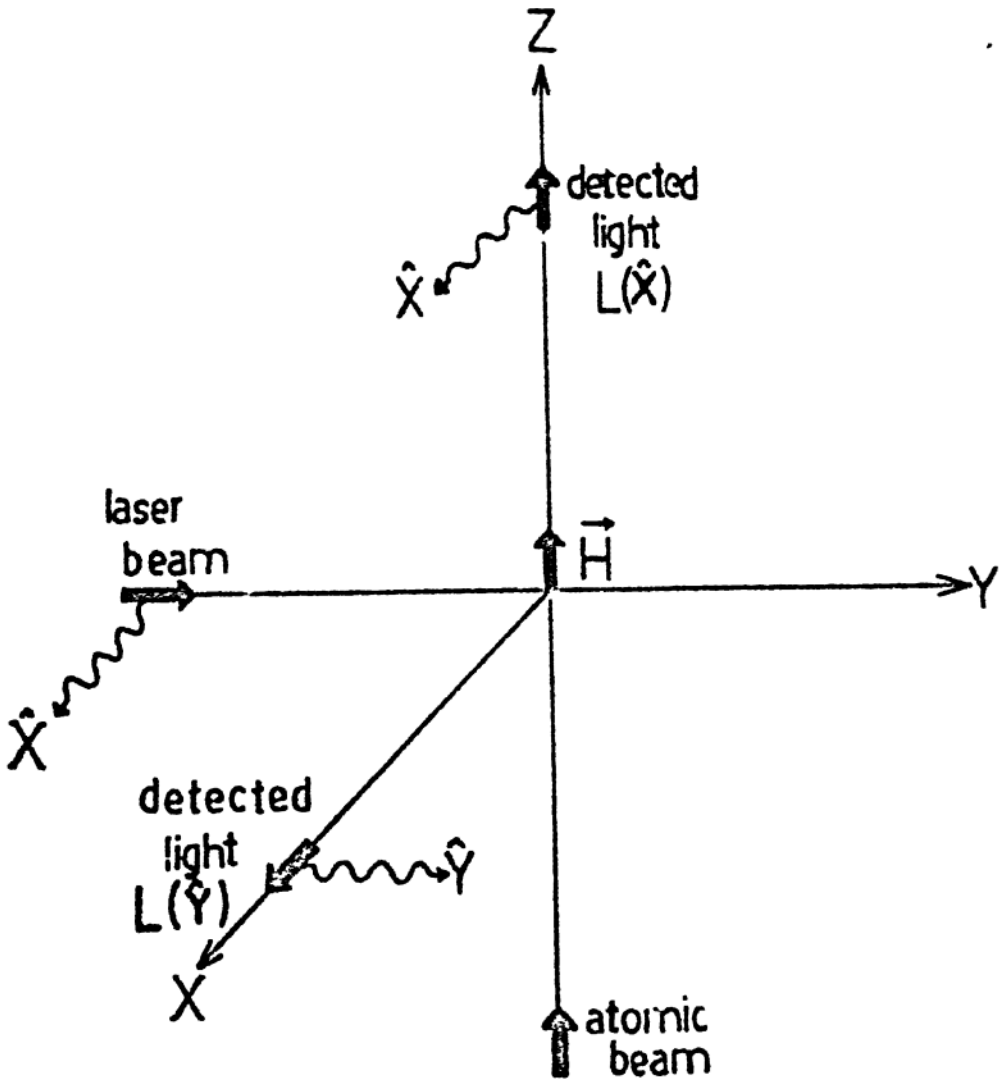

Z
detected
light
$L(\hat{X})$
$\hat{X}$
laser
beam
$\vec{H}$
Y
$\hat{X}$
detected
light
$L(\hat{Y})$
$\hat{Y}$
X
atomic
beam

FIG. 8

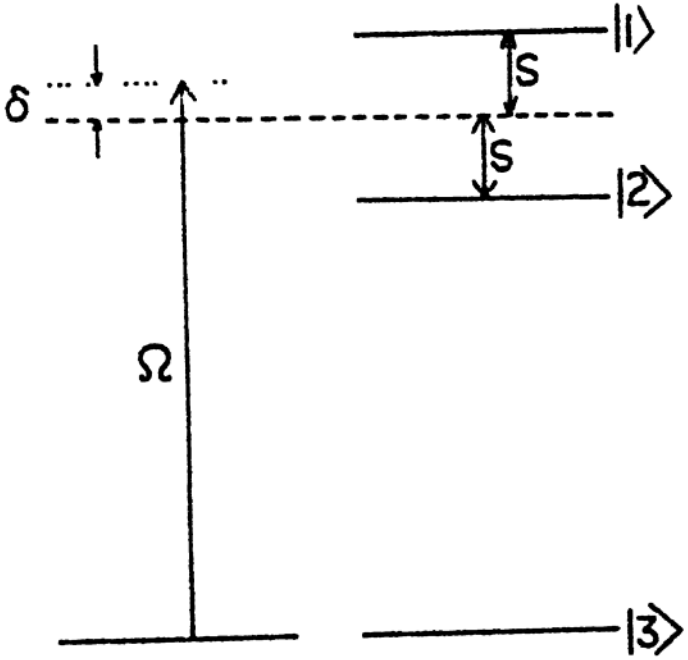

|1⟩
S
δ
S
|2⟩
Ω
|3⟩

FIG. 9

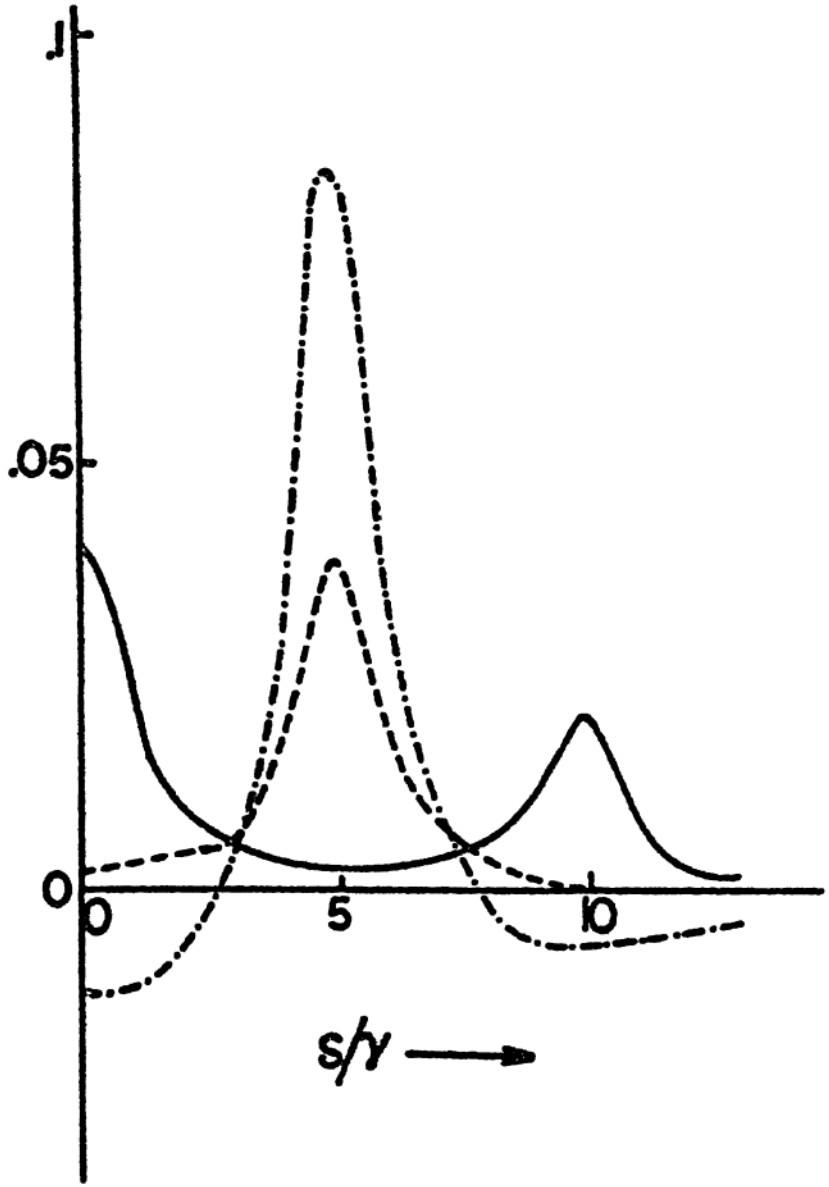


FIG. 10

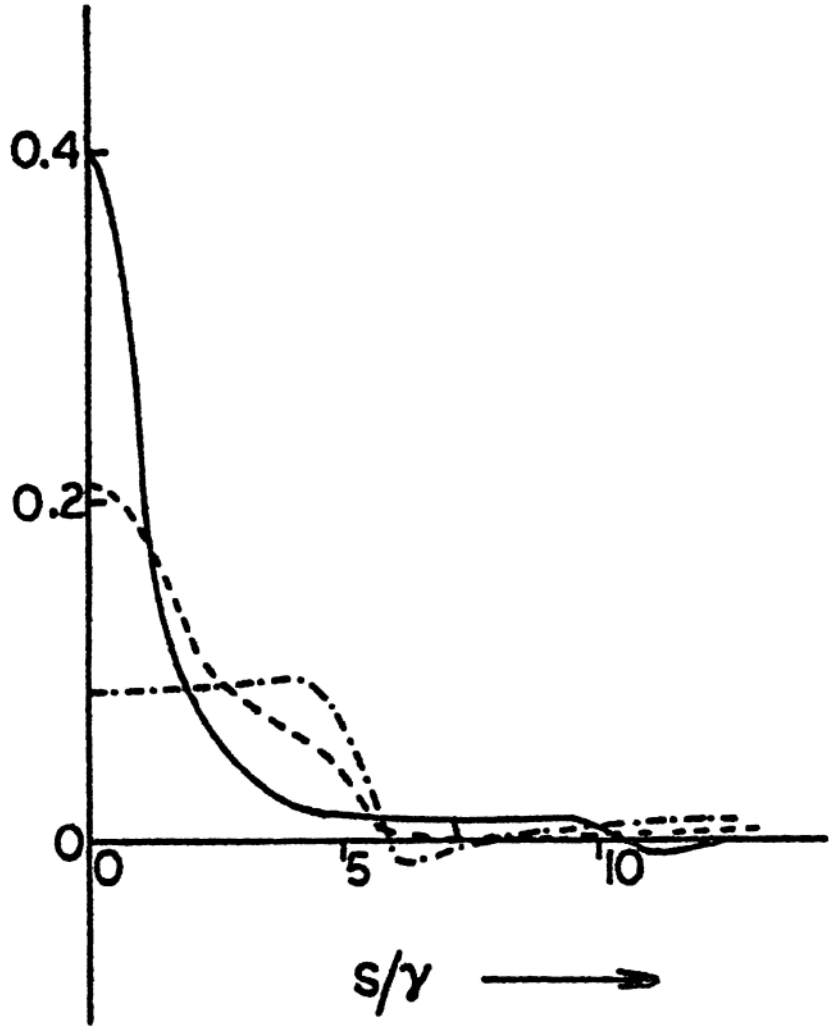


FIG. 11

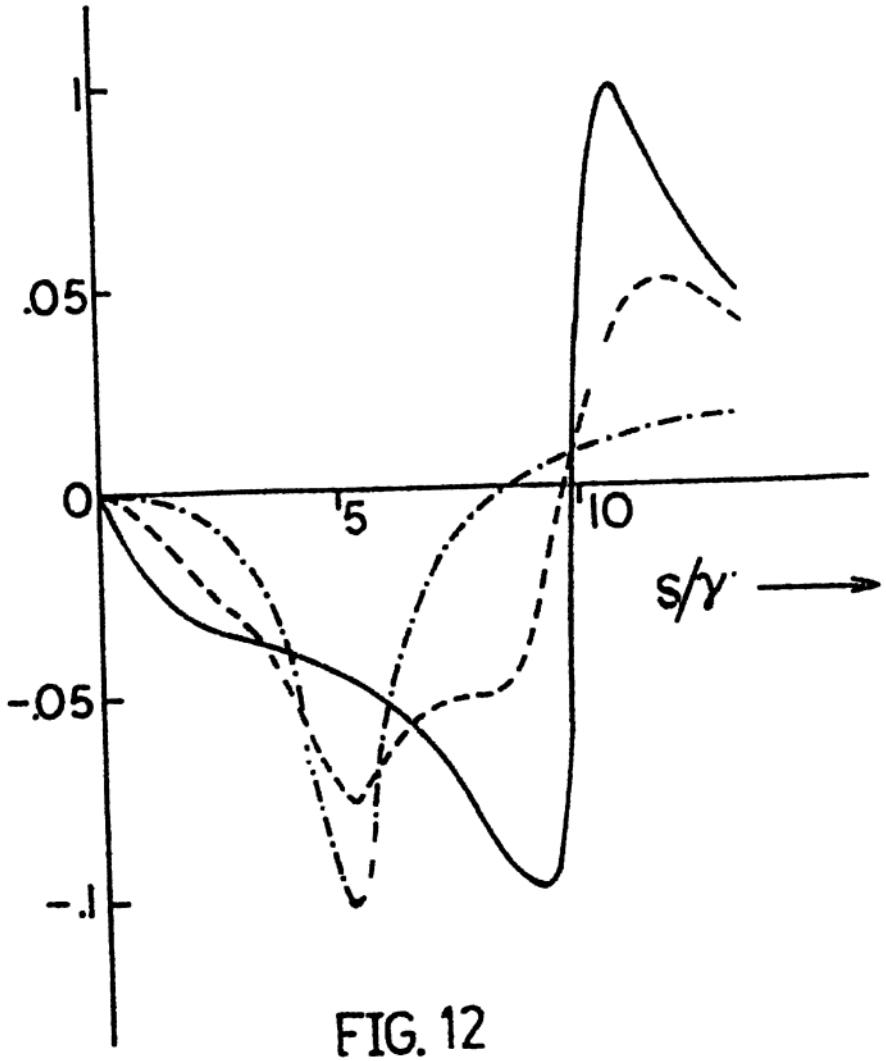


FIG. 12

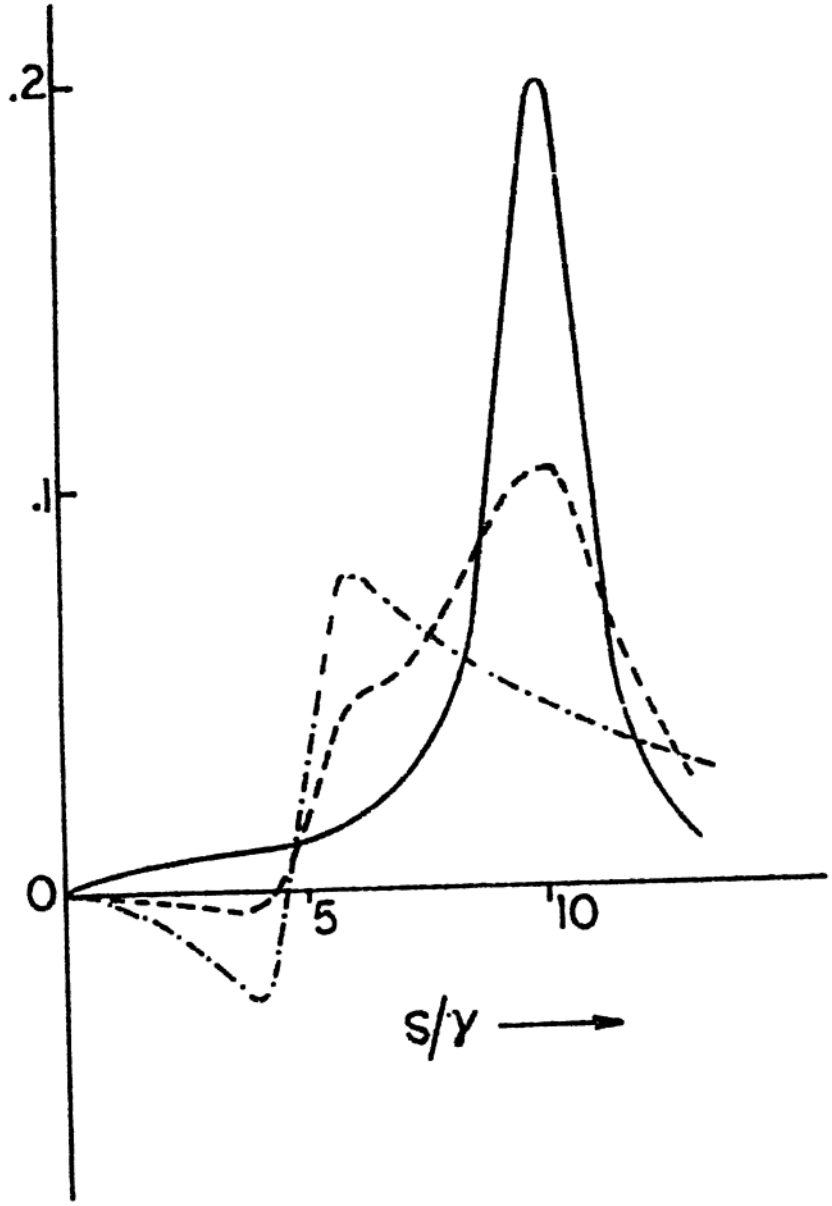


FIG. 13

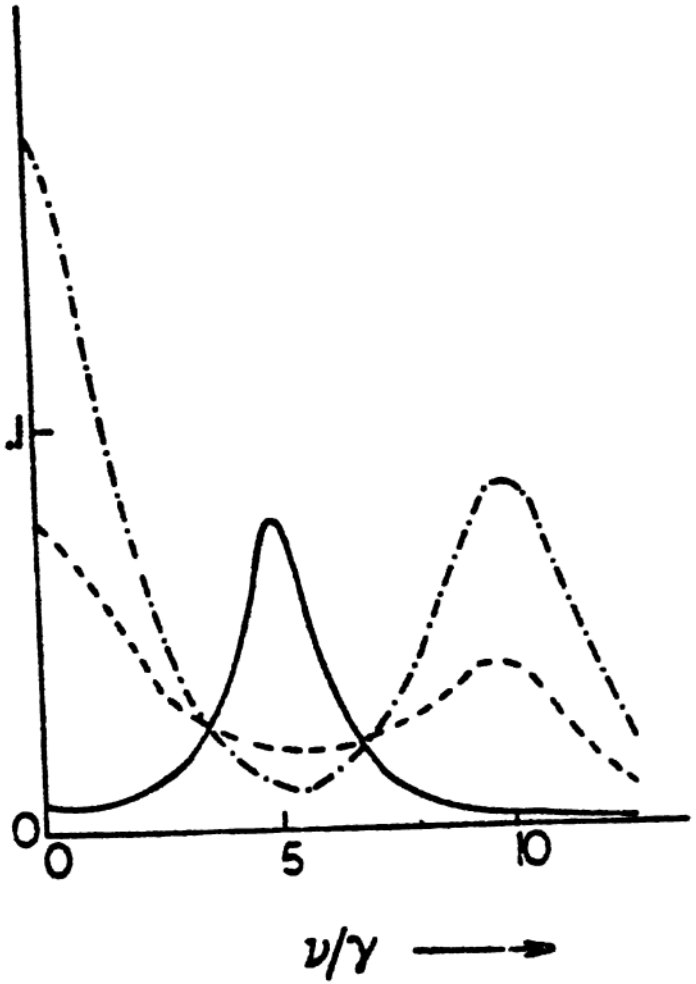


FIG. 14

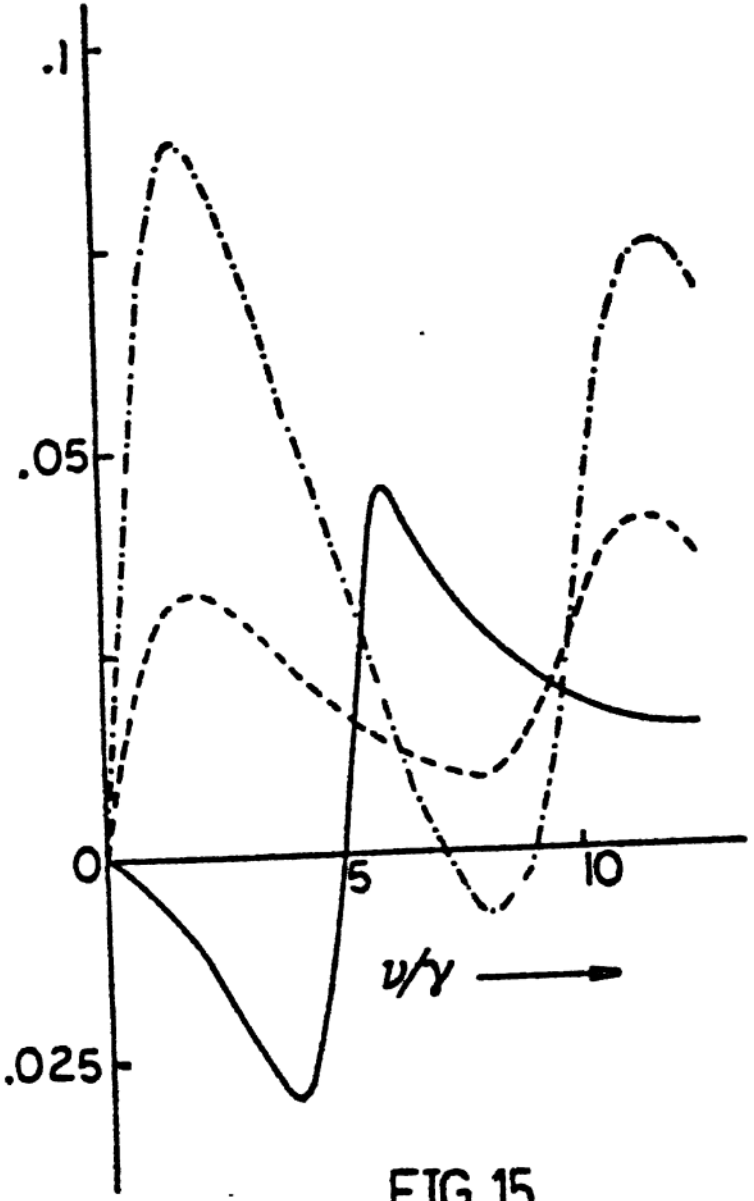


FIG. 15

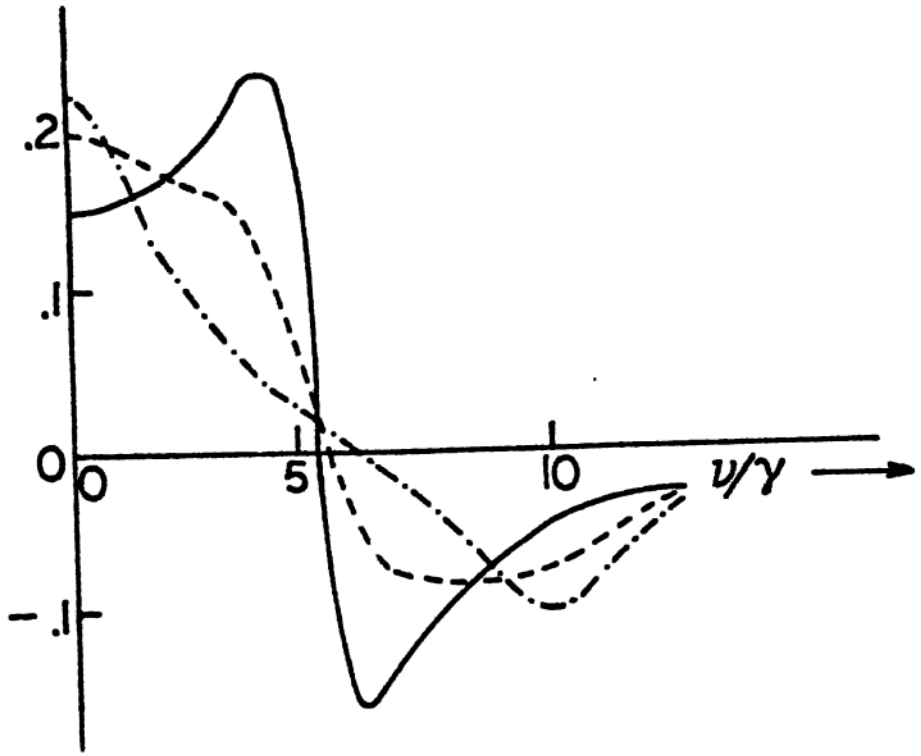


FIG. 16

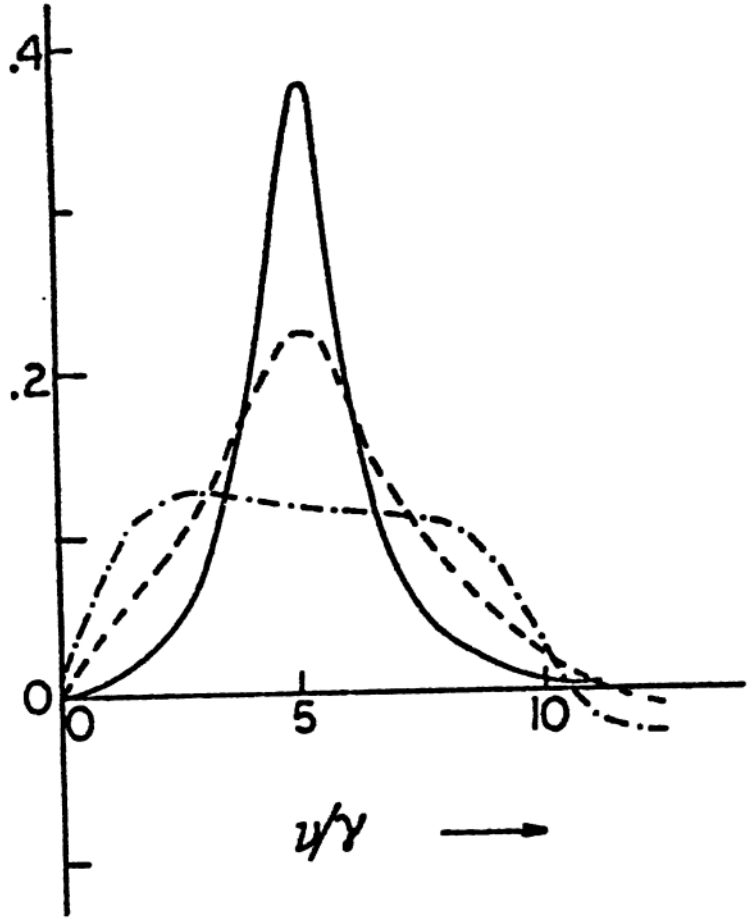


FIG. 17

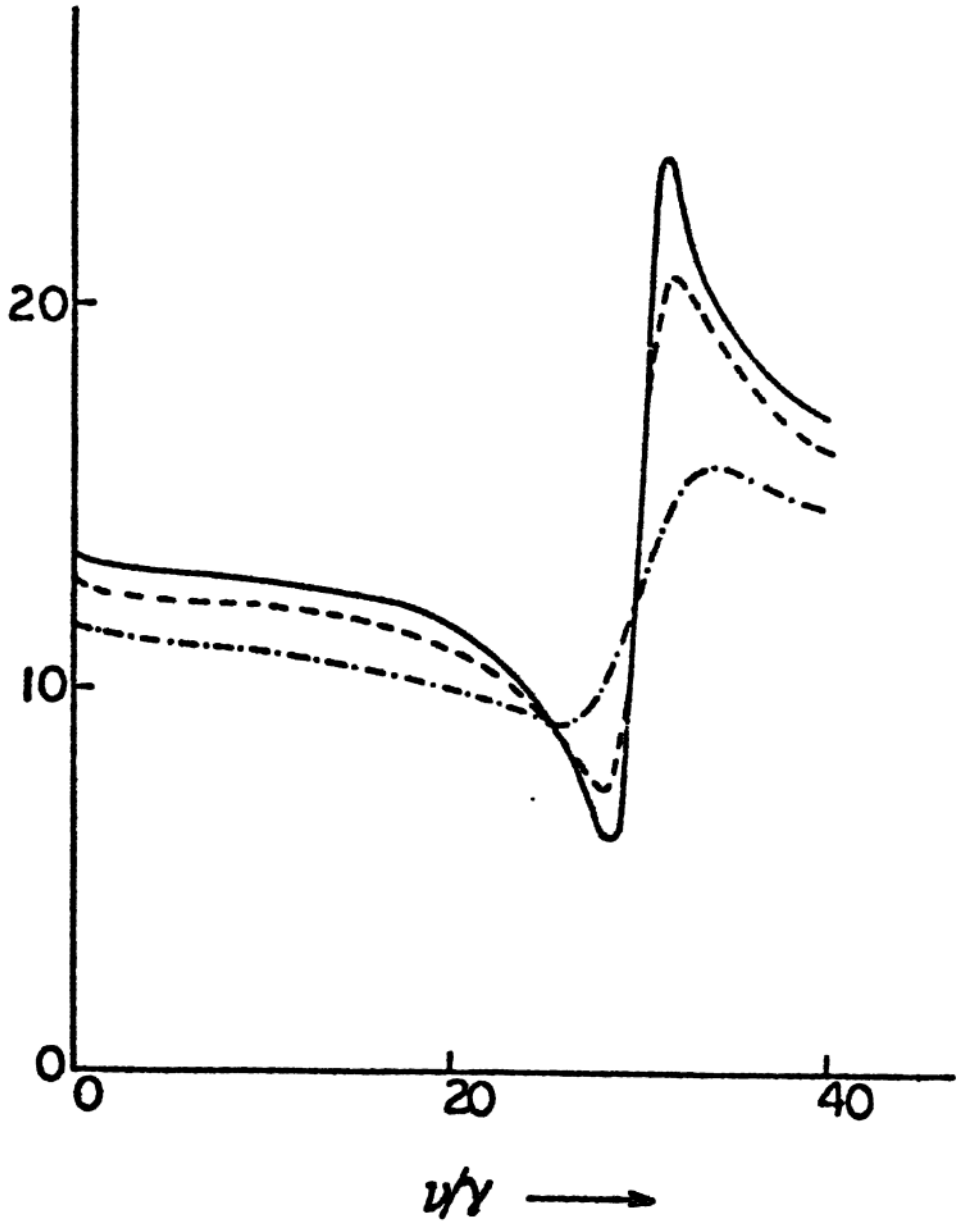


FIG 18

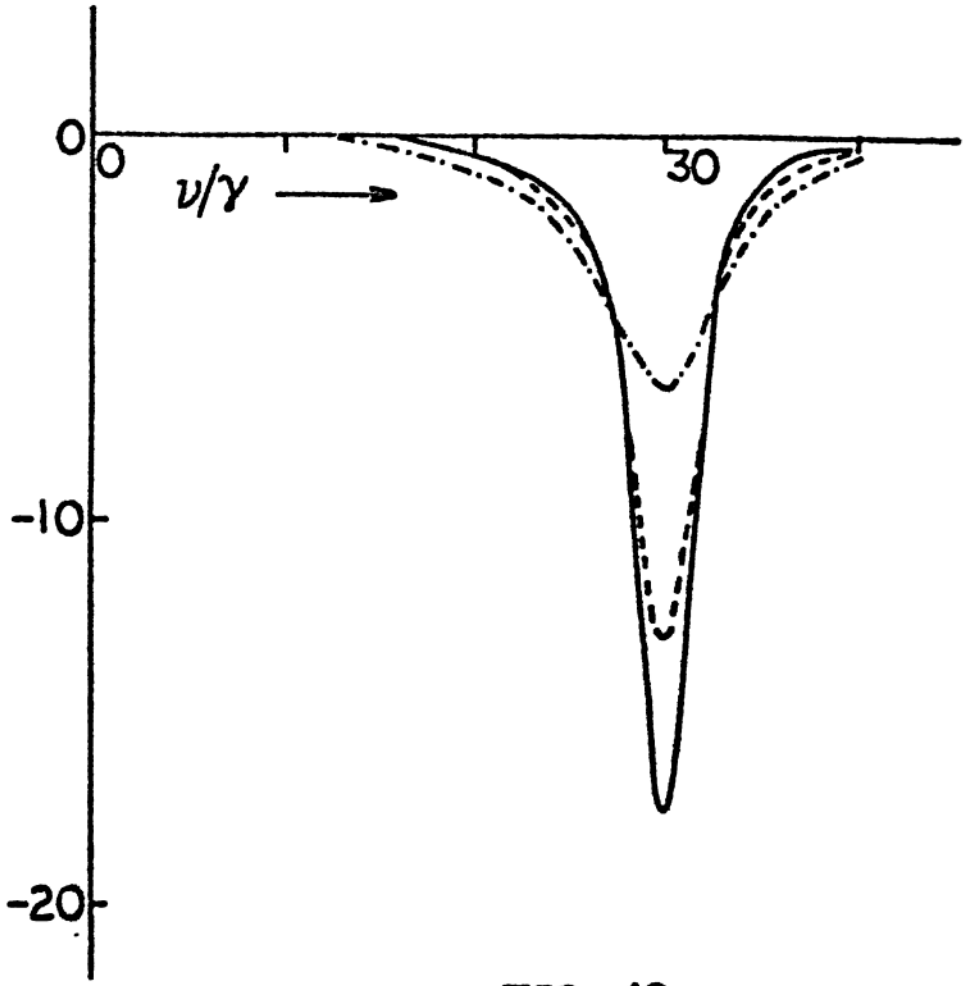


FIG. 19

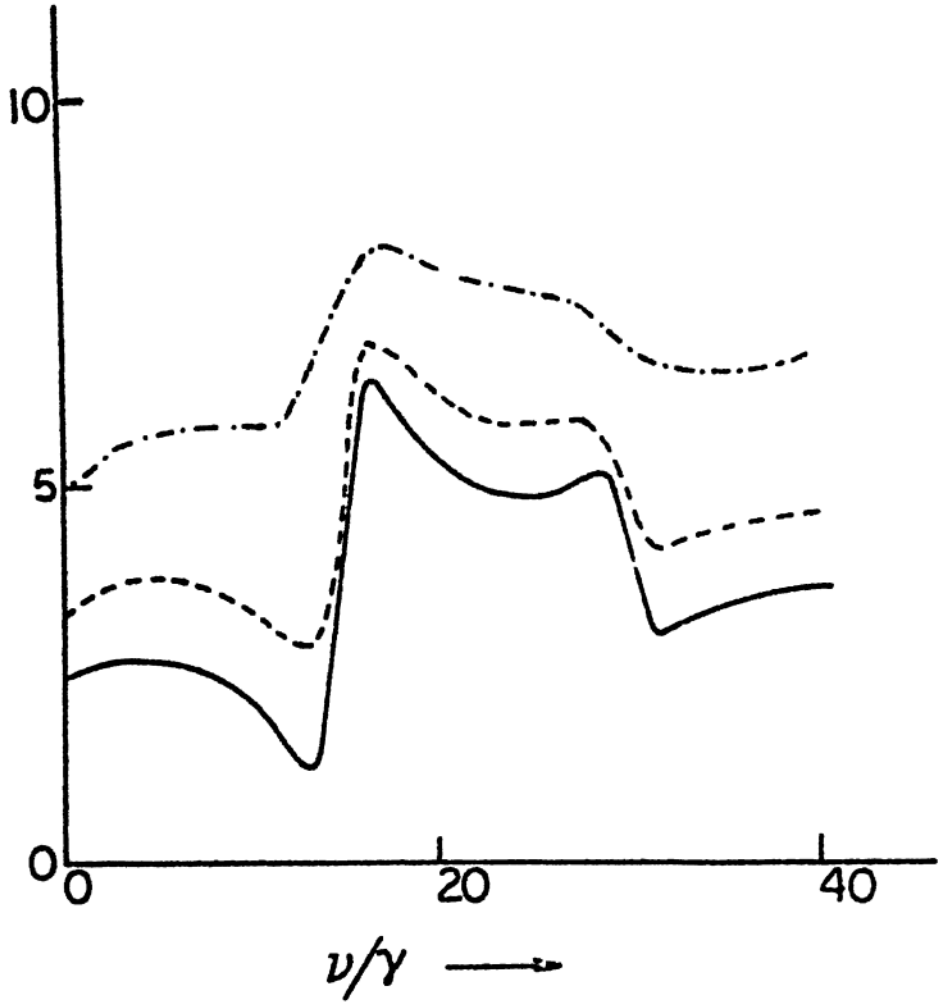


FIG. 20

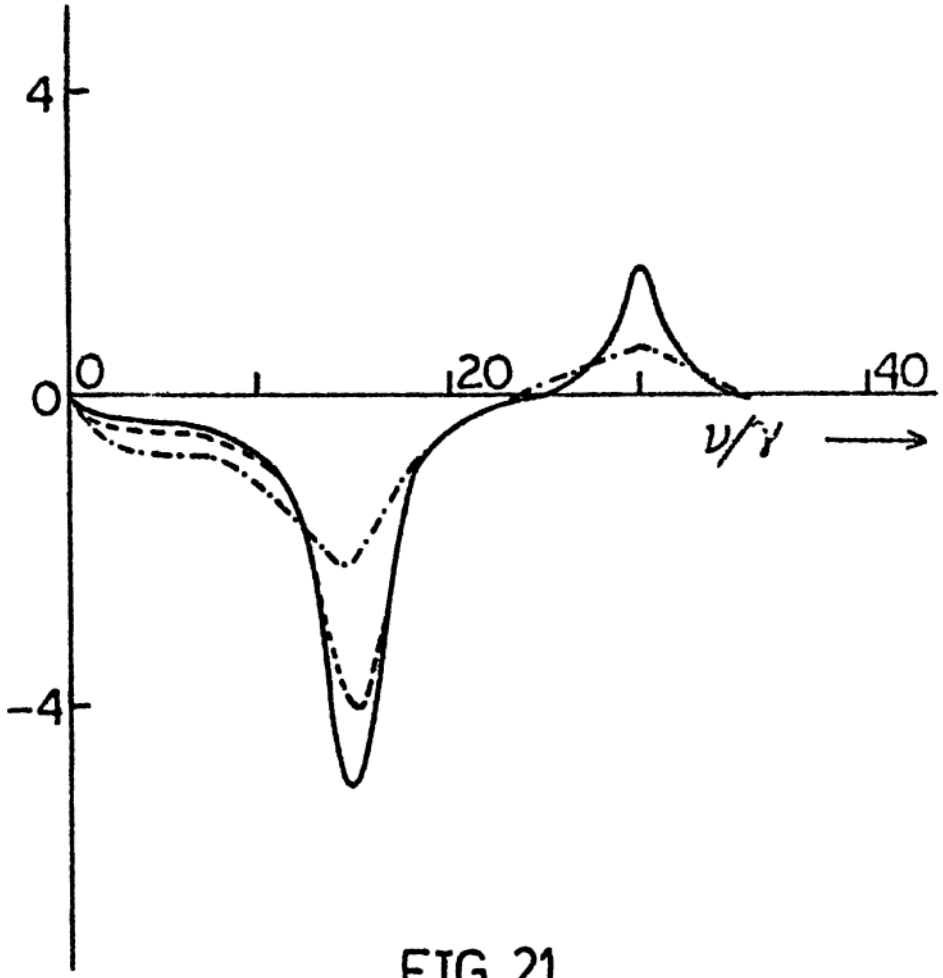


FIG. 21

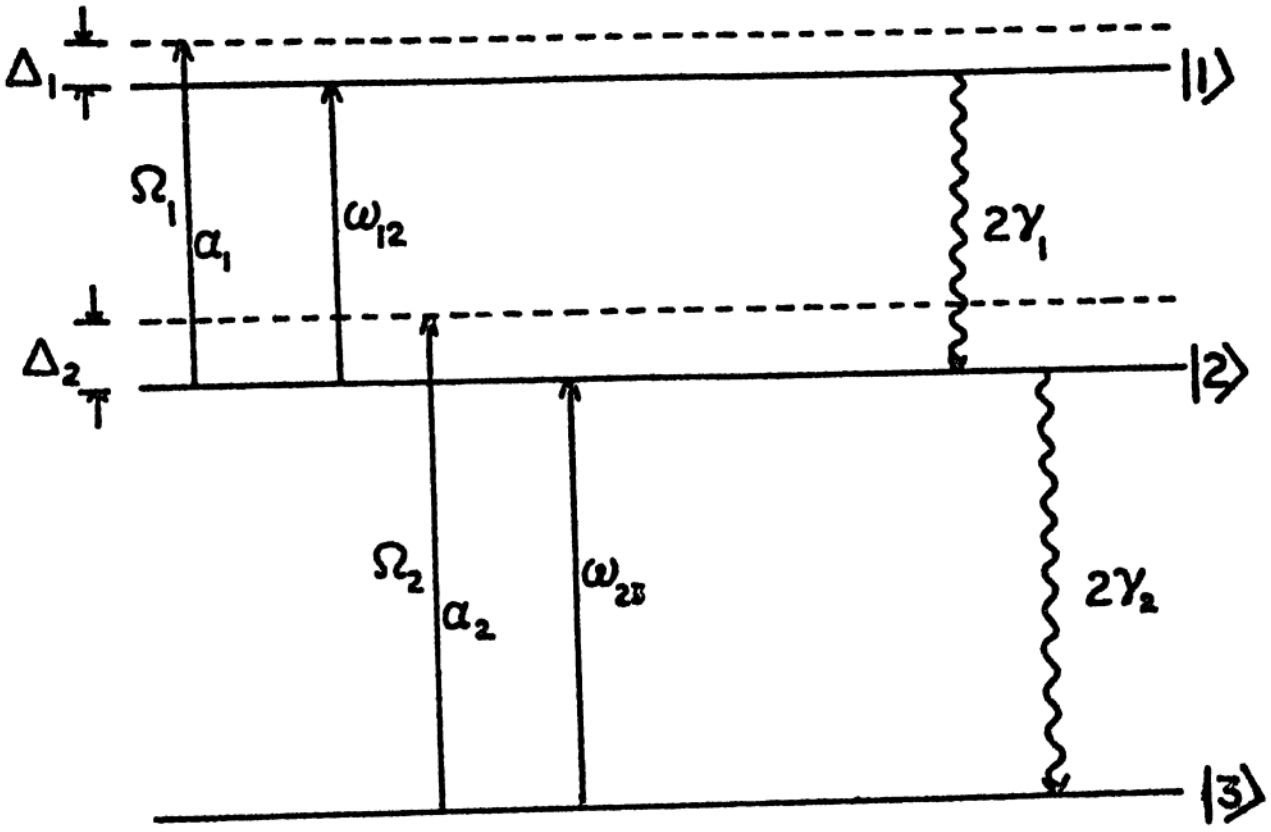

Δ_1
Ω_1
α_1
ω_{12}
$2\gamma_1$
$|1\rangle$
Δ_2
$|2\rangle$
Ω_2
α_2
ω_{23}
$2\gamma_2$
$|3\rangle$

FIG. 22

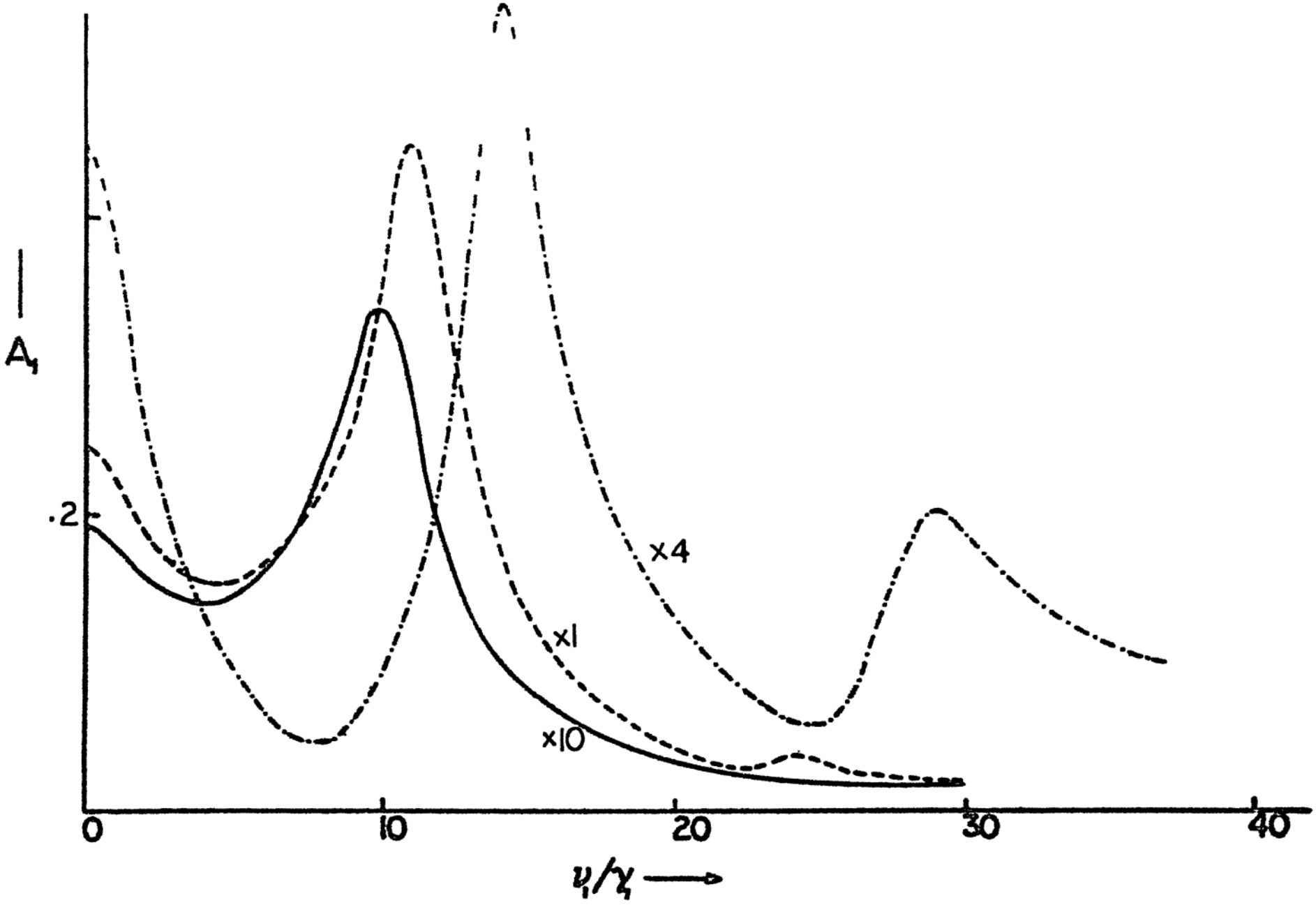
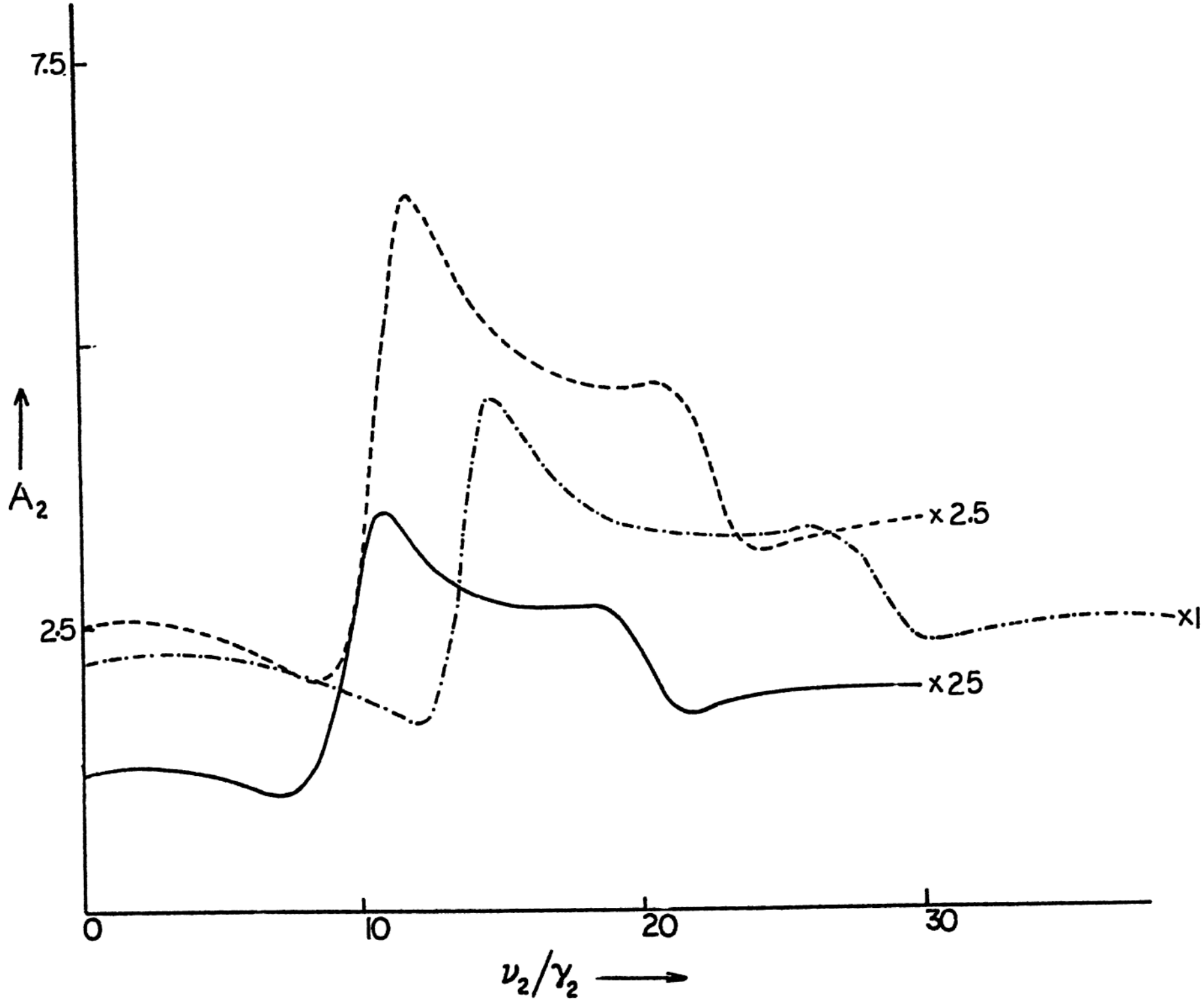


FIG. 23


7.5
2.5
A_2
x 2.5
x1
x25
0
10
20
30
ν_2/γ_2

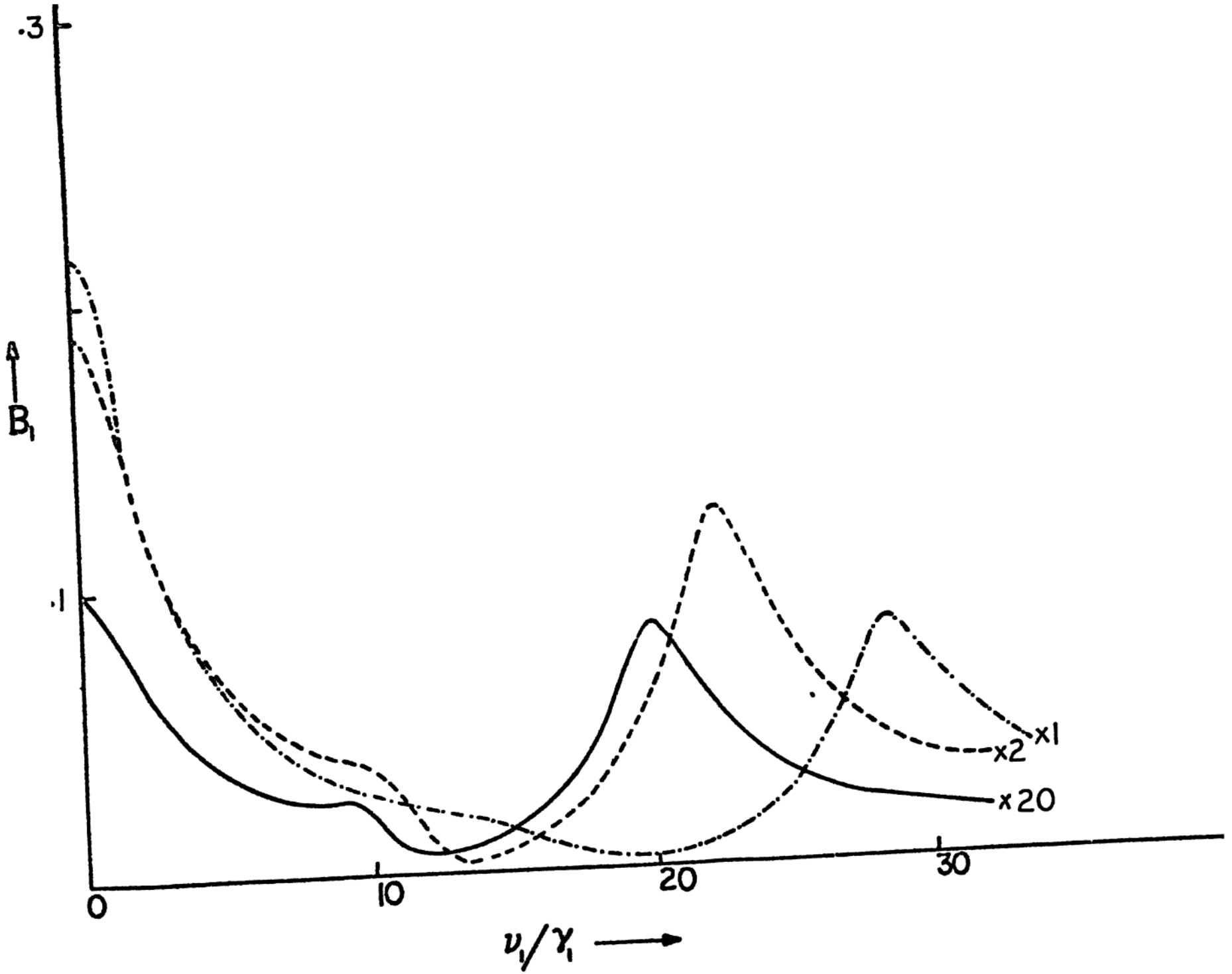
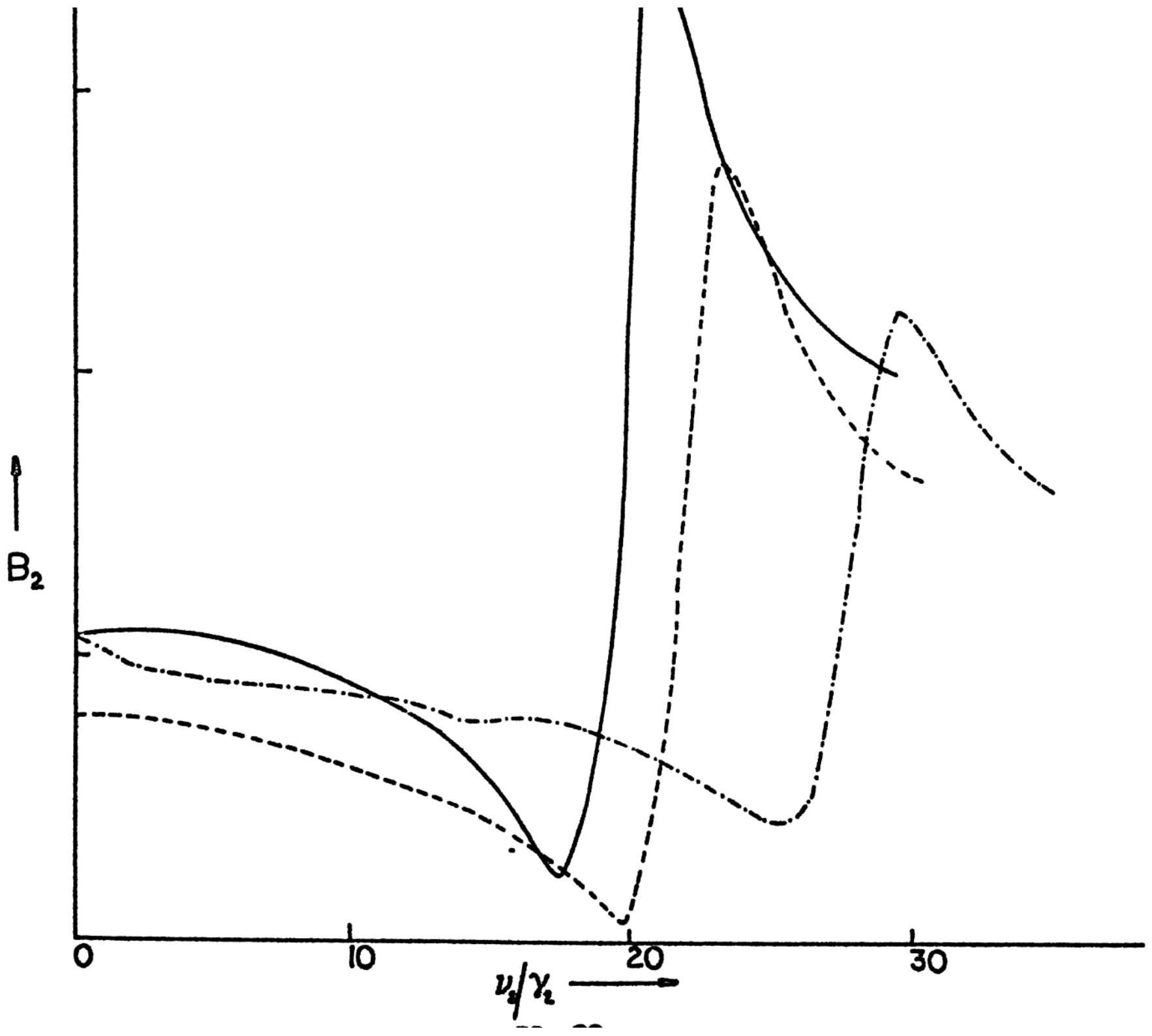
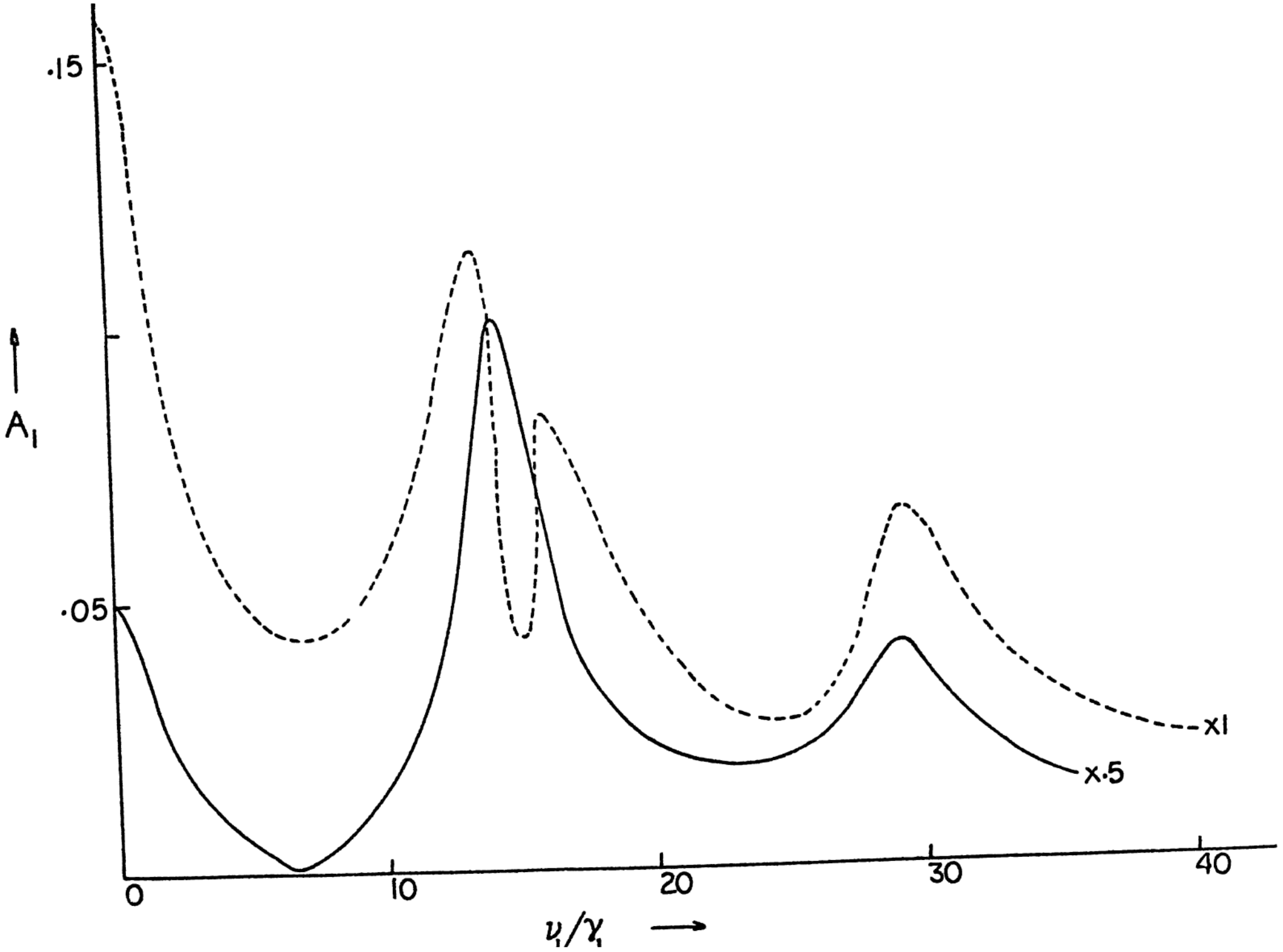


FIG. 25


B2
0
10
20
30
v2/γ2


.15
.05
A_1
0
10
20
30
40
ν_1/γ_1
x1
x.5

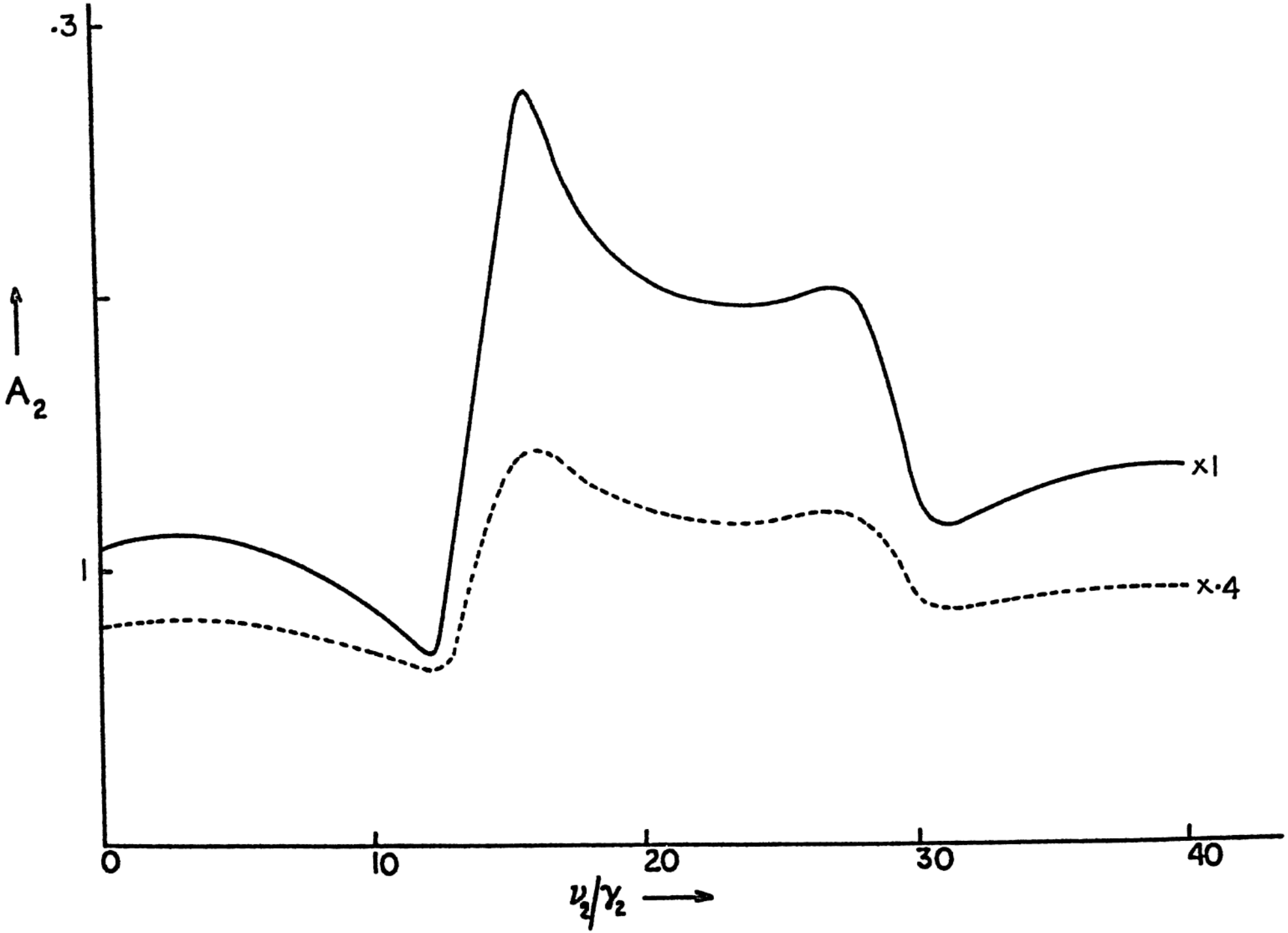


FIG. 28

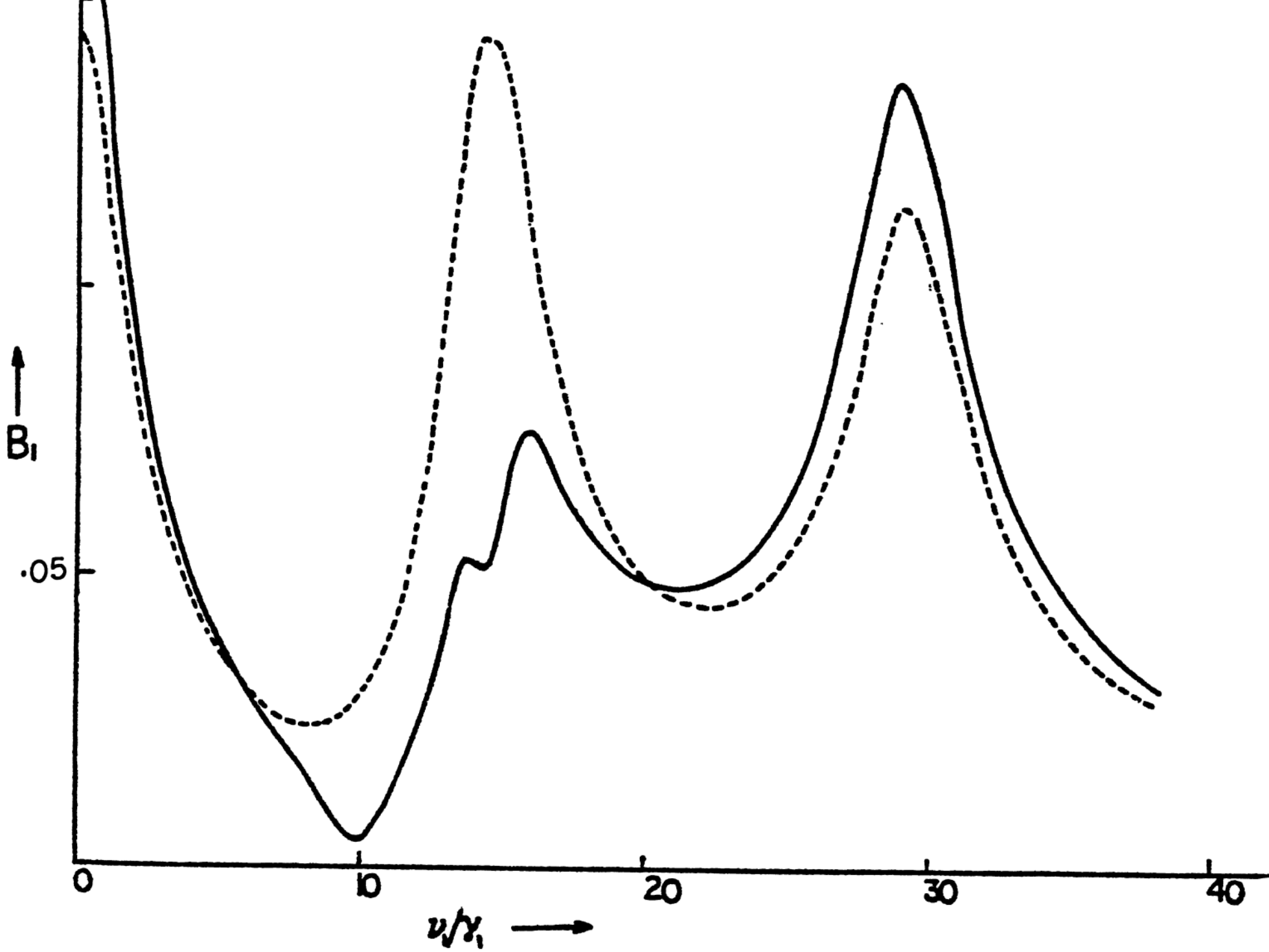


FIG. 29

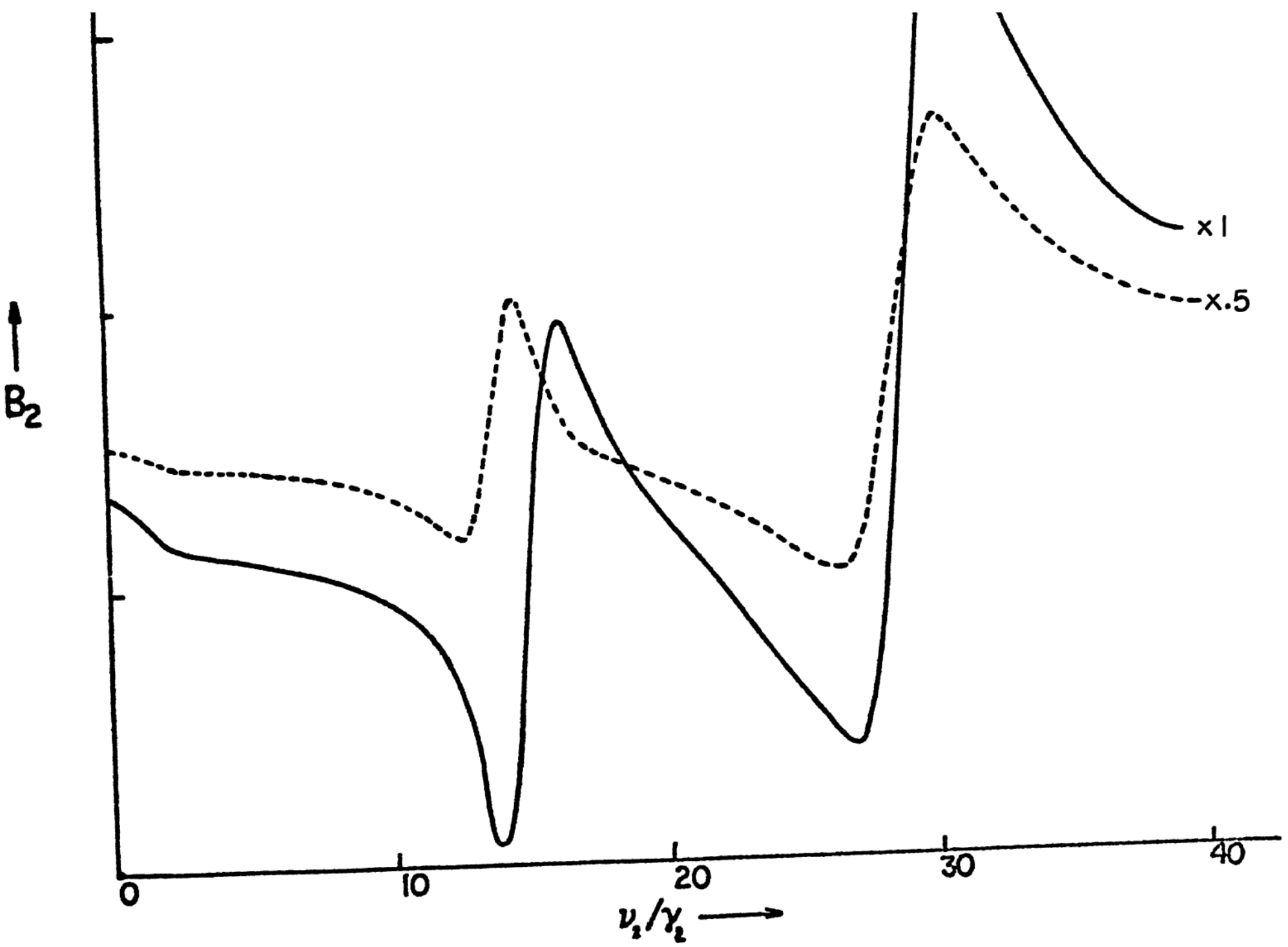


FIG. 30

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(1) Senior Cambridge (I.S.C.), University of Cambridge - Local Examination Syndicate. First class with first position in U.P.Merit List (1970).

(2) B.Sc.,(Physics Honours), Banaras Hindu University. First class with second position (1973).

(3) M.Sc., Banaras Hindu University. First class with Chancellor's medal for first position in M.A., and M.Sc., Examinations (1975).

LIST OF PUBLICATIONS

(1) " Quantum Theory of Resonance Fluorescence with Recoil " Effects" , G.S.Agarwal and R.Saxena, Opt. Commun. 26, 202 (1978).

(2) " A.C.Stark Effect and Fluorescence using Modulated " laser beams" I. Effect of laser fluctuations and arbitrary relaxation parameters" , R.Saxena and G.S.Agarwal, J. Phys. B 12, 1939 (1979).

(3) " AC Stark Effect and Fluorescence using modulated laser beams. II Two photon stepwise excitation" , R.Saxena and G.S.Agarwal, J.Phys. B13, 453 (1980).

(4) " Analytical solution for the spectrum of resonance fluorescence of a cooperative system of two atoms and the existence of additional sidebands" , G.S.Agarwal, R.Saxena, L.M.Narducci, D.H.Feng and R.Gilmore, Phys. Rev. A21, 257 (1980).

(5) " Modulated Hanle Signals in Partially coherent fields" R.Saxena and G.S.Agarwal, Phys. Rev. A (submitted).

(6) "Resolution Beyond Natural Linewidth via Fluorescence in Modulated Beams" , R.Saxena and G.S.Agarwal, submitted for publication.